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## **Textile Colouration with Natural Colourants : A Review**

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## **Abstract**

This paper provides an in-depth review of the natural colourants covering their classifications, sources, extraction techniques, application techniques in relation to different textile fibres and their advantages and challenges. The academic readers will find here an overview of the latest developments in extraction and application techniques of a variety of natural colourants. Almost all commercially produced natural fibres and the major synthetic fibres are found to be dyeable and printable with natural colourants. Although the production of natural colourants offers a lot of environmental benefits, their application techniques in textile colouration are not always sound for environment due to the need of synthetic mordants in colouration process. Advances in modern agriculture and biotechnology can play a key role in sorting the limitations of natural colourants and at the same time more and more investigations are still required to establish their cleaner application on textiles.

## **Keywords**

Natural colourants, textiles, fibres, extraction process, dyeing, printing

## 1. Introduction

Nature is superbly colourful and is an abundant source of colourants. Both living and non-living members of nature store an unending amount of colouring substances, including both dyestuff and pigments, in their constituents. The mankind, have mastered extracting and using those since the dawn of human civilisation (Zerin et al., 2019). These colourants have been used in the food, cosmetics, and pharmacology and textiles since time immemorial (Saxena and Raja, 2014; Sen et al., 2019). Soon after the arrival of synthetic dyes in 1856, the use of natural colourants in the textile industry had started to decline (Samanta and Agarwal, 2009). The practice of using natural dyes has only survived in the cottage industry sector by artisans, enthusiasts, and small entrepreneurs. Nevertheless, the vast application of synthetic dyes in textile colouration is causing alarming environmental pollution and harm to aquatic ecosystem as these are produced from petrochemical sources, which bear potentially carcinogenic and toxic compounds (Chavan, 2013; Hassaan and El Nemr, 2017). This has again geared up the interest in natural colourants among the environmentally conscious consumers.

A brief summary of recent scientific research works on the application and sustainability of natural colourants in textile substrates with important observations are revealed in this paper.

## 2. Classification of Natural colourants for textiles

Natural colourants can be classified in multiple of ways broadly depending on their hues, sources, chemical structures and their applications (Gulrajani et al., 1995; Mansour, 2018; Patel, 2011; Shahid et al., 2013; Teli et al., 2000; Yusuf et al., 2017b). They can be classified as '*substantive colourants*,' which can be directly applied to the textiles, such as indigo, turmeric and '*adjective colourants*,' which need mordant to apply to materials, such as logwood (Samanta and Konar, 2011).

### 2.1. Classification based on hue

The Colour Index, known as CI number, jointly published by the Society of Dyers and Colourists (SDC) based in the UK and the American Association of Textile Chemists and

Colorists (AATCC), classify colourants based on their application classes and chemical constitutions. Natural colourants have been grouped separately in volume 3 of the SDC Colour Index that includes 32 natural reds, 6 natural oranges, 4 natural blues, 5 natural greens, 29 natural yellows, 12 natural browns, 6 natural blacks, and one natural white in the list (Saxena and Raja, 2014) (see Table 1).

Table 1. Number of natural dyes in each hue registered in Colour

CI Natural	Number of Dyes	Percentage (%)	Examples of sources	Reference
Yellow	29	30.4	Barberry ( <i>Berberis aristata</i> ), tesu flowers ( <i>Butea monosperma</i> ), safflower yellow ( <i>Carthamus tinctorius</i> L.), kamala ( <i>Mallotus philippensis</i> ), dyer's chamomile ( <i>Anthemis tinctoria</i> ), turmeric ( <i>Curcuma longa</i> )	(Ahmed, 2009; Biertmpfel and Wurl, 2009; Teli et al., 2001, 2000)
Orange	6	6.5	Annatto ( <i>Bixa Orellana</i> )	(Adeel et al., 2019; Gupta and Gulrajani, 1994)
Red	32	34.8	Madder ( <i>Rubia tinctorum</i> ), manjistha ( <i>Rubia cordifolia</i> ), brazil wood/sappanwood ( <i>Caesalpinea sappan</i> ), morinda ( <i>Morinda citrifolia</i> ), cochineal ( <i>Dactylopius coccus</i> ) and lac <i>Coccus laccae</i> )	(Ahmed, 2009; Biertmpfel and Wurl, 2009; Vankar, 2000; Yusuf et al., 2017b)
Blue	4	3.3	Natural indigo ( <i>Indigofera</i> species), Kumbh (Manipur) and the flowers of Japanese 'Tsuykusa'	(Adeel et al., 2019; Ahmed, 2009; Yusuf et al., 2017b)
Green	5	5.5	Usually very rare, other than a combination of woad/indigo with indigo dyes	
Brown	12	13	Virtually unlimited source. Cutch is an ancient brown dye sourced from the wood of acacia trees. Lady's mantle ( <i>Alchemilla vulgaris</i> L.), Milfoil ( <i>Achillea millefolium</i> L.), Wild marjoram ( <i>Origanum vulgare</i> L.)	(Biertmpfel and Wurl, 2009; Hao et al., 2006; Yusuf et al., 2017b)
Black	6	6.5	Roots of iris plant, lac, carbon, and caramel	(Gulrajani et al., 1995; Hao et al., 2006; Yusuf et al., 2017b)

## 2.2. Classification based on sources

Four out of the five kingdoms of living things - animals (all multicellular animals), plants (all green plants), fungi (moulds, mushrooms, yeast) and prokaryotes (bacteria, blue-green algae) – are known to be the sources of extractable colourants. Among the non-living things, soil and rocks provide readily usable colourants. Based on the origin, natural dyes can be obtained primarily from three sources: plants, minerals, and animals (Chakraborty, 2014; Patel, 2011), as shown in Figure 1. These are discussed below.

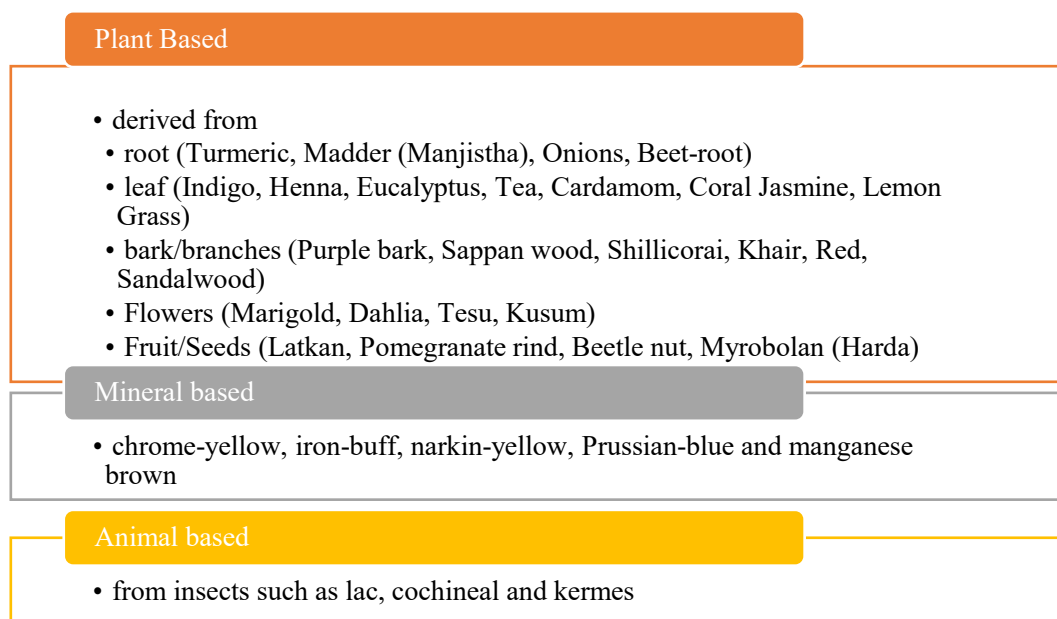


Figure 1. Broad classification of natural dyes based on sources

### 2.2.1. Dyes derived from Plants

Historically the majority of the natural dyes are obtained from the plants as various parts of several plants such as roots, leaves, twigs, seeds, bark, fruits, rinds, husks, stems, hulls, trunks etc. are found to be potentially rich in colourants (Chakraborty, 2014; Saxena and Raja, 2014; Yusuf et al., 2017b). Some plants can produce different colours in different body parts (see Table 2) depending on their age, surrounding environment and the growth conditions and the season of the year (Hancock and Boxworth, 1997). In India, around four hundred fifty dye-yielding plants have been identified (Gulrajani et al., 1995; Yusuf et al., 2017b).

Table 2. Natural dyes obtained from different parts of different plants

Botanical name	Common name of the plant	Part used	Hue	Reference
<i>Eupatorium</i>	Siam weeds	Whole plant	Yellow	(Vijaykumar and Kumar,

<i>odoratum</i>				1999)
<i>Ageratum conyzoides</i>	Goat weed	Whole plant	Yellow	(Vijaykumar and Kumar, 1999)
<i>Atrocarpus heterophyllus</i>	Jackfruit tree	Bark	Yellow	(AK and P, 2007)
<i>Delonix regia</i>	Gulmohar	Flower	Olive green	(Saxena and Raja, 2014)
<i>Tectona grandis</i>	Teak	Leaves	Yellow	(Saxena and Raja, 2014)
<i>Acacia nilotica</i>	Babool	Leaves, bark	Yellow/brown	(Chattopadhyay et al., 2013)
<i>Nymphaea alba</i>	Water lilly	Rhizomes	Blue	(Saxena and Raja, 2014)
<i>Dahlia variabilis</i>	Dahlia	Flowers	Orange	(Saxena and Raja, 2014)
<i>Emblica officinalis</i>	Amla	Bark, fruit	Grey	(Saxena and Raja, 2014)
<i>Ziziphus mauritiana</i>	Indian Jujube Ber	Leaf	Pink	(Saxena and Raja, 2014)
<i>Moringa pterygosperma</i>	Drumstick	Leaf	Yellow	(Saxena and Raja, 2014)
<i>Tamarix aphylla</i> L. Karst	Tarfa/Athl	Leaf	Brown	(Baaka et al., 2017)
<i>Brassica oleracea</i> L. var. <i>capitata</i> f. <i>rubra</i>	Red Cabbage	Leaf	Red	(Haddar et al., 2018)
<i>Tamarindus indica</i>	Tamarind	Leaves, seeds	Yellow, brown	(Saxena and Raja, 2014)
<i>Rumex maritimus</i>	Golden dock	Seeds	Brown	(Savvidis et al., 2017)
<i>Eucalyptus camaldulensis</i>	Eucalyptus	Bark	Yellow and brown	(Biertmpfel and Wurl, 2009; Mongkholrattanasit et al., 2009; Naz and Bhatti, 2011)
<i>Pterocarpus santalinus</i>	Red sandal wood	Wood	Red	(Gulrajani et al., 2003; Savvidis et al., 2017)
<i>Acacia catechu</i> wild	-	Bark	Reddish Brown	(Patil et al., 2012)
<i>Bougainvillea glabra</i>	-	Flower	Yellow/Brown/Green / Orange	
<i>Hibiscus</i>	-	Flower	Red/pink/brown/violet	
<i>Jatropha curcas</i>	-	Bark/leaves/root	Blue/Yellow	
<i>Indigofera tinctoria</i>	-	Leaves, pods	Blue	
<i>Melastoma Malabathricum</i>	-	Fruits, Flowers	Purple/Black	
<i>Rubia cordifolia</i>	-	Whole plant	Red-brown	
<i>Terminalia chebula</i>	-	Root, seeds, fruit, Bark	Dark Blue/Yellow, Gray	
<i>Toona hexandra</i>	-	Flowers/seeds	Yellow-brown	

### 2.2.2. Dyes derived from the Mineral origin

Few earth pigments composed of inorganic metal salts and metal oxides are known to be applicable as textile colourants. Some vital mineral colourants are cinnabar (a toxic mercury sulfide mineral), chrome yellow, iron buff, mineral khaki and manganese brown, malachite, ultramarine blue, azurite, gypsum, talc, charcoal black (Chakraborty, 2014; Saxena and Raja, 2014). Mineral colours are water insoluble inorganic compounds, which are precipitated onto the fibre by a process known as double decomposition (Affat, 2021; Cavalcanti et al., 2021; Choudhury, 2018; Patel, 2011; Samanta and Konar, 2011).

