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Review Paper

Exploring and Exploiting Plants Extracts as the Natural Dyes/Antimicrobials in Textiles Processing

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ABSTRACT

The large number of publications in the field of extraction, purification, modification, process optimization of natural dyes and their application on textiles demonstrates the revival of natural dyes on textile coloration. The use of natural dyes is growing considerably because of the quality of the natural dyestuff obtained, the environmental compatibility of the dyes and the substantial minimization of the processing costs. Natural dyes are extracted from different parts of plants such as bark, leaf, root, fruit, seed, and flower that contain coloring materials like tannin, flavonoids, quinonoids, etc. In this paper, it has been tried to give the latest scientific overview on extraction and application of natural colorants on textile processing. The benefits and limitations of application on textiles along with different methods of extraction of natural colorants have been discussed. Moreover, the examples of early applications of natural dyes on textile processing have been reviewed. Prog. Color Colorants Coat. 8 (2015), 87-114 © Institute for Color Science and Technology.

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1. Introduction

From the past times, natural colorants have been used for numerous purposes including tribal or funeral ceremonies, body painting, decorative art and clothing or domestic decoration. At first, colorants were mainly extracted from minerals, insects and plants [1]. But since 1856, the synthetic colorants became more important and consequently, natural colorants were increasingly replaced by synthetic ones. Almost all of the synthetic colorants, which are being synthesized from petrochemical sources through hazardous chemical processes, pose threat towards the environment and human body health [2-6].

Textile industry produces large amount of hazardous wastewater containing various synthetic colorants. Recently, numerous researches have focused on development of totally new technologies and/or modification of conventional technologies for cleaner and environmentally friendlier production and treatment processes of textiles [7]. Hence, the use of non toxic, antimicrobial and eco-friendly natural dyes on textiles, preferably on natural fiber products, has recently become more and more important [8]. One of the advantages of natural dyes is eco-friendliness, i.e., they do not produce any environmental problems at the stage of production or consumption and, maintain the ecological balance [9, 10]. In addition, most of natural dyes have inherently antimicrobial properties and consequently, could possess high medicinal activity.

Natural dyes are extracted from different parts of plants including bark (e.g. Purple bark, Sappan wood, Shillicorai, Khair, Red and Sandalwood), leaf (e.g. Indigo, Henna, Eucalyptus, Tea, Cardamon, Coral Jasmine, Lemon Grass), root (e.g. Turmeric, Madder, Onions, Beet-root), fruits or seeds (e.g. Latkan, Pomegranate rind, Beetle nut, Myrobolan) and flower (e.g. Marigold, Dahlia, Tesu, Kusum) that contain coloring materials like tannin, flavonoids, quinonoids, etc. [11] (Figure 1). There are, of course, some drawbacks associated with the application of natural dyes. These dyes have poor fastness and the resulting colors have poor quality. Moreover, it is difficult to reproduce shades using natural dyes [12].

In this report, it has been tried to overview the results of the latest publications (mostly published in the last decade) on extraction and application of natural dyes/antimicrobials as effective colorants and antibacterial agents on textiles processing. Different methods of extraction of natural dyes/antimicrobials are discussed and examples of early applications of these colorants on textile processing are presented. Moreover, resulting properties obtained along with the advantages and disadvantages are reviewed.

Figure 1: Natural dyes are extracted from different parts of plants such as **A:** bark (Purple bark), **B:** leaf (Eucalyptus), **C:** flower (Marigold), **D:** fruits (Pomegranate rind) and **E:** root (Madder)

2. Plants extracts as natural dyes/antimicrobials

Despite various benefits of natural dyes/antimicrobials, there are, however, some drawbacks restricting the application of these beneficial natural materials [13-17]. In the following paragraphs, some important benefits along with restrictions of natural dyes/antimicrobials applications have been summarized.

2.1. Advantages

The results of reviewed works revealed that the growing global interest toward the application of dyes/antimicrobials from natural resources is mainly due to the following advantages:

- Natural dyes/antimicrobials are obtained from inexpensive renewable resources with considerable potential.
- Natural dyes produce a variety of uncommon, eye-catching, and soothing shades on textiles.
- Wide range of shades can be produced with an individual natural dye either in mixture with mordants or by variation in dyeing condition.
- Different natural constituents such as auxiliary, mordant, fixing component, etc. along with the natural dye may enhance dyeing/finishing process efficiency.
- Some natural dyes have intrinsic additional properties such as antibacterial, moth proof, anti-allergy, anti-ultra violet (UV) irradiation, etc.

Natural dyes/antimicrobials are environmentally friendlier than synthetic ones because: I) in natural materials, all synthesis processes are accomplished by nature with no pollution of environment and II) these materials are readily biodegradable and do not produce hazardous effluent upon degradation in environment, and so, there is no need for further treatment of effluent before discharging into the environment.

- Color characteristics of some natural dyes mellow with time. This characteristic imparts unique properties and appearance to the stuff dyed. An old carpet piles dyed with natural dyes could be an example.
- Majority of natural dyes/antimicrobials are extracted from wild and self-growing plants that require no additional cost for the cultivation.

2.2. Limitations

Natural dyes/antimicrobials have some drawbacks that limit their application on textiles. Some important ones can be summarized as follows:

- Generally, natural dyes/antimicrobials have low wash, rub, light, sweat, and gas fastness on textiles probably due to their weak bonding and interaction to the textiles.
- Color matching and reproduction of color is another disadvantage of natural dyes since the quantity and quality of natural dyes steadily change with climate, plant genus, region, etc.
- The efficiency of the natural dyes/antimicrobials extraction process is generally low, i.e., just few grams of natural component per kilogram of raw materials.

3. Methods of extraction

Extraction of dyes/antimicrobials from the natural sources is the most important step to achieve the desired dyeing properties and/or antimicrobial activities on textiles. Moreover, obtaining a standard extraction process and optimizing the influential variables for a particular natural source are economically important and affect the price of products. The different methods of extraction of these materials from natural resources would be discussed in the following paragraphs.

3.1. Aqueous extraction

The chemical structure of the most natural dyes/antimicrobials allows the use of simple set-up and cost-effective aqueous extraction technique that is also the most widely used one. Routinely, dried and finely cut material of natural resource is ground in powdered form and then, the dye/antimicrobial is extracted in water employing different standard processes. For instance, the aqueous solution could be filtered and concentrated under reduced pressure (rotary evaporator) to give a crude extract [18].

In natural dye extraction process, selecting a proper solvent is very important. In a very recent work [19], different solvents were examined in order to optimize the natural dye extraction yield from *Gardenia jasminoides Ellis,* containing crocin as the main colorant. Fifteen kinds of solvents were examined and those which showed colors close to the target color were each mixed with water at different ratios to investigate the condition that would yield color nearest to the target color. Four solvents (methanol, ethanol, 1-propanol, 2-propanol), with Hansen solubility parameters similar to that of crocin and low chi parameter values, were chosen to achieve dyes whose color had high absorbance and were as close to the target color as possible. Four solvents were then mixed with distilled water at various volume ratios. Based on chromaticity analysis, using the pure solvent ratio of 40–60 vol%, L*a*b* values were as close to those of the target color as possible. It was therefore concluded that natural yellow dyes can be extracted by using a mixture of alcohol and distilled water.

3.2. Alcoholic/organic solvent extraction

The non-aqueous extraction methods are widely used for extraction of some organic dyes/antimicrobials from natural resources. In this method, a mixture of organic solvents, usually alcohols with hexane, is used for extraction of natural dyes. As an example, the roots of the authentic sample of *Ratanjot* were air-dried and ground to coarse powder. The powdered roots were extracted with *n*-hexane in soxhlet apparatus (Figure 2) till the color of the decoction became very light. Solvent was removed and the yield percentage of the hexane extraction from the roots was calculated [20].

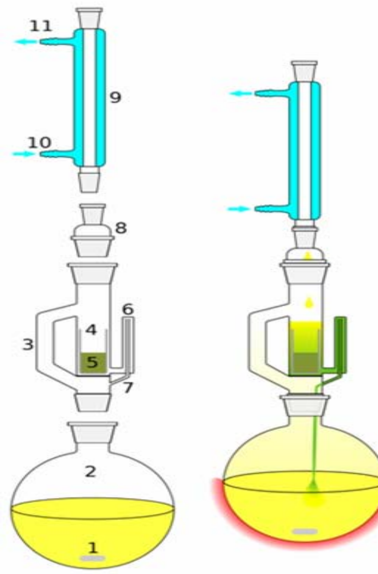


Figure 2: A schematic representation of a Soxhlet extractor; 1: Stirrer bar 2: Still pot 3: Distillation path 4: Thimble 5: Solid 6: Siphon top 7: Siphon exit 8: Expansion adapter 9: Condenser 10: Cooling water in 11: Cooling water out.

3.3. Ultrasonic extraction

In this method, the energy of ultrasonic waves enhances the yield of extraction. Ultrasound improves the efficiency of different chemical and physical processes during a known mechanism called "cavitation in liquid media", which includes the growth and explosive collapse of microscopic bubbles producing localized high temperature and pressure, shock wave, and shear force breaking chemical bonds. Numerous attempts have been paid to explore and exploit this method in the extraction and application of colorants [21]. Extraction bath pH, salt concentration, ultrasonic power, extraction time and temperature are the most influencing parameters on the process efficiency. For instance, Kamel, et al., [22] have studied the ultrasonic extraction of natural dye lac with varying percentages of the dye materials (2-12%) at different temperatures (50-80 °C) using different ultrasonic powers (100-500 W) and time intervals (20-120 min) that resulted different amounts of dyeing material.

3.4. Enzyme assisted extraction

Nowadays, this state of art extraction method is being widely used due to its high efficiency comparing the conventional methods. As a recipe for enzymatic extraction technique, a 2% solution of pectinase:cellulase (2:1) was sprayed on pomegranate rind (25 g) for achieving better soaking and contact and then, was left overnight. The enzyme treated material was washed with little amount of distilled water and pH of the solution was adjusted to 10. The sample was shaken at 150 rpm for 20-80 min at desired temperature. The contents of beakers were filtered through standard test sieve to remove solid materials and the concentrated dye extract was vacuum evaporated in a rotary vacuum evaporator to about half of the original volume and finally, the concentrated liquid was spray-dried [23].

3.5. Microwave assisted extraction

Microwave is a rather fast heating (volumetric heating) compared to conventional surface heating. The extraction time and energy input are supposed to be mostly decreased using microwave assisted extraction. Much attention has been recently paid by various researchers on the exploiting microwave energy as a modern technology in the extraction of natural materials. This is mainly because of cleaner nature of the process, lower energy consumption, and higher yield over conventional extraction methods. Dabiri, et al., [24] have optimized the microwave assisted extraction (MAE) process of two common dyes, alizarin and purpurin, from Rubiaceae plants and compared its efficacy with ultrasonic and reflux extraction methods. The superiority of MAE was concluded from the higher amount of crude material obtained (ranging from 84 to 94%), reduced extraction time (20 min vs. 6 h), and lower solvent consumption (20 vs. 100 ml). Other example for MAE process is natural dye extraction from pomegranate rind and Annatto, *Bixa orellana*, [25]. Influencing parameters, i.e., extraction time, pH of the solution and the weight of crude material have been modeled, optimized, and compared with the conventional method ones. The superiority of MAE over conventional aqueous extraction method was deduced from improved dye yield and marked decrease in extraction time.