### 2.2.3. Dyes derived from Animals

Few insects and molluscs contain extractable colourants in their bodies. Colourants carminic acid, kermesic acid and accaic acid originate from the dried bodies of insects - cochineal, kermes and lac respectively (Yusuf et al., 2017a). Cochineal (*Dactylopius coccus*) is a scale insect, and the bodies of female cochineals and their eggs contain carminic acid (C.I. Natural Red 4), which has found applications in food colouration, drug and textile dyeing. This colourant produces a variety of shades of pink, crimson red, scarlet red, and blue onto natural fibres when used with different mordants (Adeel et al., 2018). Kermes (*Kermes vermilio*) is another scale insect, and like cochineal, the bodies of female kermes and their eggs contain colouring substances. This insect is the source of a yellowish-red dye kermesic acid (C.I. Natural Red 3). Kermes powders were used for textile dyeing in ancient England, France, Spain, Turkey, and Scotland. Another red dye C.I. Natural Red 25, is obtained from the resinous secretion of different species of lac insects – *Laccifer lacca* and *kerria lacca*.

The historic dyes Tyrian purple and Tekhtelet blue are obtained from sea snails belonging to *Bolinus brandaris* (also known as *Murex brandaris*), *Hexaplex trunculus* (i.e *Murex trunculus*) and *Stramonita haemastoma*.

### 2.2.4. Colourants from Fungi



Different non-mycotoxigenic species of soil and wood-inhabiting fungi produce dyes and pigments, which have potential applications in the food, pharmaceutical, cosmetics and textile industries. In some cases, such colourants exhibit very high fastness to light and washing when applied in textiles. Soil inhabiting filamentous fungus *Monascus purpureus*, *Isaria farinosa*, *Emericella nidulans*, *Fusarium verticillioides*, and *Penicillium purpurogenum* produce red, pink, red and yellow shades on cotton substrates when dyes in the presence of alum and ferrous sulfate (Velmurugan et al., 2010, 2009). Wood inhabiting fungi *Penicillium murcianum* and *Talaromyces australis* can yield yellow and red respectively on wool (Hernández et al., 2019). Other wood-staining fungi *Chlorociboria aeruginosa*, *Scytalidiumcuboideum*, and *S. ganodermophthorum* produce green, red, and yellow pigments suitable for dyeing cotton, polyamide 6.6, polyester, polyacrylic, and wool materials (Weber et al., 2014).

#### 2.2.5. Colourants from Microbes

Interestingly different species of bacteria can produce pigments. For example, the *Rhodobacter sphaeroides*, *Flavobacterium multivorum*, *Agrobacterium aurantiacum*, *Gordonia jacobea*, *Paracoccus carotinifaciens*, and *Sphingomonas* sp. make carotenoid, a class of organic pigments with yellow-orange-red hues, in their bodies (Mumtaz et al., 2019).

### **2.3. Classification based on chemistry**

Chemistry-based classification of natural colourants identifies the particular chemical groups with specific characteristic properties exist in their chemical structures (Yusuf et al., 2017a). Based on major chemical constituents present, they are grouped as indigoids, quinonoids, flavonoids, carotenoids, tannin based dyes (Choudhury, 2017; Kumar and Sinha, 2004; Saxena and Raja, 2014). Figure 2 shows a schematic of the above-mentioned structures of natural dyes with examples.

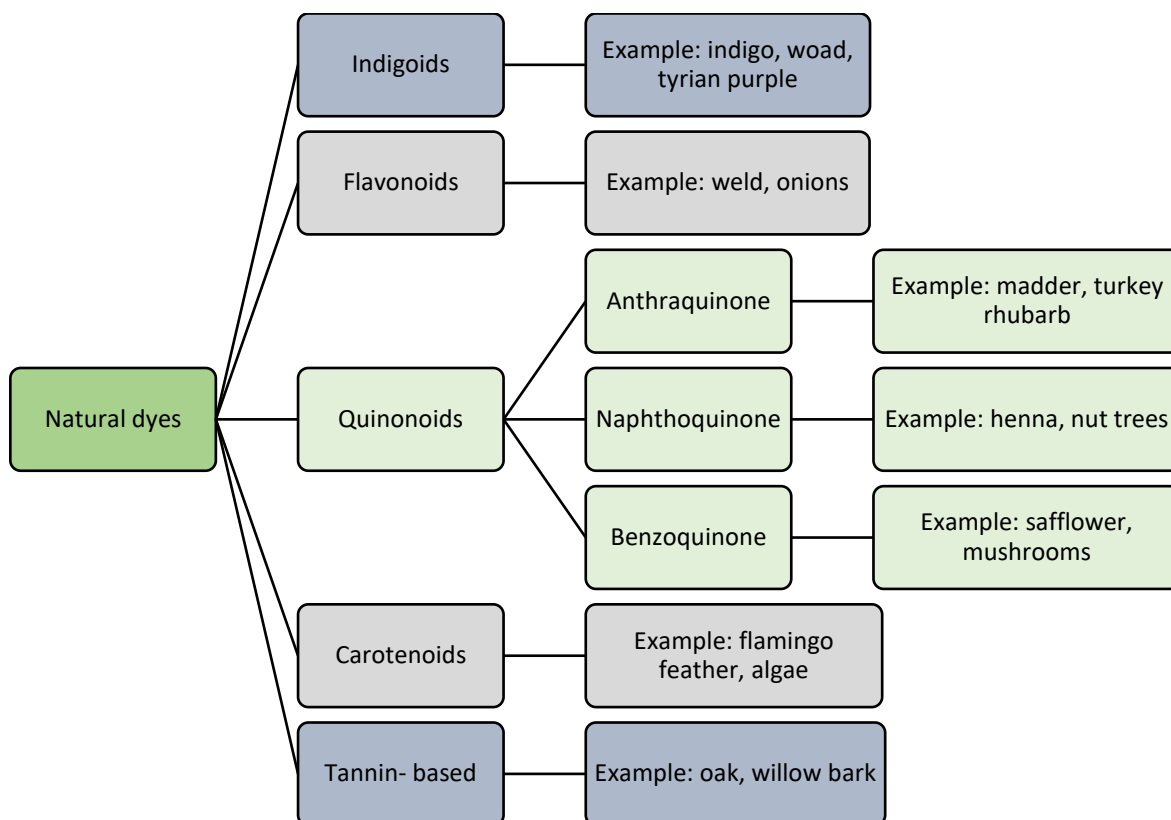


Figure 2. Natural dyes of different chemical structure and respective plant sources

### 2.3.1. Indigoid dyes

Examples of natural dyes containing indigoids are given in Table 3. Indigo is the most common dye of this group, and its main chemical ingredient is indigotin (CI Natural Blue 1, C.I. number 75780) found in the leaves of Indigo plants. The structure is very similar to synthetic indigo (C. I. Vat Blue1, C.I. 73000). Still, natural indigo contains some red dye indirubin, which can generate a prominent reddish tone to the textile materials (Saxena and Raja, 2014). The most widely used source are Indigofera species, Isatis species, and Polygonum species. The Indigofera is known to give about 60% of yield of indigo dyes (Clark et al., 1993). The other most crucial dye in this class is the Tyrian purple that can be derived from the Mediterranean mollusks of the genera - Purpura and Murex. As it is shown in the Figure 3, it is basically the 6, 6' dibromo derivative of indigo (Figure 3) including indirubin, monobromoindirubin, 6,6'-dibromoindirubin and indigotin (Mantzouris and Karapanagiotis, 2014; Vasileiadou et al., 2019, 2016). This natural source is no longer used as the shellfish

became extinct as a large number was needed to produce even a tiny amount of dyes (Daniels, 2006; Patel, 2011).

Table 3. Naturally available indigoid dyes

Common name	Commonly available species	Active ingredient	Reference
Indigo	Indigofera species such as <i>I. tinctoria</i> , <i>I. erecta</i> , and <i>I. sumatrana</i> ; pala indigo ( <i>Wrightia tinctoria</i> ), Assam indigo ( <i>Strobilanthes flaccidifolius</i> ), Japanese knotweed ( <i>Polygonum tinctorium</i> ) and common knotweed ( <i>P. aviculare</i> ), <i>Nerium tinctorium</i> , <i>Marsdenia</i> and <i>Lonchocarpus cyanescens</i>	Mainly Indigotin with indirubin  glucoside indican or isatan B including some indirubin and kaempferol	(Patel, 2011; Saxena and Raja, 2014)
Woad	<i>Isatis tinctoria</i> and other species	Indigotin	
Tyrian purple	molluscs <i>Murex trunculus</i> , <i>M. brandaris</i> and <i>Purpura lapillus</i>	6,6'-dibromoindigo also indirubin and monobromoindirubin	

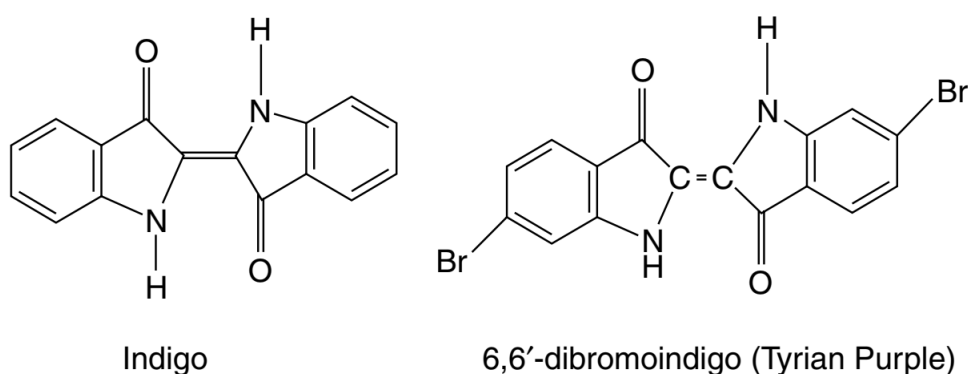


Figure 3. Chemical Structure of indigo and tyrian purple (Patel, 2011)

### 2.3.2. Quinonoid dyes

Quinonoids are the most diverse naturally occurring colouring substances. The colour gamut is widely distributed, ranging from yellow to red. Further classification of these dyes based on chemical structure includes the classes of anthraquinones,  $\alpha$ -naphthoquinones, and benzoquinones (Bechtold, 2009).

(a) Anthraquinone dyes:

A majority of the red dyes belong to anthraquinone structure (see Figure 4). They come from both plants and insects (Table 4). The most famous natural dye is the alizarin (1, 2-dihydroxy anthraquinone) obtained from the European madder (*Rubia tinctorum*). There is also Indian madder, known as manjishta/manjeet (*Rubia cordifolia*), known for its health benefits in Ayurveda. The composition of extracted anthraquinone differs from plant to plant. The main composition of Indian madder is purpurin (1,2,4-trihydroxyanthraquinone) (Gupta et al., 2001). Madder roots are known to be a source of more than 35 anthraquinoids (Schweppe, 1993). Rhubarb, another anthraquinone dye, has been known for its pharmaceutical values for over 400 years. These plants grow in Himalayas in India, China and Tibet, Siberia, and Europe (Khan et al., 2012). The dye (CI Natural Yellow 23) is extracted from the roots and contains a complex mixture of anthraquinones (Bechtold, 2009; Schweppe, 1993). Other dyes include lac, morinda, and cochineal. These dyes are known to have good colour fastness to wash and light (Chakraborty, 2014; Saxena and Raja, 2014).