It was also found that the dyeability of acrylic fibers could be significantly improved under microwave irradiation as a result of the increased adsorption of the dye into fibers due to the local overheating and an amplified reaction probability between the dye and fibers [26]. Table 1 summarizes the advantages and drawbacks of the above-mentioned extraction methods.

4. Early application of natural dyes/antimicrobials on textiles

Coloring materials are extracted from different parts of the plants such as bark, leaf, root, seed, and flower containing common coloring structures such as tannin, flavonoids, quinonoids, etc. In the following parts, the examples of early applications of these extracts are classified by according to the different parts of plants used as the natural resources.

4.1. Dyes/antimicrobials from root

Root is one of the common parts of the plants which have been extensively used for dyes/antimicrobials extraction (Table 2).

Table 1: Comparison of benefits and weaknesses of various extraction methods.

Extraction method	Benefits	Weaknesses
Conventional aqueous extraction	•Very simple method •Flexibility in the quantity of thematerial being extracted •No degradation upon extractiondue to low temperature of process	•Process efficiency is generally low •Slow and time consuming process •Utilizes high volume of solvents
Reflux (soxhlet) extraction	•Simple set-up •No further filtration needed due to simultaneous filtration upon extraction •Ability to recover solvents	•Very slow process •Large volume of solvent required •Thermal degradation at high temperatures and long times •Health and environment hazards due to use of toxic solvents
Ultrasonic extraction	•Rapid process •Highly efficient method •Energy saving through lower temperatures	•Limited extraction volume and yield
Enzyme assisted extraction	•Process eco-friendliness •Much higher yield over conventionalmethods •Very useful when selective chemical extraction required	•Enzymes performance is very sensitive to temperature, pH, metal ions, etc.
Microwave assisted extraction	•Simple method •Cleaner process •Lower extraction time •Low volume of solvent required	•High cost of microwave set-up •Limited extraction volume

As an example of application, the dyeing process of nylon with the extract of madder, the ground root of the *Rubia* species with anthraquinone substituted chemical structure, has been studied by Gupta, et al., [27]. Purpurin, which has 1,2,4-trihydroxyanthraquinone structure, was extracted from madder and was used for dyeing of nylon fibers. The authors have reported that the extracted red pigment has good affinity for the nylon fibers.

The light fastness of madder extracts applied on nylon has also been reported in a related work [28]. The authors have extracted two pigments, purpurin and munjistin, from Indian madder by soaking and extraction with alum and applied it for dyeing of nylon with ferrous sulfate, copper sulfate, alum and stannous chloride as mordants. The authors have reported a good light fastness for the fabrics while the dye fastness of the fabrics dyed with munjistin, was poor.

Also, wool yarns were dyed with madder along with aluminium potassium sulfate as the mordant and then, treated with different ammonia solutions by Montazer and Parvinzadeh [29]. During wash fastness tests, a change of color was observed on the dyed samples while the light fastness tests showed more fading of the madder-dyed yarns after ammonia treatment.

The effect of ammonia on the coloristic properties of wool dyed with madder and other natural dyes owing anthraquinone, naphthaquinone, flavone and tannin structures, has also been investigated [30]. Aluminum-mordanted wool was dyed using madder, cochineal, walnut husks, weld, red and white onion skins, and pomegranate shells and, the color changes ascribed to the extension of dye molecule conjugated system by one lone pair electron in ammonia solution.

Table 2: Characteristics of dyes/antimicrobials extracted from roots of the plants and their application properties

No.	Source	Botanic name	Chemical base	Shade on substrate	Substrate Applied	Properties achieved	Dye fastness [a]	Extraction method	Reference
1	Indian madder	*Rubia*	1,2,4-trihydroxyanthraquinone (Purpurin)	Red	Nylon and polyester	Good affinity for the fibers	---	Chelating with alum and precipitating with acid	[26]
2	Indian madder	*Rubia*	1,2,4-trihydroxy-anthraquinone (Purpurin), 1,3-dihydroxy-2-carboxy anthraquinone (Munjistin)	---	Nylon with mordant	Color	Purpurin *(G)* Munjistin *(P)*	Extraction with alum	[27]
3	Madder	*Rubia tinctoria*	Hydroxyanthraquinones	Red	Wool with mordant	Fading of the madder-dyed yarns	---	---	[28]
4	Madder, Cochineal, Walnut, Weld, White onion, Red onion, Pomegranate	*Rubia tinctoria, Dactylopius coccus, Juglens regia, Reseda luteola, Allium cepa, Allium cepa, Punica granatum,*	Anthraquinone, naphthaquinone, flavone and tannin	---	Wool with mordant	---	---	---	[29]
5	Ratanjot	*Arnebia nobilis* Rech.f	Naphthoquinonoid, hydroxynaphthoqui - none, isohexenylnaphthaz - arin	Pink, blue, purple	Polyester, nylon, cotton, wool, silk, acrylic	Color	Wash (*E*) Light (*P*) Rub (*E*)	Soxhlet extraction with n-hexane	[19, 31]
6	Turmeric	*Curcuma longa L.*	Natural phenols	Yellow	Wool	Color, antimicrobial	Wash *(G)* Light *(P)*	Aqueous ethanol extraction	[33]
7	Turmeric	*Curcuma longa L.*	Natural phenols	Yellow	Cotton coated with chitosan	Color, antibacterial	---	Aqueous ethanol extraction	[35]

Table 2: Continued.