Table 4. Some anthraquinone dyes with CI number and source

Dye name	CI Number	CI name	Natural sources	Reference
Alizarin	75330	Natural red 6, 8, 9, 10, 11, 12	Madder, chayroot	(Hancock and Boxworth, 1997; Vasileiadou et al., 2019)
Purpuroxanthin or xanthopurpurin	75340	Natural red 8, 16	Madder, munjeet	
rubuadin	75350	Natural red 8, 16	madder, <i>Gallium</i>	
morindanigrin	75360	Natural Red 19	<i>Morinda umbellata</i>	
munjistin or purpurinxanthin-carboxylic acid	75370	Natural Red 16,8	madder, munjeet	
Morindadiol	75380	Natural red 18	Morinda root	
Soranjidiol	75390	Natural red 18	Morinda root	
chrysophanic acid	75400	Natural Red 23	turkey rhubarb	
Purpurin	75410	Natural red 16, 8	Munjeet, madder	
Pseudopurpurin	75420	Natural red 14, 9, 8	Gallium, madder	
morindon	75430	Natural Red 19, Yellow 13	oungkouda	
emodin or frangulaemodin	75440	Natural Green 2, Yellow 14	Persian berries	
Laccaic acid	75450	Natural red 25	<i>Coccus laccae</i> (Lac dye)	
Kermesic acid	75460	Natural red 3	<i>K. licis</i>	
Carminic acid or cochineal	75470	Natural red 4	<i>C. cacti</i>	

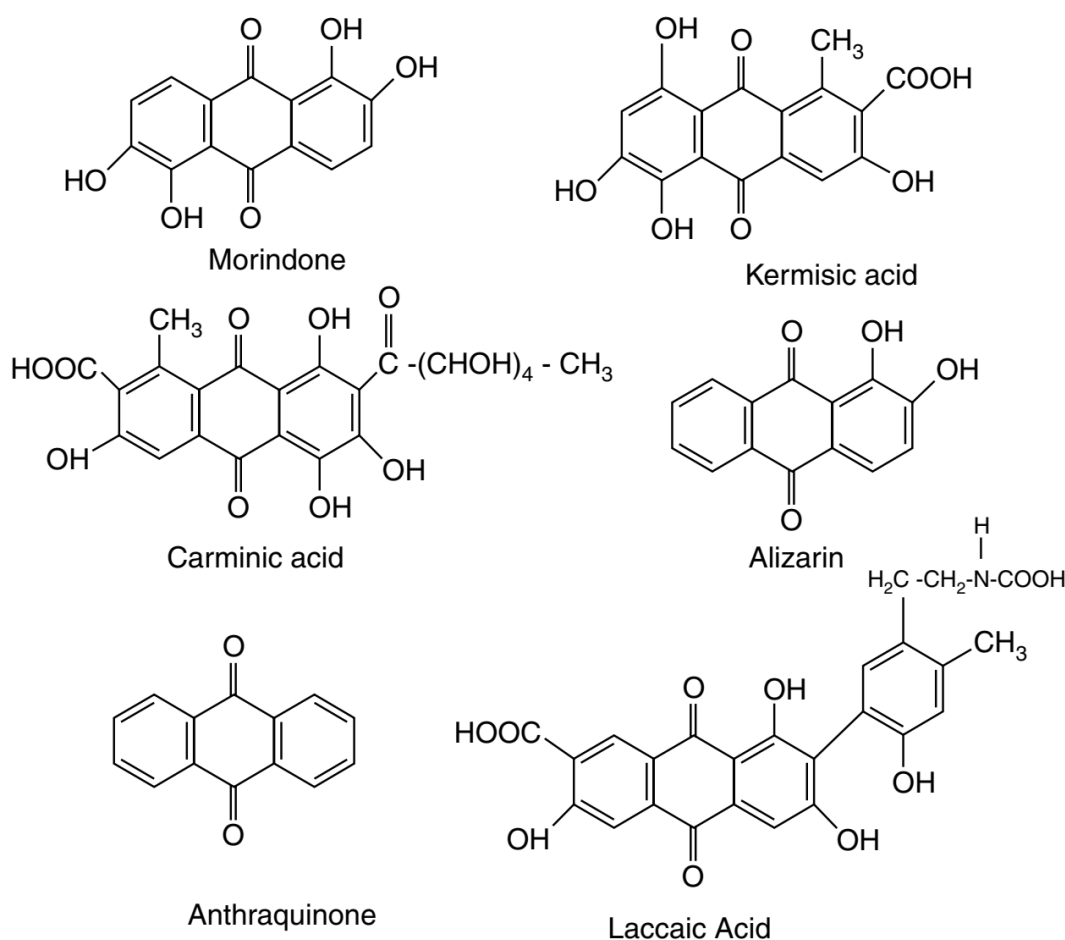


Figure 4. Structures of some anthraquinone dyes (Patel, 2011)

#### (b) Naphthoquinone dyes:

Like anthraquinone dyes, naphthoquinones also give away different shades of orange, red, or reddish-brown hues (Saxena and Raja, 2014). The two most prominent members of this class are lawson (2-hydroxy-1,4-naphthoquinone) and juglone (5-hydroxy-1,4, naphthoquinone) (see Figure 5). The commonly known henna (*Lawsonia inermis* L.), cultivated and used in Bangladesh, India, Northern and Eastern Africa, and Australia is the prime source of lawson (Cartwright-Jones, 2006; Chakraborty, 2014) for the traditional use of colouring hair and nails, also in dyeing wool and silk in orange to brown colours (Obara and Onodera, 1979) directly or in combination with different mordants (Agarwal et al., 1992). Henna can also be used to dye polyester with better affinity than polyamide (Gupta and Gulrajani, 1994). Another naphthoquinone, Juglon, is available in plenty in different parts of nut trees

(*Juglandaceae*) depending on the variety of trees. The main component of juglone is a glycoside, e.g. 5-hydroxynaphthohydroquinone-4-b-D-glycoside (Schweppe, 1993).

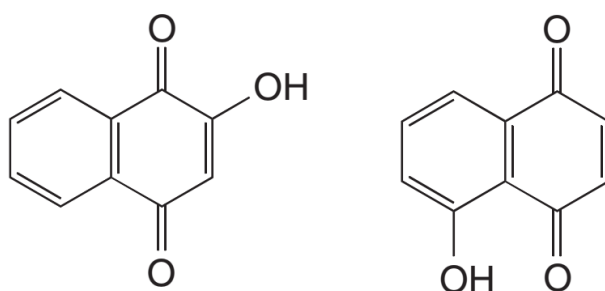


Figure 5. Chemical structure of henna (2-hydroxy-1,4-naphthoquinone) and juglone (5-hydroxy-1,4-naphthoquinone)

### (c) Benzoquinone dyes

This dye range is relatively small. The most important dye is the red dye found in safflower (*Carthamus tinctorius* L., CI Natural Red 26), known as Carthamin (see Figure 6). The dye structure is quite complex and sensitive to hydrolysis (Obara and Onodera, 1979). The oxidation of yellow cartharmin produces the red-orange dye to produce carthamon. Safflower is cultivated in subtropical regions of Egypt, Iran, India, China, Africa, North and South America from ancient times for food, cosmetics, pharmaceuticals, and colouration of wool, silk, and cotton (Oda, 2001). Mushrooms and lichens are also known to be source of Benzoquinone dyes (Bechtold, 2009).

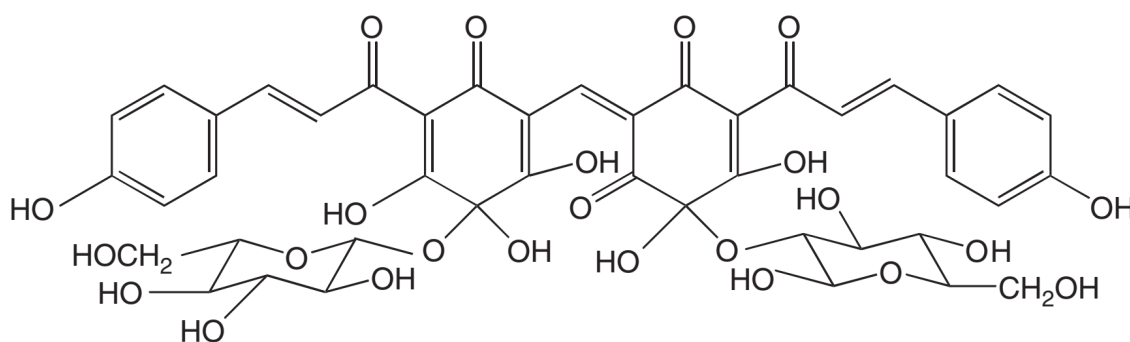


Figure 6. Structure of carthamin (Bechtold, 2009)

### 2.3.3. Flavonoid dyes

Flavonoid dyes consist of the largest colour palette ranging from pale to deep yellow, orange to reds and blues through plant sources (Markham, 1982). These dyes are derivatives of phenylalanine and the acetate coenzyme A esters, which possess a C6-C3-C6 structure (Guinot et al., 2006). The plant uses flavonoids as a defence mechanism and signalling

compounds for reproduction, symbiosis, and pathogenesis. These can substantially absorb UV-B light and thus can prevent damage of leaf tissues by UV radiation. These compounds are produced in large quantities in the plant and substantially affect soil composition during decomposition. The role of flavonoids as a food content has been extensively studied due to their antioxidant properties, anti-ulcer and anti-viral infections, and prevention of chronic age-related diseases, including cardiovascular disease and cancer (Cook and Samman, 1996; D. Li et al., 2019; Perez-Vizcaino and Fraga, 2018). An average human diet, based on an estimation, includes a flavonoid intake ranging between 100 and 1000 mg/day (Aherne and O'Brien, 2002). Approximately 6000 naturally occurring flavonoids, most of which are pale, yellow or colourless, are known to have been isolated from different plants (Wätjen et al., 2005). Figure 7 presents the chemical structures of common flavonoids.

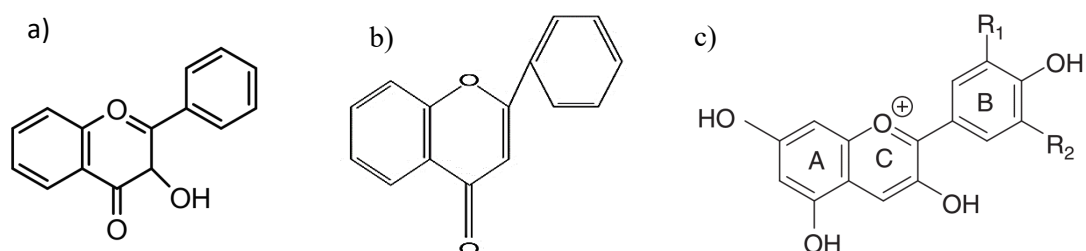


Figure 7. Structure of a) flavonol (pale yellow) b) flavone and c) anthocyanidin (Orange to violet) (Giusti and Wallace, 2009; Gulrajani and Gupta, 1992; Patel, 2011)

#### 2.3.4. Carotenoid dyes

These are red, orange or yellow pigments that are commonly seen in the most photosynthesising organisms. Around 750 different carotenoids (not including cis-trans isomers) have so far been separated from different natural sources (Britton et al., 2004). Most carotenoids are xanthophylls and carotenes and are responsible for attractive colours in many red and yellow fruits and vegetables, the feather of birds such as flamingos and wild cock. Carotenoids also occur in algae, fungi, yeasts, moulds, mushrooms, and bacteria. All the sources of orange pigment have shared standard structural features such as polyisoprenoid structure and a series of centrally located bonds (Chakraborty, 2014; Chandrika, 2009). The basic carotenoid structure is linear and symmetrical, containing around 40 carbon atoms with one or two cyclic structures at the end of their conjugated chains. The long alternating single and double bonds systems give distinct molecular shapes, light-absorbing properties, and

chemical reactivity (Chandrika, 2009). Two unique type of carotenoids that are commonly used for textile colourtaion are Saffron (*Crocus stativus*) and annatto seeds (*Bixa Orellana*). Crocetin, the yellow pigment is the main colourant in saffron flowers, while bixin and norbixin are the main colourant for annatto seeds (Chandrika, 2009; Saxena and Raja, 2014). Other dye in this class also includes Indian mahogany (*Cedrela toona*) and harshingar (*Nyctanthes arbor-tistis*) flowers.

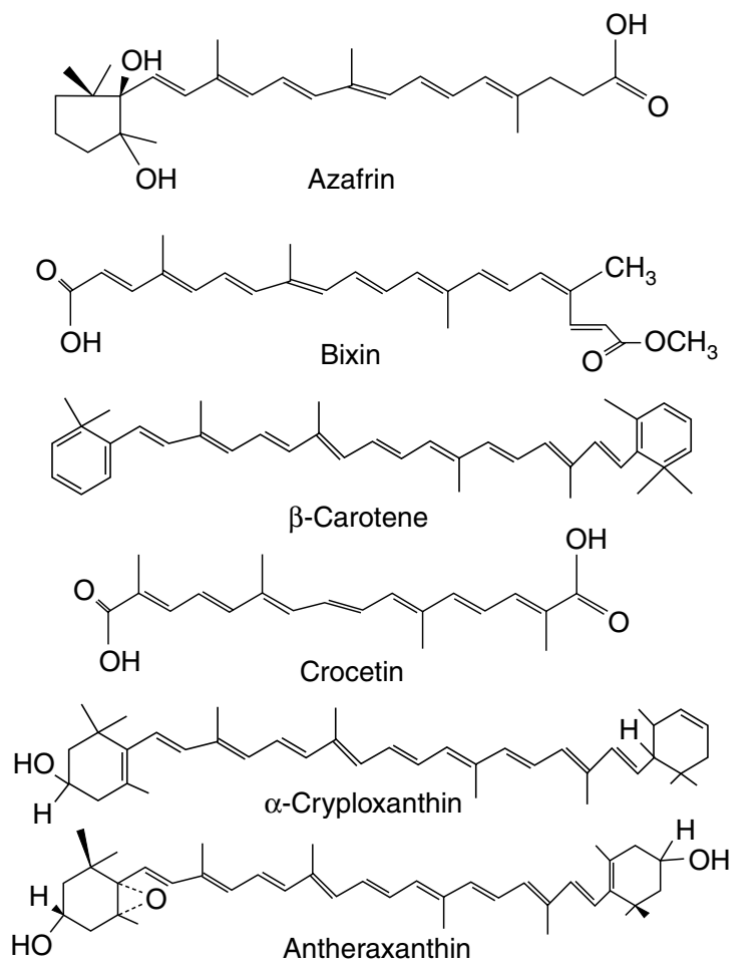


Figure 8. Structures of some carotenoid dyes (Patel, 2011)

### 2.3.5. Tannin-based dyes

Tannins are chemically polyphenolic compounds having molecular weights between 500 and 3000 and are water-soluble. Tannins are obtained from the various parts of the plants, such as fruit, pods, plant galls, leaves, bark, wood, and roots (Saxena and Raja, 2014; Yusuf et al., 2017b). The colour ranges are from light yellow to light brown, which contributes to food



and beverages. Tannins are capable of precipitating proteins (Hagerman et al., 1997). They are usually classified into three groups-hydrolysable (gallotannins and ellagitannins, derived from fustic, oak, waterlilies etc.), condensed tannins (proanthocyanidins and derived from oak, willow bark, cutch and waterlilies etc.) and the phlorotannins (Giusti and Wallace, 2009). Tannins improve the affinity towards natural dyes by working as a natural mordant for textile dyeing (Mussak and Bechtold, 2009), and essential ingredients to produce yellow, brown grey, and black colours (Gulrajani, 2001).

#### ***2.4. Classification based on application methods***

Based on the method of applications in textile colouration, natural colourants can be classified into two distinct groups: substantive and non-substantive. Substantive colourants do not require any pretreatment of the fabric before dyeing, such as indigo, orchil, and turmeric. On the other hand nonsubstantive colourants require a chemical, called mordant, to increase between colourants and fibre.

Most natural dyes are nonsubstantive. The dyes that are capable of formation of complex with metal mordants also called mordant dyes (Yusuf et al., 2017b). Unlike animal fibres, mordant plays a significant role for the dyeing of vegetable fibres such as cotton, which does not have carboxylic acid or amino group to provide attachment sites for reaction (Saxena and Raja, 2014). With the same source of dyes, different mordants produce different shades or colours with different hues or tones (Abdel-Kareem, 2012; Yusuf et al., 2017b). Mordants can come from natural sources such as pomegranate peel (*Punica granatum* L), gallnut (*Quercus infectoria* L) and catechu (*Acacia catechu*) or can be synthetic (Shahid-ul-Islam et al., 2019). Possible use of biomordants cited in different literature is given in (İşmal, 2017). Monogenetic dyes produces only one colour with mordants, whereas polygenetic dyes produce different colour based on the mordant used in the dyeing process (e.g., logwood, alizarin, fustic and cochineal) (Choudhury, 2017). Mordant dyes are mostly limited to wool, which provides deep shades with excellent wash and light fastness when applied with metallic mordants. Mordant dyes contain ligands (OH, H<sub>2</sub>, COOH), which helps to form a stable, coordination complex inside the wool fibre (Baaka et al., 2017).

Mordants are classified to three groups, which are metallic, oil mordants, and tannins (Saxena and Raja, 2014). Metal salts of aluminum, tin, chromium, copper, and iron are commonly

used in traditional textile dyeing. Chromium are black listed due to environmental concern, and copper is also in restricted category with certain limits, while the presence of tin in wastewater is a challenge. Alum (in the form of basic aluminium sulphate) or potash alum (double sulphate of potassium and aluminium) is the most widely used aluminium mordant, which as high as 10–20 % of alum can be used on the weight of the material can be used in textile dyeing (Saxena and Raja, 2014). Iron sulphate and copper sulphate are used as mordant in textile materials. Oil mordants are mainly used in dyeing processes using madder to produce Turkey red colour, which forms a complex with alum (Patel, 2011). On the other hand, tannic acid or tannins are first choice of mordants for cotton and cellulosic fibres and are important to produce yellow, brown, grey and black colours with natural dyes (Patel, 2011).

Unlike mordant dyes, extracted dyes from natural sources can be of direct, acid, cationic, vat dye (Patel, 2011; Saxena and Raja, 2014) and disperse dye in nature (Yusuf, Shabbir and Mohammad, 2017, Chattopadhyay, 2011). These dyes can be applied in exhaust and continuous/semi-continuous method. In exhaust dyeing technique, the dyes are transferred from dyebath into the fibre polymer by adsorption and absorption and finally get fixed into polymer network. On the other hand, padding (mostly pad-dry-cure in natural dyeing) has some distinct advantages over exhaust method (Maulik et al., 2011; Syamili et al., 2012).

### **3. Advancement in Dye Extraction Processes**

The amount of colourants present in natural sources are very limited. They need specific extraction technique to separate dye-bearing components from their original sources (Gupta, 2019). Elimination of non-colourant components like carbohydrate, protein, and chlorophyll is vital in extracting colouring substances from natural sources to get pure colourants. There are various methods of extracting dye-bearing materials from natural sources, as shown in Figure 9.

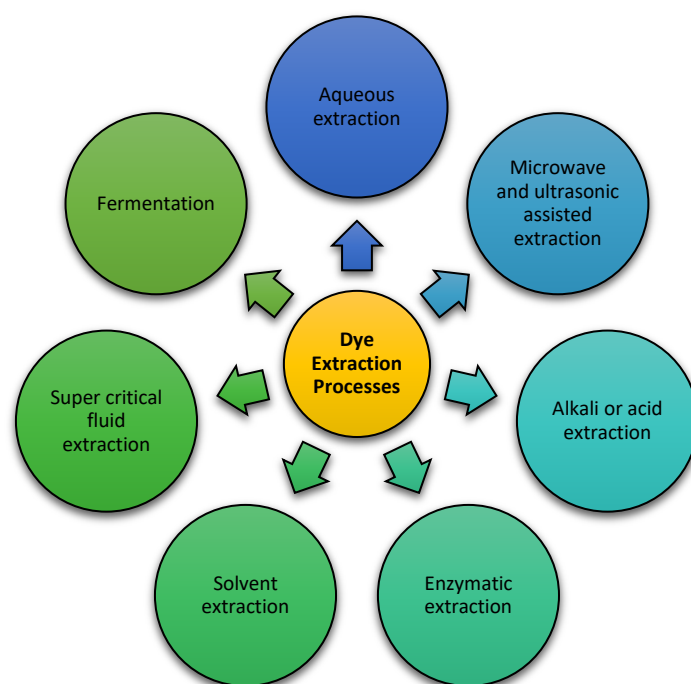


Figure 9. Various Extraction processes of natural colourants (Mansour, 2018)

Natural dyes can also be extracted using the acidic or alkaline aqueous medium. The dyes containing a phenolic group in their structures are appropriate for alkaline extraction. In addition, the extraction of lac dye and red dye from saffron can also be done by the acid/alkaline method (Gupta, 2019). The aqueous extraction process usually takes a long time; in that case, the microwave-assisted extraction process can be useful. It is reported that the liquor from *Coleus atropurpureus* leaves containing tannin and flavonoid can be extracted at an optimized condition of temperature 100°C, microwave power of 400 watts, and an M: L ratio of 0.001g/mL (Selfina Gala, 2018). Ultrasound, a good source of energy, can make the extraction process even faster. A comparative analysis on using the dye extracts by traditional and ultrasound method is observed in a work, where 0.5 %, 1 %, 2 % stock extracts were obtained from 0.5 g, 1 g, 2 g of dye fine particles in 100 ml water and extracted in the ultrasound sonicator at 35 KHz frequency for varying time. Here harda and tamarind seed coats were used as a natural mordant, and turmeric, henna was used to dye wool fabric. The ultrasound method gives effective extraction and higher colour value for both natural colourants and mordants than the simple aqueous method (Javed Sheikh, 2016). Another well-known and cost-effective process is fermentation where the bio enzymes help to extract dye bearing compounds. Various enzymes like cellulase, pectinase can be used for this purpose. Indigo can be extracted by the fermentation process. (Gupta, 2019) . The dyes extraction process from Nila leaves is optimized by the maceration method in work. 12-hour

of maceration at pH 11 is found to be the most optimal process to get a sharp blue shade from Nila leaves. The maceration method generates sharper dye than the boiling method in cotton material. (Lisna Hidayati, 2018).

#### **4. Development in Colouration Techniques with natural colourants**

As natural dyes are mostly non-substantive, it is challenging to reproduce shades and to standardise a recipe and it requires a skilled artistry. Almost all of the natural dyes require the use of mordants to get fixed into textile substrates (Samanta et al., 2016). The factors that influence textile colouration with natural dyes are pH, ionic strength of dyebath, molecular size of dyes, and dyebath temperature (Vankar, 2017; Ben et al., 2021). The following subsections highlights the recent advancements in colouration of different fibre types with natural colourants.

##### ***4.1. Dyeing and printing of Cotton***

Cotton is widely investigated as the most frequently used natural fibre to explore its suitability in dyeing with a range of natural dyes (El-Sayed et al., 2022; Kavyashree, 2020; Yildirim et al., 2020). Several studies are unfolding the kinetic modelling of different natural dyes on cotton. In a recent study, kinetic experiments of curcumin (dye sourced from turmeric rhizome) on cotton showed a strong affinity between dye and cotton. The dyeing process followed the pseudo-second-order model, and the whole process was reported exothermic (A. N. M. Haque et al., 2018). Moreover, the value of enthalpy was matched in chemisorption mechanism, which suggests the possible chemical interaction of curcumin with cotton. Similar results were found with lac dye (Chairat et al., 2008). Furthermore, similar findings were reported when lac dye was used on chitosan pre-treated cotton, and curcumin was used on regenerated cellulose such as modal (Ahsanul Haque et al., 2018; Rattanaphani et al., 2007). Hence, a certain interaction between cotton and a range of natural dyes are well established as a favourable condition for dyeing. However, one of the critical problems with natural dyes is the low fastness properties, which might be due to their heterogeneity and often limits their application (Kasiri and Safapour, 2014). To offset this problem, several advanced dyeing techniques have been developed over the years to enhance the applicability of natural dyes and make the dyeing process environmentally more

sustainable. Table 5 lists the research works on cotton and other fibres with different natural dyes in various methods.

Table 5. Major studies on cotton, silk and wool with natural dyes

Source of Natural Clourant	Fibre dyed	Mordant	Mordanting and Dyeing process	Reference
African marigold ( <i>Tagetes erecta</i> L.)	Cotton	Alum, ferrous sulphate, copper sulphate, stannous chloride	<ul style="list-style-type: none"> <li>- Pre-mordanted for 30 min, at 80°C, m:l 1:40, 2% mordant, rinsed, squeezed, dried;</li> <li>- cotton is dyed at aquous dye extract, m:l 1:40;</li> <li>- silk sample is dyed at pH 4 with sodium acetate and acetic acid buffer;</li> <li>- washed in cold water and dried at room temperature,</li> <li>- dipped in saltwater for dye fixing.</li> </ul>	(Jothi, 2008)
	Silk			
Bacteria ( <i>Serratia sakuensis</i> )	Cotton	-	<ul style="list-style-type: none"> <li>- cotton is dyed for 60 min, 80°C, m:l 1:25 with Glauber's salt;</li> <li>- silk and wool are dyed using acid dyeing method for 60 min, 80°C, m:l 1:25;</li> <li>- cold washed with non-ionic detergent and air dried.</li> </ul>	(Vaidyanathan et al., 2012)
	Silk			
	Wool			
Gulzuba flower ( <i>Hibiscus mutabilis</i> )	Cotton	Alum, ferrous sulphate, copper sulphate, stannous chloride, potassium dichromate, stannic chloride	<ul style="list-style-type: none"> <li>- Pre-mordanted for 30 min, 60°C, m:l 1:50;</li> <li>- rinsed, squeezed, dried;</li> <li>- cotton sample is dyed with dye extract at m:l 1:40, 60 min;</li> <li>- silk sample is dyed at pH 4 by adding a buffer solution (sodium acetate and acetic acid), m:l 1:40, 60min;</li> <li>- washed with cold water and dried at room temperature;</li> <li>- dipped in brine for dye fixing.</li> </ul>	(Shanker and Vankar, 2007)
	Silk			
	Wool			
<i>Lac insect</i> ( <i>Luccifer lacca</i> )	Cotton	-	<ul style="list-style-type: none"> <li>- dyed at pH 3, m:l 1:100, varying time 5 min, 60 mim, 120 min, at different temperature 10°C, 30°C, and 60°C</li> </ul>	(Chairat et al., 2008)
Mangosteen hulls ( <i>Garcinia mangostana</i> )	Cotton	Ferrous sulphate, calcium	<ul style="list-style-type: none"> <li>- Cotton and silk are dyed for 60 min at 60°C, m:l 1:25;</li> <li>- Post-mordanted by</li> </ul>	(Chairat et al., 2007)