No.	Source	Botanic name [a]	Chemical base	Shade on substrate	Substrate Applied	Properties achieved	Dye fastness	Extraction method	Reference
8	Aloevera, Chitosan, Curcumin	———	--- Polysaccharide Curcuminoid	———	Cotton, wool, rabbit hair	Antimicrobial	Wash *(G)*	———	[36]
9	Chinese herb	*Rhizoma coptidis*	Berberine	Brownish yellow	Wool with mordant	Deeper shades and darker colors, good antibacterial property	*(*Wash *P)*	Extraction with boiling water	[37]
10	Amur cork tree, Goldenseal coptis, Oregon grape, Barberry, Tree turmeric, Yerba mansa	*Coptis chinensis, Berberis aquifolium, Berberis vulgaris, Berberis aristata, Anemopsis californica*	Isoquinoline alkaloid	Yellow	Nylon 66	Color, antimicrobial	---	---	[38]
11	Dolu	*Rheum emodi*	Anthraquinone	Yellow, Olive green	Wool, silk with metallic, mordants	Color	Wash *(G)* Light *(M)*	---	[39]

[a] Excellent *(E)*, Very good *(VG)*, Good *(G)*, Medium *(M)*, Poor *(P)*

In a similar work, the dyeing behavior of wool fabrics with madder was studied [31]. The effect of different mordants (potassium dichromate, stannous chloride, alum, ferrous sulfate, lime, copper (I, II) sulfate, cobalt chloride, and nickel chloride) with three mordanting procedures (pre-, meta-, and post-mordanting) on color characteristics of the dyed samples was also investigated. The results showed that all color characteristics of the dyed samples were in the first quadrant of CIE L*a*b* color space. Mordanting with potassium dichromate, ferrous sulfate, and copper (I, II) sulfate showed the darker shades and lower chromaticity values while stannous chloride and alum gave the lighter shades and higher chromaticity values. The authors have reported that the wash and light fastness of the mordanted samples were higher than those of the un-mordanted one.

Ratanjot, *Arnebia nobilis* Rech.f, is the root of an herbal plant that has been widely used as a potent resource for obtaining both herbal drugs and coloring matter [20]. It contains a variety of naturally occurring chemicals, mainly a naphthoquinone derivative called alkannin (Figure 3). Alkannin has a deep red color in a greasy or oily environment and a violet color in an alkaline environment [32].

Extraction of coloring agents from ratanjot has been examined by Arora, et al., [20]. For this purpose and due to variety of plant species, they used different identification methods such as macroscopic, microscopic, chemical, and thin layer chromatography (TLC) analysis of the roots. The extraction was conducted by a soxhlet system with n-hexane as solvent and, the TLC analysis showed the presence of five naphthoquinone derivatives with red and pink colors.

In another related investigation by Arora, et al., [33], ratanjot extract has been used for dyeing of various textiles, i.e., cotton, wool, silk, nylon, polyester, and acrylic fibers and interestingly, the strong sensitivity of extracted natural dye to pH and temperature changes was observed. With gradual increasing of pH in the range of 3 to 12, continuous changes in color from red to purple and then, to dark blue was observed. This was attributed to the conversion of dye structure from quinonoid in acidic solution to benzenoid in alkaline solution. Dyeing results revealed the decrease in dye uptake with increase in pH, which was ascribed to the decomposition of dye structure in alkaline media.

Interestingly, by means of oxidation/reduction test, the reason of completely different resulting shades was addressed to the structure transformation of dye from normal state (quinonoid) to the reduced reversible state (benzenoid) on all substrates. Fastness tests results demonstrated poor light fastness whereas wash, rub and perspiration fastnesses were excellent. Finally, the findings of the study showed that *Arnebia nobilis* Rech.f can be used as a good natural dye resource for dyeing and production of variety of eco-friend textiles where light fastness is not a key among the dye selection criteria [33].

Turmeric, *Curcuma longa*, is a rhizomatous herbaceous perennial plant of the ginger family native of tropical south Asia. The dried rhizomes are ground into a deep orange-yellow powder commonly used as spice in cuisine and for dyeing of fabrics. Turmeric's active ingredient is curcumin, one of the natural phenols that are responsible for the yellow color of turmeric. Other two curcuminoids presented in turmeric are desmethoxycurcumin and bis-desmethoxycurcumin, known as curcumin I-III [34]. Chemical structures of curcuminoids are shown in Figure 4.

Turmeric has been used to impart either color or antibacterial property on textiles [35, 36]. Han and Yang [35] used curcumin as antimicrobial finish for wool against two bacteria *Staphylococcus aureus* (*S. aureus*) and *Escherichia coli* (*E. coli*) with no additional treatment through conventional dyeing process. Very low concentration of curcumin was effective in bacterial growth inhibition on wool, e.g., only 0.05% and 0.2% curcumin were able to impart inhibition rate of 70% and 95% against *E. coli*, respectively.

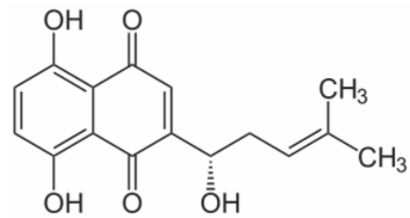


Figure 3: Chemical structure of alkannin extracted from Ratanjot (IUPAC name: 5,8-Dihydroxy-2-[(1S)-1-hydroxy-4-methylpent-3-en-1-yl]naphthalene-1,4-dione).