Linn)		hydroxide,	impregnating in mordant solution at room temperature for 30 min, - washed with water and air-dried.	
	Silk	Alum, zinc tetrafluoroborate		
Senna Tora ( <i>Cassia tora L.</i> )	Cotton	Ferrous sulphate, Copper sulphate, Zinc sulphate, Manganese sulphate , aluminium sulphate Nickel sulphate	- both cotton and silk were pre-mordanted with any mordant for 60 min at 40°C, m:l 1:100, - both are dyed for 60 min at 60°C, m:l 1:100	(Lee and Kim, 2004)
	Silk			
Turmeric rhizome ( <i>Curcuma longa</i> )	Silk	Ferrous sulphate	- pre-mordanted with 0.5 g/L FeSO <sub>4</sub> for 10 min, at 70 °C, m:l 1:20; - dyed for 100 min at different temperatures (70°C, 85°C, 100°C), pH 7, m:l 1:20; - hot washed with water at 80°C, then cold wash at room temperature, squeezed, dried for 10 min at 60°C.	(A. N. M. Haque et al., 2018)
Turmeric rhizome ( <i>Curcuma longa</i> )	Cotton	Ferrous sulphate, aluminium sulphate, zinc chloride, tannin	- Pre-mordanted for 10 min at room temperature, m:l 1:40; dried for 5 min at 130°C; -dyed for 60 min at at 60°C, m:l 1:40; - rinsed with warm and cold deionised water, air dried	(Mulec and Gorjanc, 2015)
Pomegranate ( <i>Punica granatum</i> ) and Tea leaf ( <i>Camellia sinensis</i> )	Cotton	-	- acrylamide monomer was padden on cotton fabric pre-soaked with K <sub>2</sub> S <sub>2</sub> O <sub>8</sub> , dried at 95°C for 5 min and cured at 140°C for 5 min; - dyed for 60 min at 90°C, m:l 1:50, pH 4.2 ± 0.2; - washed with non-ionic detergent for 10 min at 60°C, then cold-washed and dried.	(Maulik et al., 2011)
<i>Lac insect</i> ( <i>Luccifer lacca</i> )	Cotton	-	- cotton pretreated with chitosan was dyed for 180 min at 30°C, pH 3.0, m:l 1:100;	(Rattanaphani et al., 2007)

<i>Green tea leaf (Camellia sinensis)</i>	Cotton	-	- Ethanol extract of green teal leaf mixed with 8% citric acid is applied on cotton by pad-dry-cure at 50°C, m:l 1:20, 3psi, 100% wet pick up; - air dried and cured for 3 min at 140°C	(Syamili et al., 2012)
<i>Henna leaves (Lawsonia inermis)</i>	Cotton	copper sulphate	- UV irradiation of dye powder and cotton fabric for 60 min, - pre-mordanted with 4% copper sulphate for 120 min at 65°C, - dyed in presence of 3% NaCl for 120 min at 65°C	(Iqbal et al., 2008)
<i>Lac insect (Laccifer lacca)</i>	Cotton	-	- cationised cotton fabric was dyed for 60 min at 80°C, pH 2.5. m:l 1:40, sonic power (100-500W)	(Kamel et al., 2007)
<i>Dom sheng (Symplocos spicata)</i>	Cotton	stannous chloride, stannic chloride, ferrous sulphate, copper sulphate, and potassium dichromate	- Cotton was pretreated with 2% tannic acid for 4-5 hours and then pre-mordanted with any one mordant for 60 min at 40° C; - Silk and wool were pre-mordanted without tannin acid treatment, - pre-mordanted fabrics were dyed in sonicator process for 60 min at pH 5.09, 30-40° C, varying m:l 1:10 to 1:40	(Vankar et al., 2008a) (a)
	Silk			
	Wool			
<i>Berberine (Mahonia napaulensis DC)</i>	Cotton	alum, ferrous sulphate, stannic chloride, stannous chloride, copper sulphate, potassium dichromate.	- Cotton was pre-treated with 2% tannic acid and then pre-mordanted with 1-2% mordant (any one). - silk and wool fabrics were directly pre-mordanted - all fabrics were individually dyed for 180 min at 30-40°C without sonicator and for 60 min at 30-40°C with sonicator	(Vankar et al., 2008a) (b)
	Silk			
	wool			
<i>Catechu (Acacia catechu) and Tectona (Tectona grandis)</i>	Cotton	Ferrous sulphate, alum, copper sulphate, potassium dichromate, stannous chloride, stannic chloride	- enzyme treated cotton fabric was pre-mordanted with 2% mordant solution of for 30 mins at 45°C; - dyed at sonicator bath for 60 min at 45°C, m:l 1:30	(Vankar and Shanker, 2008)

<i>Red cabbage (Brassica oleracea)</i>	Cotton	-	- cationised for 60 min at 50°C, dried at room temperature; - dyed by ultrasonic energy for 60 min at 80°C, pH 9	(Ticha et al., 2016)
<i>Onion (Allium cepa)</i>	Cotton		- microwave plasma treated cotton was dyed in onion pulp juice and skin extraction separately for varying time 10 - 30 min at 70°C	(Chen and Chang, 2007)
<i>Pomegranate (Punica granatum)</i>	Cotton	-	- Pre-treated with plasma and chitosan - dyed for 60 min at 100°C, pH 7	(Haji, 2017)
<i>Red Calico leaves (Alternanthera bettzikana)</i>	Cotton	-	- cotton and calico leaves were gamma irradiated, - dyed for 60 min at 60°C, m:l 1:30, varying pH 5-10	(Khan et al., 2014)
<i>Note: m:l = material to liquor ratio</i>				

Chemical pre-treatment of cotton (apart from conventional mordanting) before dyeing process is a common method for improving dye exhaustion and fastness properties of textiles. In a study with berberine, a significant increase in dye exhaustion on cotton together with very good colour fastness properties (washing fastness rating of 4 and dry cleaning fastness of 4-5) was achieved when the fabric was pre-treated with an anionic bridging agent containing dichloro-s-triazinyl reactive group (Kim et al., 2004). The bridging agent surpassed the weak interaction between cotton and berberine and therefore was able to provide good results. A few other chemicals like citric acid, hydrogen peroxide, chitosan also reportedly has a better influence on the dyeability and durability of cotton when the colouration was performed with green tea leaf, curcumin, and lac, respectively (Ammayappan and Moses, 2009; Rattanaphani et al., 2007; Syamili et al., 2012).

More advanced technologies like sonication, UV irradiation, and gamma-ray radiation are also introduced to enhance dyeability from natural dyes. Several studies using the sonication method reported notable improvement in dyeing performance. This method is also called sonicator dyeing (ultrasound-assisted dyeing), which has attracted researchers in recent years due to its known profile of providing greater dye yield in a reduced amount of electrolyte, dyeing time and temperature (Kamel et al., 2007). In a study with lac dye on cotton, the



ultrasonic technique was found to have increased the dye uptake around 66% increase compared to the conventional method (Kamel et al., 2007). In a relevant study, the efficiency of dyeing was found to increase about 7-9% when cotton was dyed with eclipta dye using the sonication method instead of the conventional method (Vankar et al., 2007). Along with the dye uptake property, the fastness characteristics of dyed fabric was also found to improve by using sonicator dyeing on cotton with *Symplocos spicata* (Vankar et al., 2008b). However, it should be noted that in all these cases, chemical pre-treatment, cationization or enzyme treatment was required before the sonication (Ghorpade et al., 2000; Senthilkumar et al., 2002; Ticha et al., 2016; Vankar and Shanker, 2008).

Among the other techniques, UV irradiation of both dye and cotton was found effective for increasing colour strength with henna dye (Iqbal et al., 2008). The irradiation helped the colouring component in henna leaf (lawsone) to be degraded hydrolytically and produce bigger spaces inside cellulose structure, which synergistically resulted in a high exhaustion property of the dye onto cotton (Iqbal et al., 2008). Besides, gamma-ray radiation on both natural dye and cotton before the dyeing step was reported as an efficient technique not only because it can reduce the amount of mordants required for the process but also can boost the colour fastness (Khan et al., 2014).

The plasma treatment of fabric surface before dyeing is reported while dyeing cotton with natural dye. This, in fact, is a non-destructive modification of fabric surface which influences the dye exhaustion properties. A report on onion dye on cotton revealed a significant increase in cotton fabric hydrophilicity after plasma treatment, ceasing the necessity of using metallic salt as a mordant (Chen and Chang, 2007).

Apart from dyeing, a few studies on printing on cotton with natural dyes, i.e., alkanet, rhubarb, henna, and curcumin. Naturally sourced thickener such as meypro gum was found helpful to achieve better colour strength than commonly used synthetic thickeners in pigment printing technique (Rungruangkitkrai and Mongkholrattanasit, 2012). Application of binders instead of mordanting agent was also proved effective in case of pigment printing. Additionally, mercerized cotton showed better printability with natural dye as well as displayed higher colour efficiency (Bahtiyari et al., 2017). In case of screen printing guar gum was used as thickener. Moreover, Antibacterial property of printed cotton with natural dyes was also studied by researchers (Teli et al., 2013). They applied functional biopolymer chitosan as eco-friendly mordant with different concentration on cotton fabric before printing

with natural dyes like catechu, turmeric, marigold. The chitosan mordanted printed samples were found to have good colour values as well as excellent antibacterial activity, which opened a new door for eco-friendly printing with antibacterial finishing of textiles. The colour-fastness of printed cotton was reported between 4 and 5 for alkanet and rhubarb dyes (Rekaby et al., 2009). Besides, several natural dyes sourced from, i.e., turmeric, indigo, pomegranate, henna were reported in the literature to become successfully printed on cotton fabric in different methods, including resist printing and screen printing and having good fastness properties (Hakeim et al., 2005; Hebeish et al., 2006; Karolia and Buch, 2008). At present digital printing with water based ink-jet inks is gaining popularity around the world and offers benefits over conventional printing methods. In a recent study, a novel eco-friendly process was offered to investigate the potential application of four natural natural pigments (annatto, catch, pomegranate fruit rind, and golden dock) for the preparation of water-based ink-jet inks for digital printing on cotton fabric (Savvidis et al., 2014). Colour consistency and fastness results of printed samples after fixation were promising and this paves the way of developing suitable eco-friendly inks using natural dyes for the digital printing of cotton.

#### ***4.2. Dyeing and printing of wool***

Different natural dyes from various natural sources like Iranian and Indian madder, Gulzuba, Lac, Anthraquinone, Rutin, Weld, Tea, Walnut bark, Marigold etc. used for dyeing and printing of wool are reported. Different types of mordants and methods of mordanting techniques significantly affect the rate of fading and colour fastness property. (Shanker and Vankar, 2007) has reported that pretreating with 2 to 4% metal mordants and maintaining a material to liquor ratio of 1:40 is optimal to obtain very good fastness properties for wool, silk and cotton samples (see Table 5). A study conducted by (Gong et al., 2020) has shown the impacts of different natural mordants like pomegranate peel, citric acid, Arjun bark, chlorophyll extract etc. and the metal mordants like sodium dichromate, stannous chloride, copper sulfate, ferrous sulfate etc. on different fastness properties of wool fabrics dyed with 50% (on the weight of material) Cinnamomum camphora natural dye.

Low temperature plasma treatment is a water-free and clean process to modify the topography of of textile fibres. It can eliminate the hydrophobic nature and can partly degrade the surface scales of wool fibre without affecting its bulk properties (Haji and Qavamnia,

2015; Zhang and Wang, 2015). Plasma treatment before dyeing is found to be very effective in increasing the dye and mordant penetration into the wool by increasing the -C=O groups and by removing the lipid layers on the surface of the fibres (Haji, 2020a, 2020b; Haji et al., 2016a, 2016b, 2014; Haji and Qavamnia, 2015).

The application of ultrasound is becoming a very useful technique for many chemical processes and reactions like chemical synthesis, fabric dyeing, water treatment, materials production etc. (Cravotto and Cintas, 2007). Comparison between conventional and ultrasonic dyeing processes for dye exhaustion of wool fabric using betanin dye was carried out, were characterized by mass techniques and fractional crystallization methods were applied to extract and isolate dyes from *Opuntia ficus-indica* plant (Guesmi et al., 2013). This study revealed that the dye exhaustion rating was improved from 30% to 60% by the sonication process within a shorter dyeing period. Another study showed that the dye extraction and the dye uptake percentage of wool fabrics with lac (*Kema lacca*) dye were increased by 41% and 47%, respectively, in the ultrasonic dyeing method than conventional process (Kamel et al., 2005).

Along with the dyeing of wool fabric with natural dyes, printing is also reported in different studies. Printing of wool fabric with henna (2-hydroxy-1,4-naphthoquinone) as the natural colourant is reported by (Hakeim et al., 2003), where different fixation techniques like steaming, microwave heating, thermofixation etc. were applied, and the influence of different printing conditions like thickening agent type, mordant type, glycerine concentration, pH value of printing paste, dye concentration etc. on the fastness properties and the colour strength of the printed wool fabric was studied.

### ***4.3. Dyeing and printing of silk***

Silk, one of the delicate natural fibres, is also well examined in dyeing with different natural dyes. It is a natural protein fibre that is prone to an alkaline environment like wool, especially when the pH value is over 9. Besides, silk persists cationic nature by an isoelectric point at pH 5 (Samanta and Konar, 2011). Due to this reason, dyeing on silk with natural dyes is preferably conducted at a slightly acidic condition (see Table 5) (Jothi, 2008). Curcumin, extracted from turmeric, was successfully used to dye silk at a range of temperatures between 60-100°C using several mordants, e.g., FeSO<sub>4</sub>, C<sub>4</sub>H<sub>6</sub>O<sub>6</sub>, CuSO<sub>4</sub> (ANMA Haque et al., 2018).

It was well reported that mordanting of silk has a significant impact on the colour performance, brightness and fastness of dyed silk (Rungruangkitkrai and Mongkholrattanasit, 2012). However, among the metallic mordants, Ferrous sulphate was claimed to produce better results because of colour and fastness than other mordants (Chairat et al., 2007; Lee and Kim, 2004). A combination of two or more mordants was also found useful to achieve very good to excellent colour fastness properties (Pruthi et al., 2008).

Advanced dyeing methods are also reported for dyeing silk with natural dyes, like other major natural fibres (cotton, wool, etc.). Pre-treatment with enzymes, mainly lipase, diastase and protease–amylase before dyeing showed the enhancement in fastness property for dyed silk (Vankar and Shanker, 2009). Pre-mordanting followed by sonication was reported as an efficient method for silk to achieve better colour properties (Vankar et al., 2008b, 2008a). An increased dye diffusion was reported when silk fabric was treated with plasma before microwave-assisted dyeing (Dayioglu et al., 2015). Both the time and microwave energy in plasma treatment contribute in enhanced dye molecule diffusion into the silk polymer. In a different study, silk was dyed with natural dye with and without oxygen and argon plasma pre-treatment (Boonla and Saikrasun, 2013). The surface treatment enhanced the roughness of the silk surface, which resulted in higher adsorption capacity, especially for argon plasma treatment.

Apart from the silk dyeing approaches, there were a few attempts to print on silk fabric by natural dyes. A study of silk printing with alkanet and rhubarb colours reported the steam fixation method to be better than thermo-fixation in achieving higher colour strength (K/S value) (Rekaby et al., 2009). Besides, printing on silk fabric was also conducted using natural agents as thickeners like mango kernel gum and cassia seed gum, guar gum and a wide range of colours (varying mordant selection) and good fastness properties were reported (Babel and Gupta, 2016; Yadav et al., 2010).

#### ***4.4. Dyeing and printing of bast fibres***

Bast fibres are obtained from the bast or outer cellular layers of the stems of various plants. They have found numerous applications in traditional and technical textiles. Their advantages include local supply chain, low price, environmental friendliness, satisfactory mechanical properties and recyclability.

#### 4.4.1. *Jute fibre*

The golden fibre jute is a coarser variety of agro-renewable and eco-friendly natural fibre (Samanta et al., 2012, 2008). Most of the natural dyes applied on jute fibre are extracted from leaf, bark, wood, rind, seed, root, etc. Mordanting methods (pre, post, meta) and the types of mordant (bio or chemical) can change the depth of shade, dye uptake and fastness behaviour of jute fibre dyed with natural colourants. Pre-mordanting route favours dyeing of jute fabric with direct types of natural dyes in presence of chemical mordant aluminium sulphate (Samanta et al., 2003). In contrast, post-mordanting with chemical mordants exhibits good colour yield and fastness properties after the dyeing of bleached jute with annatto seeds (Chattopadhyay et al., 2014).

A notable number of research works has been carried out to determine the effect of bio-mordant (myrobolan) and eco-friendly chemical mordant with or without mordant assistant like metal salts such as ferrous sulphate, aluminium sulphate  $[Al_2(SO_4)_3 \cdot 18H_2O]$ , potash alum  $[K_2SO_4, Al(SO_4)_3 \cdot 24H_2O]$  on bleached jute fabric (Samanta, 2018; Samanta et al., 2006; Samanta and Agarwal, 2007). The effect of mordant on bleached jute fibre has been analyzed by using single or double mordants Myrobolan, aluminium sulphate, ferrous sulphate, potassium aluminium sulphate, stannous chloride and EDTA at different concentrations and assessed the resultant variation in surface colour strength (K/S), colour difference and mechanical properties like breaking strength, breaking extension, bending length of bleached Jute fabric. Generally, the application of any kind of mordant irrespective of its application percentage or method causes loss in fabric strength with some increase in warp way breaking extension. The value of bending length dyed fabric is found to increase when mordanted with  $FeSO_4$  or  $Al_2(SO_4)_3$  and to reduce to some extent when mordanted with EDTA. Among all the mordants, highest colour strength (K/S) and colour differences value are found for a single application of  $FeSO_4$  as mordant (without dyeing) on jute fabric as it is a transition metallic salt bearing an inherent colour. In the case of double mordanting, sequential treatment of Harda and  $FeSO_4$  on bleached jute fabric exhibits high colour difference as Harda itself is a light yellow dye that adds additional colour tone on fabric (Samanta, 2018). Some major challenges in continuing the natural dyeing process on jute fabric are reproducibility of colour, dyeing variables, types, methods and concentration of mordant, fastness properties, dye uptake, etc.

Decorative look in jute fabric can be achieved by printing. However, there is hardly any research work found on the printing of jute with natural colourants. In one research, the eco-friendly chemically processed jute fabric is printed with natural dyes extracted from roots of *manjistha*, bark of *ratanjot* and seed of *Annatto* (particle size 400-800nm) by screen print method (Chattopadhyay and Pan, 2018). The printing paste constituted different natural resins like guar gum, gum arabic, sodium alginate, and gum indulka. Bio-scouring by mixed enzyme (cellulase and xylanase) and the non-ionic surface acting agent followed by bleaching was done to make jute fabric white, bright and soft and ultimately more suitable for printing. Myrobolan and potash alum both are applied separately and sequentially (double mordant) on bleached jute fabric. After dye extraction, the printing of jute fabric is done by the screen print method, where 20 to 40 mesh size screens exhibit better performance. After assessing all the printed fabrics, it was revealed that double mordanting jute fabric and application of guar gum as a thickener in print paste shows better colour yield and fastness characteristics on printing (Chattopadhyay and Pan, 2018).

#### 4.4.2. Flax fibre

Among all bast fibres, flax fibre has fascinated the textile market because of its distinctive features such as high strength, smoothness, elegance in appearance, quick moisture absorbency, rapid dryness, hygroscopicity, high air permeability, and comfortability. However, dye penetration in flax fibre becomes very difficult because of its high crystallinity in its cellulose structure. Few research works have been conducted on dyeing of flax fibre with natural dyes. Most cases, flax fabric, commonly known as linen, was dyed at boiling temperature (Grifoni et al., 2014; Sarkar and Seal, 2003; Wang et al., 2009). Potash alum is mostly applied for mordanting in all mordanting methods (pre, post and simultaneous) on linen fabric during dyeing with different natural dyes because of its ability to form coordinated complex and to chelate with natural dyes readily (Grifoni et al., 2014; Sarkar and Seal, 2003; Teli and Pandit, 2018). Bio-mordants such as Harda and Chestnut are also used for mordanting of flax fabric. Influence of mordants on colour yield, colour strength (K/S values) and other fastness properties of *Sterculia foetida* dyed linen fabric was analysed where bio(harda) mordanted linen fabric exhibits higher colour depth and good fastness properties than alum or without mordanted fabric both in pre and post mordanting method (Teli and Pandit, 2018). Moreover, Ultraviolet radiation properties of linen fabric can also be

improved by the application of some natural dyes (extract from *Helychrysum*, *Rubia*, *Daphne*, *Lavandula*, *Cynara*, *Chelidonium majus*, *Sterculia foetida*) (El-Hennawi et al., 2017; Grifoni et al., 2014; Teli and Pandit, 2018).

#### 4.4.3. Ramie, Hemp and Banana Fibres

Besides jute and flax, other bast fibres like ramie, hemp and banana are also known to the textile industry because their versatile applications expanded from luxurious clothing material to bio-composite.

The dyeing behaviour of Ramie fibre is influenced by its high crystalline structure, which thwarts the exertion of dyestuff into fibre molecules. Four natural dyes extracted from *caesalpinia sappan*, *rhizoma coptidis*, *gardenia*, and *areca catechu* contain brazilin, berberine, crocin, and crocetin and catechin colourants respectively have been applied on ramie fabric at 90°C for 50 to 70 min in neutral condition (Zheng et al., 2011). In this work, rare earth was employed as a metallic mordant in pre, meta and post mordanting method where post mordanting showed a high dye pickup percentage. Compared to other metallic mordants, rare earth metals increased fastness properties and stabilized colour shade at different p<sup>H</sup> of naturally dyed ramie fabrics because of its formation of coordinated bond with natural dyes and fibre (Zheng et al., 2011). Ramie fabric was also dyed with curcumin in aqueous solution of NaOH/urea at low temperature without any use of mordant. The colour strength (K/S value) of ramie fabrics dyed in NaOH/urea solution found to have increased in compared to ramie fabrics dyed in water. This is because of the insertion of Na<sup>+</sup> in the macromolecular structure of ramie fibre and swelling it, resulting in the decrease of fibre crystallinity and an increase of entrance of curcumin molecules into the cellulose macromolecules' space. It was also revealed that better colouration of dyed ramie fibre obtained at low temperature as hydration of cellulosic fibres by Na<sup>+</sup> is an exothermic process and most of the curcumin decomposes in NaOH/urea aqueous solution at high temperature (S. Li et al., 2019).

Hemp fibre is known to the luxury textile industry, unlike flax and ramie. Fabric made from processed hemp fibre bears unique criteria such as proper ventilation, moisture absorbency, antimicrobial and anti-ultraviolet properties. Owing to higher strength and stiffness, it is also used in bio-composites. Hemp fibre's colour yield and colour fastness properties were found to be good, which makes it eligible for producing high value-added clothing. But very little

work has been found on the application of natural dyes on hemp fabric. This fibre was successfully dyed with a tannin-rich natural dye extracted from neem bark using water at 100°C for 60 min after pre-mordanted with  $\text{AlK}(\text{SO}_4)_2$  (Inprasit et al., 2018). Neem dyed hemp fabric got an excellent rating in colour fastness properties as hydrophobic (aromatic) and hydrophilic (-OH) groups of polyphenol compound of tannin, hydrophilic(-OH) cellulose groups of hemp and  $\text{AlK}(\text{SO}_4)_2$  mordant formed insoluble complex on hemp fabric surface which prevented leaching of dye molecule and preserve colour stability (Inprasit et al., 2018). Besides mordant, cationization process can also increase dye uptake and colour yield by changing ions on the fabric surface. Cationization on hemp fabric is applied during dyeing with the extract from Arabica coffee (Butthongkum et al., 2016).