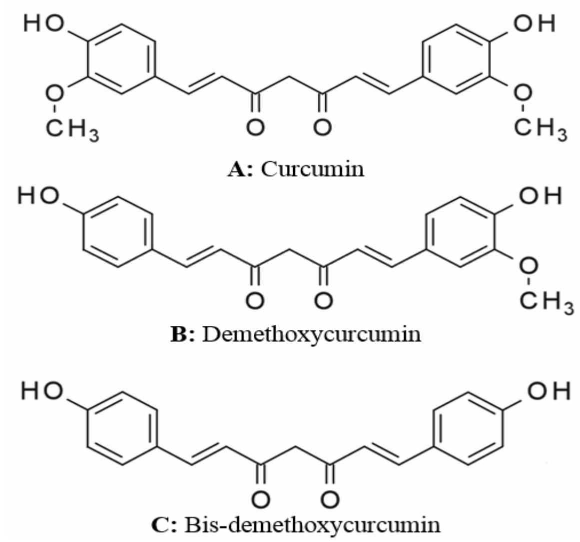


Figure 4: Chemical structure of curcuminoids extracted from turmeric (IUPAC name: **A:** (1E,6E)-1,7-bis (4-hydroxy-3-methoxyphenyl) -1,6-heptadiene-3,5-dione; **B:** (1E,6E)-1,6-Heptadiene-3,5-dione, 1-(4-hydroxy-3-methoxyphenyl) -7-(4-hydroxyphenyl); **C:** (1E,6E)-1,7-bis (4-hydroxyphenyl) hepta-1,6-diene- 3,5-dione).

An attempt has also been made to develop environmentally friendly textiles for medical application using chitosan and turmeric. Kavitha, et al., [37] have reported the results of turmeric dyed cotton yarns that has been pretreated with chitosan. Improvements in the mechanical properties such as tensile strength, flexural rigidity, and shear strength as well as natural dye uptake were achieved upon chitosan treatment with additional excellent antibacterial activity against two pathogenic bacteria, *E. coli* and *S. aureus*.

Recently, in an interesting attempt to develop an eco-friend antibacterial finish using materials from green resources, curcumin along with two other natural bioactive agents, aloevera and chitosan, have been applied lonely or in binary or in ternary combinations on cotton, wool and rabbit hair [38]. Their bactericidal properties in both pretreated and untreated substrates were examined and, it was found that antibacterial activity of substrates finished after pretreatment with selective chemicals is higher than directly finished fabrics.

Also, in a research work by Ke, et al., [39], the plant roots of a chinese herb, *Rhizoma coptidis* having natural cationic colorant called berberine, was minced and dipped in distilled water for 30 min, then boiled in distilled water to extract the brownish yellow dye, that was then used for dyeing of wool fibers with mordanting. The wool fabrics dyed under different conditions showed various yellow colors with deep shades and good antibacterial properties while washing fastness was poor.

Berberine has been similarly used as colorant and antimicrobial finish on nylon 66 fibers [40]. By using berberine, excellent bactericidal property was imparted to nylon fibers and consequently, it was introduced as practical, eco-friendly, very simple and effective finish. An interesting investigation on the coloration of protein fibers was carried out using a natural dye by Das, et al., [41]. Wool and silk fibers were dyed with extract of *Rheum emodi* using metallic mordants, manganese (Mn), aluminum (Al), iron (Fe), and consequently, various shades from yellow to olive green were obtained. Chrysophanic acid, an anthraquinone derivative, is the main coloring matter of *Rheum emodi* with the anthraquinone chemical structure (Figure 5). Interestingly, dyeing results exhibited no changes in the dye uptake of this dye in the pH range of 4-8. Another finding was the unusual higher dye up-take, dye affinity, and dyeing rate of silk than that of wool fibers.

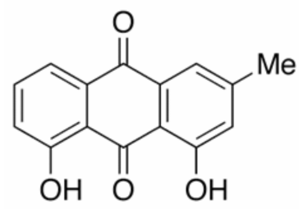


Figure 5: Chemical structure of Chrysophanic acid the main color matter of Rheum emodi (IUPAC name: 1,8-dihydroxy- 3-methyl- 9,10-anthracenedione).

4.2. Dyes/antimicrobials from seed

Seed of the plants could be a rich resource of natural dyes/antimicrobuals (Table 3). For instance, bixin, a natural dye with carboxylate groups found in annatto seed, could be considered as a natural acid dye. It was discussed that with decreasing pH, the amino groups in protein fibers are protonated, ionic interaction of bixin carboxylate anions with amino cations increases, and consequently the affinity and dye uptake increase [42]. Interestingly, the color imparting ability of mordants for both wool and silk fibers was found to be in descending order with bixin dye: ferrous sulfate > aluminum sulfate > magnesium sulfate > no salt, which was attributed to the easily and rapid salt formation of iron and aluminum with bixin compared to magnesium.

Sathianarayanan, et al., [43] have tried to impart bactericidal finish on cotton fabrics using the bio-active ingredients extracted from ban-ajwain seeds, *Thymus serpyllum*. The extract was incorporated through different techniques such as direct application, micro-encapsulation with acacia gum, cross-linking with resin, and the mixture of them. Durability of finish against the washing was very low in direct application. Wash fastness was enhanced with cross-linking and micro-encapsulation, where, some physical/mechanical properties were lost. Combined method gave the best fastness results so that the finish was durable up to 15 washing cycles.