Another most promising and eco-friendly bast fibre is banana fibre, which is collected from the pseudostem of banana plant and exhibits similar chemical properties to jute fibre. The study on application of natural dyes on banana fibre is very limited. Catechu extracted from the wood of acacia catechu can be applied on banana fibre without mordant or with different chemical mordants (alum, ferrous sulphate, stannous chloride). Experiments with banana fabric dyed at 90°C for 60 min in around neutral  $\text{p}^{\text{H}}$  and m:l 1:50 showed that mordanted banana fabric produced better colour depth after dyeing than without mordanting (Singh, 2020). Pre-mordanting method of dyeing revealed to be better in fastness properties as compared to post-mordanting and without mordanting. Banana fibre is also dyed with turmeric (*Curcuma longa*) via ultrasonic and conventional methods using various mordants (Canpolat et al., 2015). Ultrasonic methods revealed better colour yield than conventional dyeing technique, whereas Ferrous sulphate treated fabric gave better results than copper sulphate and alum.

#### ***4.5. Dyeing and printing of polyester***

Polyester is synthetic fibre with higher tensile strength, better crease recovery and higher biological resistance in comparison to natural fibres (Edlund and Albertsson, 2003; Gulrajani et al., 2008). An attempt has been made by (Baig, 2011) in dyeing of polyester (PET) with natural indigo (*Indigofera tinctoria*) in exhaust method at different pH values. The outcomes of the study suggest that in the acidic pH range polyester fabrics could best be dyed, and hydrogen bonding was found as the key contributor to dye exhaustion into the polyester fabrics. Madder (*Rubia tinctorum*) is widely used for dyeing cotton, silk, wool etc. mainly,



but not typical for polyester dyeing. However, it was reported that dyeing 100% polyester fabric with madder at different temperatures ranging from 60 to 130°C along with five different natural mordants and seven different chemicals (Gedik et al., 2014).

A novel water-free and mordant free and resource-efficient dyeing of polyester fabric with natural colourant 'curcumin' in super-critical carbon dioxide (scCO<sub>2</sub>) is trialed by (Abate et al., 2019). The dyeing behaviour of polyester fabric with natural dyes is found to be more influenced by the system pressure than the dyeing temperature. To achieve the maximum colour strength and levelness, the mentioned dyeing parameters were 120°C, 25 MPa and 60 min. Walnut (*Juglans regia*) shells are also testified as a natural source of colour for polyester and polyester/viscose blended fabric dyeing (Eser et al., 2016). To improve dyeability and activate the polyester fibre, UV/ozone pre-treatment for 5 to 120 min is reported to apply prior to the dyeing procedure. The polyester fabric was dyed with Curcumin and saffron as a source of natural dye (Elnagar et al., 2014). Apart from the polyester dyeing approaches, there are several works on polyester fabric printing with natural dyestuffs. In a bio-technique approach, laccase enzyme was employed as an alternate of mordants to fix three different natural colourants, i.e. rutin, morin and quercetin, onto the polyester fabric (El-Hennawi et al., 2012). The laccase enzyme was reported to undergo enzymatic oxidative polymerization of the dyestuffs and helps achieve very good to excellent colour fastness. In another study, hiren alkanet (Root of alkanet plant) dye nanoparticles were reported to use for printing of polyester fabric by dye-printing technique and pigment-printing technique (Taha et al., 2020). Both the printing techniques were reported to provide very good to excellent fastness properties with higher colour strength. As a part of obtaining antimicrobial properties in printed polyester fabrics, chitosan biopolymer is reported to use in making a print paste with *Curcuma tinctoria* as a natural colourant and gelatinized starch (Abdou et al., 2013). The study reported that the use of chitosan helped to improve the dye uptake percentage of polyester fabric along with providing antimicrobial characteristics.

#### **4.6. Dyeing and Printing of Nylon**

Nylon is a synthetic fibre that contains amide groups in the polymer chain. Generally, acid dyes are used to colour nylon, but scientists are trying to apply vegetable dyes to nylon. It is reported that plant dyes like Turmeric and Rhubarb can be used to print nylon fabric using cellulase enzyme, which helps to extract the dye and besides this presence of benzyl alcohol

or dimethylformamide in the printing paste quicken the dye diffusion in the fibre during steaming (Maamoun et al., 2007). Effect of different mordants on nylon is observed using onion, lac, and turmeric as natural colourants, where nylon was pre-mordanted at 100°C for 45 minutes, and the simple aqueous extraction process was used to extract colouring materials. Dyeing of nylon fabric was done in the open bath process, and high temperature high pressure (HTHP) process and all dyes give a good depth of shade at acidic conditions. Safflower, Madder, Curcumin, Saffron and Ratanjot also provide good colour values and colour fastness properties on nylon using different mordants. Besides these plant dyes, liquid waste of Eucalyptus from lumber steaming also gives excellent wash fastness on nylon (Purwar, 2016).

#### ***4.7. Dye-fabric interactions and mordants***

The interaction between natural colourants and textile fabrics can vary based on the chemical nature of the colourants and the types fibre. In cases where the dye molecules are water soluble and have a planer molecular structure and possessing conjugated bonds (such as turmeric, pomegranate, harda), and are substantive towards cellulose, they can directly be attached onto fabric at a high temperature (e.g., 100 °C), similar to the application of synthetic direct dyes (Samanta and Konar, 2011). However, some of the natural dyes possess either sulphonic or carboxylic groups that are more suitable for producing interconnection with the amino groups of wool or silk (such as saffron), while some dyes deliver cations (e.g., berberine) and produce covalent bonds with the carboxylic groups of protein fibres (Gupta, 2019). Several other natural dyes (such as indigo) are water insoluble and require transformation into water-soluble form by reduction reaction and then further oxidation onto the fabric during the final dyeing (Samanta and Konar, 2011). Nevertheless, natural dyes overall show a relatively low affinity (compared to synthetic dyes) towards textiles, and some of them have least or no affinity. As mentioned earlier in Section 2.4, often mordants are used to overcome this problem. Mordants produce a dye-mordant complex and thus enhance the affinity towards the fabric. This phenomenon not only results in a higher colour build-up, but also promotes a better colour fastness (Purwar, 2016). As the mordant, a number of tannins (such as hydrolysable and condensed) and metallic salts (such as ferrous sulphate, aluminum sulphate, zinc chloride) are frequently used. However, these synthetic mordants are at the end

responsible for polluting the effluent water, which lessens the greener prospect of using natural dyes.

#### **4.8. Role of bio-mordants**

Currently, a decent interest is growing on the use of bio-mordants that are sourced from completely natural origins. These mordants have already delivered promising results and comprehensive reports are already available in the literature. For example, a recent report from Habib et al. showed improved colour depth while using either zeera (*Cuminum cyminum*), harmal (*Peganum harmala*), illaichi (*Elettaria cardamomum*) or neem (*Azadirachta indica*) leaves extract as the mordant (Habib et al., 2021). In another study, turmeric (*Curcuma longa*) as a bio-mordant showed a higher colour strength (K/S 13.8) than synthetic mordants such as iron (K/S 2.09) during silk dyeing with coconut coir (Adeel et al., 2021). In a further report of silk dyeing, both the colour strength and fastness properties achieved by different natural mordants (such as individual extract of pomegranate, henna, acacia, rose or turmeric) were comparable with those achieved by using synthetic mordants (Adeel et al., 2020). Similar successes of using bio-mordants were also confirmed in case of protein-based natural dyes, such as cochineal (Amin et al., 2020). More encouragingly, a significant amount studies have validated the effective role of different bio-mordants in dyeing with natural dyes where a number of different dyeing techniques, such as common exhaustion method (Haji, 2010), microwave radiation (Zuber et al., 2019), ultrasonic treatment (Arifeen et al., 2021) and plasma treatment (Haji and Naebe, 2020) were used. Thus, the reported findings show the suitability and applicability of the bio-mordants in a varied range of application.

*Table 6 Categories of synthetic and bio-mordants*

<b>Categories</b>	<b>Examples</b>	<b>Sustainability issues</b>	<b>Reference</b>
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Synthetic mordants	metallic salts such as sulfates (magnesium, aluminum, zinc, copper, cobalt, nickel, manganese or tin), chlorides (stannic, ferric, copper, zinc, aluminum and even neodymium or zirconium), hydroxides (calcium) and oxides (ferric or lanthanum)	Except alum and iron, most mordants are not environmentally safe and restricted by eco-certification. The generated effluent can cause water and soil contamination.	(İşmal, 2017; İşmal and Yıldırım, 2019; Rahman et al., 2013; Rovira et al., 2015; Shahid et al., 2013; Shahid and Mohammad, 2013; P S Vankar, 2017)
Biomordants	Vegetable tannin-containing substances such as myrobolan (Harda, <i>Terminalia chebula</i> ), oak galls, sumac, or pomegranate rind, Zeera ( <i>Cuminum Cyminum</i> ), Hermal ( <i>Peganum Harmala</i> ), Illaichi ( <i>Elettaria Cardamomum</i> ) Neem ( <i>Azadirachta Indica</i> ) extract.	Environmentally alternative of synthetic mordants, however, colour performance and fastness are mostly unsatisfactory. In some cases, biomordants have therapeutic and ayurvedic properties.	(Cunningham et al., 2011; İşmal, 2017; Prabhu and Teli, 2014)

## 5. Functional treatments with natural colourants

Major chemical compounds found in natural dyes are indigoid, pyridine, carotenoid, quinonoid, flavonoids, betalains, anthocyanin, anthraquinone, and tannins provide several functional properties when applied on textiles. The interaction between these dyes and the functional groups ( $-OH$ ,  $-SO_3H$ ,  $-COOH$ ,  $-C_6H_5OH$ ,  $-NH_2$ ) of different textile fibres can make a complex structure responsible for achieving such characteristics. A summary of natural dyes with a responsible chemical component for the specific functional property is presented in Table 7, but not discussed with further details in this section as it will go beyond the scope of this review.

Table 7. Natural dyes with the responsible chemical components for defined property

Functional Property	Natural Dye with Botanic Name	Responsible component for functional properties	Substrate tested with	References
Antimicrobial	Pomegranate peel ( <i>Punica granatum</i> )	Hydrolysable tannins((pyrogallol)	Lyocell	(Rehman et al.,

				2018)
Mosquito repellence	Pomegranate peel ( <i>Punica granatum</i> ) with different conc. of polyvinyl alcohol	<ul style="list-style-type: none"> <li>• Ellagic tannin</li> <li>• Polyvinyl alcohol</li> </ul>	Cotton	(Rimpi and Archana, 2017)
Antimicrobial, Mothproofing property	Henna ( <i>Lawsonia inermis</i> )	Lawsonone (2-hydroxy-1,4-naphthoquinone)	Wool	(Nazari, 2017; Yusuf et al., 2015)
Antibacterial and antioxidant	Chitosan treated Henna dyed ( <i>Lawsonia inermis</i> )	Polysaccharide of (1-4)-linked D-glucosamine and N acetyl-D-glucosamine with its polycationic structure Lawsonone (2-hydroxy-1,4-naphthoquinone)	Jute	(Bhuiyan et al., 2017)
Antimicrobial	Chitosan treated tea ( <i>Camellia sinensis</i> )	Polysaccharide of (1-4)-linked D-glucosamine and N acetyl-D-glucosamine with its polycationic structure. Polyphenol	Wool	(Shahid-ul-Islam, B.S. Butola, 2018)
Antibacterial	Bakayan ( <i>Melia Composita</i> )	Alkaloids and limonoids	Silk, wool, Cotton	(Pal et al., 2016)
Antibacterial and UV protection	Madder ( <i>Rubia tinctorum</i> ) and Safflower ( <i>Carthamus tinctorius</i> )	di- and tri hydroxyl anthraquinones and Carthamin respectively	Polyamide 6	(Ibrahim et al., 2013)
Antibacterial	Neem ( <i>Melia azedarach</i> )	Phenolic compounds and flavonoids	Hemp	(Inprasit et al., 2018)
Antimicrobial	Golden dock ( <i>Rumex maritimus</i> ) Indigo ( <i>Quercus infectoria</i> )	Tannin	Wool	(Singh et al., 2005)
Antibacterial	Berberine ( <i>berberis vulgaris</i> )	Berberine (a quaternary ammonium structure)	Wool	(Aminod din, 2012)
Antibacterial (but no activity against <i>Klebsiella pneumonia</i> )	Peony ( <i>Paeonia officinalis</i> )	Paenol/paenoside/paeonolide/paenoniflorin	Cotton, Wool, Silk	(Lee et al., 2009)