Table 3: Characteristics of dyes/antimicrobials extracted from seed of the plants and their application properties

No.	Source *(Local name)*	Botanic name	Chemical base	Shade on substrate	Substrate Applied	Properties achieved	Dye fastness[a]	Extraction method	Reference
1	Annato *(Jorot)*	*Bixa orellana*	Carotenoids (Bixin, Norbixin)	Yellow, Orange, Brown	Wool, silk with metallic mordants	Color	Wash *(M)* Light *(P)*	---	[40]
2	Ban-ajwain	*Thymus serpyllum*	---	---	Cotton	Antibacterial	Wash *(VG)*	Soxhlet extraction with methanol	[41]

[a] Excellent *(E)*, Very good *(VG)*, Good *(G)*, Medium *(M)*, Poor *(P)*

In another study using natural dyes, the renewable environmentally benign soybean protein fibers were dyed with madder, Weld and Walnut seed husk extracts [44]. Wool fabrics were treated at similar conditions for comparison purposes. Then, the dyeing and antimicrobial properties were characterized and compared. It was found that the amount of the amino acid content of the fiber plays a dominant role on the color depth. Moreover, an unusual change in the color hue of the madder on the soybean textiles was observed when dyed fabrics were washed in an alkaline solution. Additional antimicrobial activity against the gram positive bacterium was imparted using Walnut husk and madder dyes while the effect of Weld dye was almost negligible. However, the natural dyes examined did not exhibit any significant bactericidal properties on soybean fabrics against gram negative bacterium.

4.3. Dyes/antimicrobials from leaf

In some studies, leaves of different plants have been used as the color resource [45, 46] (Table 4). For Example, eucalyptus, *Eucalyptus camaldulensis*, is a rich resource of natural polyphenols and tannins. Leaves containing 11% quercetin as the major component along with other minor components such as rutin, gallic acid, ellagic acid, etc (Figure 6) are very useful in coloration yield and durability of substrate [47].

Mongkholrattanasit, et al., [48] demonstrated that aqueous extract of eucalyptus leaves could be effectively used in exhaustion coloration process of wool and silk, where aluminum, iron, copper, and tin were used as mordants. Wool fibers had higher color strength than silk fibers and wide variety of shades were achieved on both wool and silk, i.e., yellowish brown with aluminum and copper, dark grayish brown with iron and bright yellow with tin. Protection against UV irradiation was additional property imparted for both substrates, and fabrics showed good to very good wash fastness and medium to good light and rub fastnesses.

Tea plant, *Camellia sinensis*, is the rich resource of different beneficial natural components such as antioxidants, flavanols (flavan-3-ols), flavonoids, polyphenols, and catechins. It is processed through different methods to produce variety kinds of teas, i.e., green, yellow, dark, white, oolong, and black tea. Deo and Desai [49] have investigated the dyeing of cellulosic fibers, cotton and jute, with aqueous extract of black tea where the tannins were the main coloring agents. The solution of black tea was stirred, heated, held at the boil for 30 min, allowed to rest for 15 min and then, filtered and used as black pigment for dyeing of fabrics together with ferrous sulfate heptahydrate, alum and copper sulfate pentahydrate mordants. Deep color shade on jute fabrics was produced with good to excellent wash and light fastnesses.

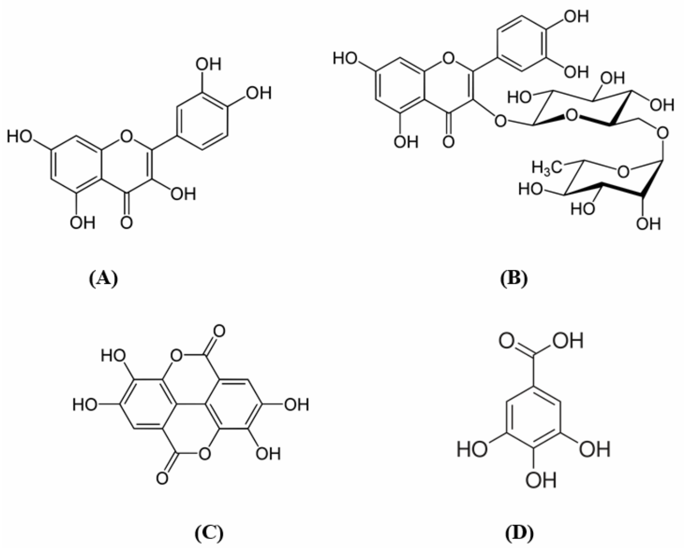


Figure 6: Chemical structures of A: quercetin, B: rutin, C: ellagic acid and D: gallic acid, extracted from eucalyptus leaf (IUPAC name A: 2-(3,4-dihydroxyphenyl)-3,5,7-trihydroxy-4H-chromen-4-one; B: 2-(3,4-dihydroxyphenyl)-5,7-dihydroxy-3-[α-L-rhamnopyranosyl-(1→6)-β-D-glucopyranosyloxy]-4H-chromen-4-one; C: 3,4,5-trihydroxybenzoic acid; D: 2,3,7,8-Tetrahydroxy-chromeno[5,4,3-cde]chromene-5,10-dione).

Table 4: Characteristics of dyes/antimicrobials extracted from leaves of the plants and their application properties.

No.	Source	Botanic name	Chemical base	Shade on substrate	Substrate Applied	Properties achieved	Dye fastness[a]	Extraction method	Reference
1	Eucalyptus	*Eucalyptus camaldulensis*	Polyphenol (Quercetin, Rutin, Ellagic Acid, Gallic Acid)	Yellow to brown, Grey	Wool, silk with metallic mordants	Color, UV protection	Wash *(G to VG)* Light *(M to G)* Rub *(M to G)*	Aqueous extraction	[45]
2	Tea	*Camellia sinensis*	Tannin	Black	Cotton, Jute with mordants	Deep shad on jute	Wash *(G to E)* Light *(G to E)*	Extracted with boiling water	[46]
3	Green tea	*Camellia sinensis*	---	---	Cotton	Antibacterial	Wash (M)	Methanolic extraction	[47]
4	Deciduous	*Silver oak, Flame of the forest, Tanner's senna, Wattle, Serviceberry*	Tannin	Yellow, Brown, Yellow, Orange, Dark grey	Wool with mordants	Color	Light *(M to G)*	Extraction with boiled water	[48]
5	Dom Sheng	*Symplocos spicata*	Flavonol	Yellowish brown	Cotton, wool, silk with metallic mordants	Color	Wash *(VG)* Light *(VG)* Rub *(VG)*	Aqueous extraction	[49]

[a] Excellent *(E)*, Very good *(VG)*, Good *(G)*, Medium *(M)*, Poor *(P)*

In another related study, with the aim of production of eco-friendly medical textiles, Syamili, et al., [50] have tried to apply green tea leaves extract onto cotton to impart bactericidal property. The effective ingredients of green tea were evaluated using various analyzing methods. The ingredients were extracted through ethanol, chelated with copper to exploit intrinsic bactericidal property of copper, and applied on cotton lonely and with citric acid as X-linking agent. Results demonstrated considerable inhibition against *S. aureus* and *E. coli* bacteria growth on finished substrates.

Similarly, the leaves of deciduous plant with high tannin content have been used as a natural dye sources in coloration of wool [51]. The coloring agent of five different deciduous plants, namely, silver oak, flame of the forest, tanner's senna, wattle and serviceberry, was extracted through boiling 250 g of the leaf material with 5 lit of water for 1 h and were used along with mordants in wool dyeing. The authors have introduced the use of plant leaves more eco-friendly and economical than using other parts, showing a moderate to good fastness.

Also, ultrasonic coloration of cotton, wool, and silk have also been performed using the extract of *Symplocos spicata* plant leaves along with Al, Fe, Cu, Cr, Sn(*II*) and Sn(*IV*) mordants [52]. In the case of cotton fibers, an additional pretreatment with tannic acid was also conducted to provide carboxylic acid groups on the fiber surface for better mordanting yield. Dye uptake was markedly improved and became almost double in ultrasonic dyeing compared to conventional exhaustion dyeing process. Yellow to brown shades were produced on fibers with high color fastness properties. Lower time, energy, and dye amount required to obtain the same results compared to the conventional dyeing process proved the superiority of ultrasonic natural dyeing method.

In another related work, the potential of red calico (Alternanthera bittzickiana (Regel) G. Nicholson) leaves gamma ray assisted extracts as natural dye source for coloration of cotton textiles was explored [53]. The results demonstrated that red calico plant leaves could be an excellent source of natural dyes for textile dyeing. Copper sulfate (1%) and tannic acid (1%) were identified as best pre- and post-mordants, respectively, to improve the color strength and color fastness properties. Gamma ray dose of 15 kGy was effective for extraction of dye and surface modification of cotton fabric. Moreover, the natural dye can be best extracted from irradiated powder of red calico leaves using alkaline medium. Optimum condition for dyeing produced good color strength was 60 ºC for 50 min using dye bath of pH 7.0 and salt concentration of 6 g/L.

In a relevant study, natural colorant from the leaves of fennel (*Foeniculum vulgare*) was valorized in the dyeing of cationized cotton fabrics [54]. The influence of the main dyeing conditions (dye bath pH, dyeing temperature, and dyeing duration) on the performances of this dyeing process was studied. A surface design with the help of Minitab 15 software was used for optimization of the dyeing process and evaluation of interaction effects of different operating parameters. The optimum conditions for dyeing process were found to be pH of 8.22, dyeing temperature of 99.83 ºC and dyeing duration of 59.38 min. It was concluded that the dye bath of cationized cotton with Croscolor DRT absorbed polyphenols compounds and reduced the Biological Oxygen Demand, COD and BOD_5 of the dyeing bath.

4.4. Dyes/antimicrobials from fruit

Fruits of the plants are another resource of natural dyes and have been extensively studied in textile coloration (Table 5). As an example, pomegranate rind extract was used for bi-functional treatment of cotton, i.e., coloring and bactericidal finish [55]. Pomegranate is the fruit of deciduous shrub or small tree, *Punica granatun*. The dye was extracted by water/ethanol solvent and showed good affinity for cotton. Good wash, light, rub, and perspiration fastnesses and excellent antibacterial property against common bacteria, *S. Aureus* and *E. coli,* were resulted. However, antibacterial property steadily decreased by successive washing cycles.

Similarly, pomegranate rind has been utilized for dyeing of wool and silk with using or without mordants [56]. The best dyeing pH was found to be 4, where iron and aluminum were resulted in grey and yellow shades on fibers, respectively. Mordanting improved dye uptake and light fastness, while, wash fastness was not markedly changed [57].

Table 5: Characteristics of dyes/antimicrobials extracted from fruits of the plants and their application properties.