Antimicrobial	<ul style="list-style-type: none"> <li>• Goldthread (<i>Coptis Chinensis</i>)</li> <li>• Clove (<i>Syzygium aromaticum</i>)</li> <li>• Gallnut</li> <li>• Pomegranate</li> </ul>	<ul style="list-style-type: none"> <li>• Alkaloid berberine</li> <li>• Eugenol (2-methoxy-4 allyl-phenol)</li> <li>• Penta-m-digalloyl-<math>\beta</math>-glucose, gallic acid</li> <li>• Ellagic acid and tannin</li> </ul>	Cotton, Wool, Silk	(Lee et al., 2009)
UV protection, Antibacterial, Antioxidant	Modified Curcumin ( <i>Curcuma Longa L.</i> ) by reactive UV absorber	Curcumin react with oxalanilide and sulfatoethylsulfone group of UV absorber	Silk	(Zhou and Tang, 2016)
Mothproofing property	Walnut hull ( <i>Juglans regia L.</i> )	Juglone (5-hydroxy-1,4-naphthoquinone)	Wool	(Nazari, 2017)
Antibacterial, Antioxidant, Flame retardant, Waterproof	<i>Dioscorea cirrhosa</i> tuber	Condensed Tannins (proanthocyanidins)	Silk	(Yang et al., 2018)
Antibacterial, Antioxidant	<i>Lycium ruthenicum Murray</i>	Acylated Anthocyanins	Wool	(Dong et al., 2019)
Antimicrobial and UV Protection	<i>Quercus infectoria</i> and <i>Acacia Catechu</i> , <i>Acacia nilotica</i> , <i>Kerria lacca</i> , <i>Mallotus philippinensis</i> <i>Rumex maritimus</i> and <i>Rubia cordifolia</i>	Tannin	Cotton	(Gupta et al., 2005)
UV Protection	<ul style="list-style-type: none"> <li>• Eucalyptus leaf extract (<i>Eucalyptus camaldulensis</i>)</li> <li>• Orange Peel</li> <li>• Blossoms of broom (<i>Cytisus scoparius</i>)</li> <li>• Dandelion (<i>Taraxacum officinale</i>)</li> </ul>	<ul style="list-style-type: none"> <li>• Tannin as major and flavonoids as minor</li> <li>• Phenolic compounds (Narirutin and Hesperidin)</li> <li>• Flavonoids</li> <li>• carotenoids</li> </ul>	Wool	(Hou et al., 2013; Křížová and Wiener, 2016; Mongkh olrattana sit et al., 2014)
UV Protection	Cannonball mangrove ( <i>Xylocarpus granatum</i> ) bark extract.	Tannin	Cotton	(Pisitsak et al., 2016)
UV Protection	Flos Caryophyllata (clove) and acutissima shell	Tannin	Silk	(Cao et al., 2013)

UV Protection	Weld ( <i>Reseda luteola L.</i> ), Woad ( <i>Isatis tinctoria L.</i> ), Logwood( <i>Haematoxylon campechianum L.</i> ), Lipstick tree ( <i>Bixa orellana L.</i> ), Madder( <i>Rubia tinctoria L.</i> ),Brazil wood ( <i>Caesalpinha brasiliensis L.</i> )	Flavonoid	Hemp and Flax	(Grifoni et al., 2009)
UV Protection	Henna ( <i>Lawsonia inermis</i> )	Lawson; 2-hydroxy-1,4-naphthaquinone in Henna leaves	Cationic cotton	(Omer et al., 2015)
UV Protection	Turmeric( <i>Curcuma longa</i> )	Curcumin	Polyester	(Sriumao et al., 2014)
UV Protection	<ul style="list-style-type: none"> <li>• Cochineal (<i>Dactylopius Coccus costal</i>)</li> <li>• Weld (<i>Reseda luteola</i>)</li> </ul>	<ul style="list-style-type: none"> <li>• Carminic acid (a large glucose units attached by anthraquinone moiety)</li> <li>• Luteolin and some Apigenin</li> </ul>	Silk and Wool	(Gawish et al., 2016)
UV protection, deodorization, antimicrobial properties	<ul style="list-style-type: none"> <li>• Gallnuts (Cynipidae)</li> <li>• Areca nuts (<i>Areca catechu L.</i>)</li> <li>• Pomegranate peels(<i>Punica granatum L.</i>)</li> </ul>	<ul style="list-style-type: none"> <li>• Tannin (Gallnut tannin)</li> <li>• Tannin (Catechuins and Epicatechuins)</li> <li>• Pomegranate Tannin (Ellagic tannin)</li> </ul>	Silk	(Jung, 2016)
UV Protection and antibacterial	<ul style="list-style-type: none"> <li>• Gromwell (<i>Lithospermum erythrorhizon</i>)</li> <li>• Gallnut</li> <li>• Banana Peel (<i>Musa, cv. Cavendish</i>)</li> </ul>	<ul style="list-style-type: none"> <li>• Deoxyshikonin</li> <li>• Tannin</li> <li>• Luteolin</li> </ul>	Cotton	(Hong et al., 2012; Salah, 2013)
AntiMoth	Silver oak, Walnut husk and Pomegranate rind	More than 40% tannin	Wool	(Shakya et al., 2015)
Deodorizing	<ul style="list-style-type: none"> <li>• Gardenia</li> <li>• Coffee sludge</li> <li>• <i>Cassia tora. L.</i></li> <li>• Pomegranate extract</li> </ul>	<ul style="list-style-type: none"> <li>• Crocin</li> <li>• Caffeine</li> <li>• Antraquinones</li> <li>• Ellagic acid</li> </ul>	Cotton, Silk, Wool	(Hwang et al., 2008)

Although natural dyes can improve the functional properties of textile materials, they are not free from limitations that demand more research and development. The imparted functional properties like antimicrobial, antioxidant, antimoth or UV protection tend to decrease after several washing. So, more research should be done to enhance the stability of functional characteristics of natural dyes after laundering.

## **6. Advantages and challenges**

Production of natural colourants encourages plantation, which can reduce CO<sub>2</sub> from the atmosphere and increase the level of O<sub>2</sub> in the atmosphere. Historically natural colourants were the primary colourants for textiles. Gradually synthetic colourants have grabbed the whole market of commercial textile colouration. Still, the associated industrial activities for producing synthetic colourants are harmful to the environment and climate. Time has come to move towards the natural colourants. Figure 10 represents the comparative positions of natural dyes over synthetic dyes. Increasing the plantation of colourant-bearing plant species can contribute to socio-economic benefits like poverty elevation through job creation, utilisation of barren lands, maintaining the fertility of arable lands through crop rotation, and greenhouse gas reduction. However, to achieve this, a range of technical, logistical and commercial challenges need to be overcome first (Zerin et al., 2019).



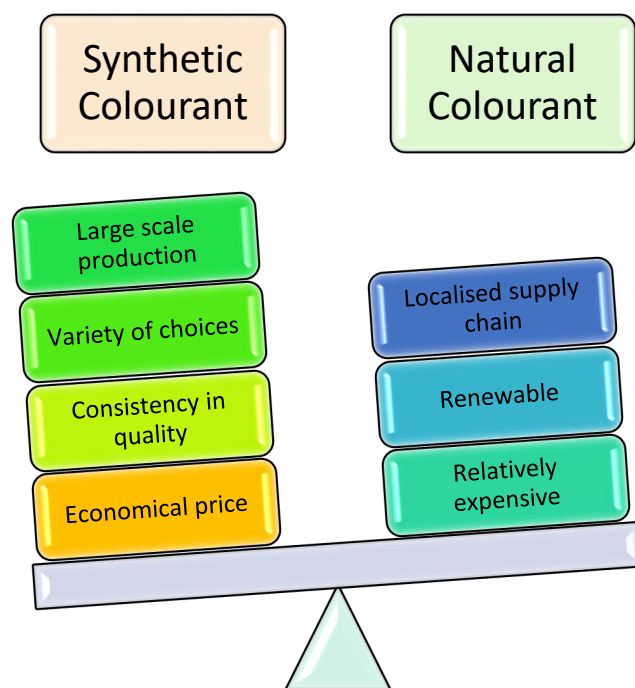


Figure 10. Comparative positions of natural colourants over synthetic colourants.

### ***6.1 High volume production***

Yield of the natural colourants is usually very low in their sources. Adopting non-traditional extraction methods such as ultrasonic, microwave and enzyme-assisted extraction (Samanta et al., 2018) and biotechnological interventions such as genetic engineering (Saxena and Raja, 2014) can improve the production rate and volume of natural colourants.

### ***6.2 Consistency in colourants and colouration***

As natural sources of colourants are not commercially produced at a large scale, proper grading and sorting of sources and colourants are not practised widely, like it is done for commercial crop production. As a result, the natural colourants available on the market nowadays are not consistent in quality, which results in inconsistent quality in coloured materials. This required a combined initiative from all stakeholders, including governments, farmers and industry leaders, to establish standard production and colouration processes and promoting them throughout the industry to get the true benefit of natural colourants.

### **6.3 Need of mordanting**

As it is mentioned earlier, the non-substantive characteristic of natural colourants requires the use of mordants in colouration process. Synthetic mordants are responsible for generating harmful effluent. This is an area of improvement to popularise natural colourants. There have been plenty of research works on the extraction and application of natural colourants in the past. Now it is high time to concentrate on making the colouration process eco-safe too. Promoting the use of natural mordants and making them available for the industry is a crucial challenge to meet.

### **6.4 Colour Fastness**

Achieving high colour fastness to wash and light with natural colourants are sometimes difficult with or without mordants and it varies depending on the natural sources and types of mordant used (Samanta and Konar, 2011). As reported in section 2.4, mordant dyes usually impart deep shades with excellent wash and light fastness onto wool. However, cotton fibre seems to achieve low colour fastness to wash from natural colourants (see section 4.1). Pre-treated cotton with an anionic bridging agent containing dichloro-s-triazinyl reactive group was found to increase the colour fastness properties (washing fastness rating of 4 and dry-cleaning fastness of 4-5) when dyed with berberine (Kim et al., 2004). Sonicator dyeing on cotton with *Symplocos spicata* also can increase dye uptake and fastness properties in comparison to traditional exhaust dyeing (Vankar et al., 2008b). Gamma-ray radiation on both natural dye and cotton before the dyeing can boost the colour fastness (Khan et al., 2014). Pre-treatment of silk with lipase, diastase and protease–amylase enzymes before dyeing showed the enhancement in fastness property for dyed silk (Vankar and Shanker, 2009). Textile substrate dyed with marigold extract using iron and copper sulphate as mordant exhibit very good light and wash fastness at first instance but it starts to degrade after a few washes with soap (Jothi, 2008). On the other hand, wash and light fastness of dyed cotton, wool and silk fabric with dye extract from *Serratia marcescens* bacteria found to be low (grades 1 for light and 1–2 for washing) but the fastness against rubbing was found to be satisfactory (grade 4–5 for silk and cotton and grade 3 for wool) (Vaidyanathan et al., 2012).

### ***6.5 Price and efficiency***

Natural colourants usually have low yield, which ultimately increases the net cost of production (Zerin et al., 2019). Due to relatively low demand and supply, their prices are also relatively high when compared with their synthetic counterparts. The supply chain of natural colourants are not well established globally. In most cases they can be only sources locally in certain places and countries. This problem cannot be solved until any large volume production of natural colourants takes place. Consumers and textile professionals cannot solve this by themselves unless governmental policies and financing institutions supporting and promoting natural colourants do not come into existence.

Similarly the efficiency of colouration with natural colourant is also not very notable. The efficiency in case of natural colourant depends on the efficiency of the extraction process, the yield of colourants, efficiency of the dyeing process including use of ingredients, water and energy and ultimate colour strength of textiles and fastness. There is hardly any literature that report the complete efficiency of colouration with natural colourants. However, innovations in extraction process and dyeing processes are said to be able to improve the efficiency of natural dye extraction, the ultimate yield, and dye uptake and colour strength (Zerin et al., 2019). For example, the use of ultrasonics in extraction and dyeing can save energy by reducing requirements of temperature and time, reduce pollution by decreasing the need to auxiliary chemical and increasing dye uptake and minimise the fibre damage, thus increases the overall efficiency through quality enhancement and lowering of processing cost (Shahid et al., 2013)

## **7 Conclusion**

Natural colourants have a glorious past but a gloomy present in the textile industry. As the production of natural colourants promote plantation and the substances themselves are biodegradable, they attract a lot of sympathies from the environmentalists and show a lot of promise for future. But in fact, the efficiency of colouration with natural colourant is not very much notable and the need of mordants result into the production notorious effluent as well. The sections 3 and 4 identified the innovations in extraction of natural colourants and dyeing processes with those colourants that can ensure higher yield and better quality in coloured textiles. At present, natural colourants are only surviving with in cottage industries.

To revive the glory of the natural colourants in textiles and enjoy true environmental benefits out of them, it is essential to have national and international policy support and financial investment to set up large scale production of colourants and colouration facilities with selected colourants, which are found to be more promising than others.

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