No.	Source *(Used part)*	Botanic name	Chemical base	Shade on substrate	Substrate Applied	Properties achieved	Dye fastness [a]	Extraction method	Reference
1	Pomegranate *(peel)*	*Punica granatum*	Alkaloid	---	Cotton with metallic mordants	Color, antimicrobial	Wash *(G)* Light *(G)* Rub *(G)* Perspiration *(G)*	Aqueous extraction, Ethanolic extraction	[50]
2	Pomegranate *(rind)*	*Punica granatum*	Tannin, Alkaloid	Yellow, grey	Wool, silk with metallic mordants	Color	Wash *(G)* Light *(G)* Rub *(G)*	Aqueous extraction, Ethanolic extraction	[52]
3	Fruit	*Opuntia ficus-indica*	Indicaxanthin, Betanin	---	Wool with mordant	Color	Wash *(G)*	Extraction with ethanol	[53]
4	Fruit	*Opuntia ficus-indica*	Indicaxanthin	Orange-yellow	Modified acrylic fibers	Improvement of dye uptake by sonicator dyeing	Light *(M)*	Extraction with ethanol	[55]
5	Prickly pear *(fruit)*	*Opuntia Lasiacantha* Pfeiffer	Betalain	Red	Wool with mordants	Showing high bacteria inhibition activity	Light *(G)*	Extraction with water	[56]
6	Mangosteen *(hull)*	*Garcinia mangostana Linn*	Anthocyanin	Red	Cotton, Silk with metallic mordants	Color	Wash *(G)* Light *(G)* Rub *(G)*	With aqueous citric acid solution	[57]
7	Amla *(fruit)*	*Emblica officinalis G.*	Tannin	---	Cotton, Silk with natural and metallic mordants	Antibacterial	Wash *(G)*	Ethanolic extraction	[58]
8	Green chili	*Capsicum annum*	Oleoresin	Yellow	Cotton with modrants	---	Wash *(M)* Light *(G)* Rub *(G)*	Solvent extraction method	[59]
9	Banana *(peel)*	*Musa cavendish*	Flavonoid	---	Cotton with metallic mordants	Color, antibacterial, UV protection	---	Alkaline extraction	[61]
10	Orange *(peel)*	*Citrus sinensis*	---	Yellow	Wool with mordants	Color, UV protection	Wash *(G)* Rub *(G)* Light *(M)*	Extraction with water	[62]

[a] Excellent *(E)*, Very good *(VG)*, Good *(G)*, Medium *(M)*, Poor *(P)*

Also, Guesmi, et al., [58] have extracted indicaxanthin natural colorant from *Opuntia ficus-indica* fruit and used it for dyeing of textiles. This plant contains mainly two dyes, indicaxanthin and betanin (Figure 7).

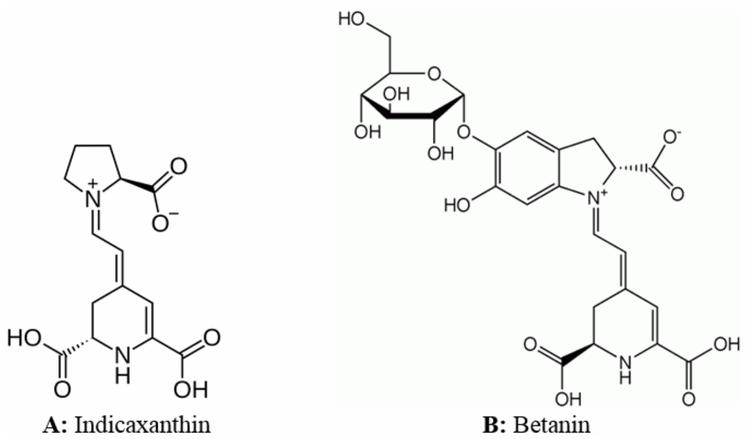


Figure 7: Chemical structures of indicaxanthin and betanin extracted from *Opuntia ficus-indica* fruit (IUPAC name **A:** 4-[2-(2-Carboxy-pyrrolidin-1-yl)-vinyl]-2,3-dihydro-pyridine-2,6-dicarboxylic acid; **B:** 4-(2-(2-carboxy-5-(beta-D-glucopyranosyloxy)-2,3-dihydro-6-hydroxy-1H-indol-1-yl)ethenyl)-2,3-dihydro-(S-(R*,R*))-2,6-pyridinedicarboxylic acid).

Extraction method was the mixing of 50 g of juice from cactus pears with 100 ml of 80% aqueous ethanol as solvent. Extracted colorant was used for dyeing of wool fibers together with different mordants and the results showed good washing fastness, where aluminum potassium sulfate ($KAl(SO_4)_2$), manganese sulfate ($MnSO_4$) and cobalt sulfate ($CoSO_4$) mordants gave the best results. Also, it was suggested that at different pH values, the betanin molecule may exist in cationized or monoanion forms [59].In a similar work, the parameters for extraction of natural dye from Chinese gall were optimized using a fractional factorial experimental design using Taguchi's orthogonal array and the optimum conditions of 90 ºC, 1:30 liquor ratio and two extraction cycles each 60 min long were identified [60]. Then, the extracts obtained at optimum conditions were used in wool fabrics dyeing. Color characteristics analyze revealed pre-mordanting as the best method for mordant usage. Highest color change, good color fastness against wash, crocking and perspiration, as well as effective bactericidal activity against typical Gram positive and Gram negative microorganisms such as S. aureus and E. coli were achieved using dye liquor concentration of 200%, mordant concentration of 2.5%, pH value of 8 at 98 ºC. Based on the results obtained and from environmentally friendly, economical, sustainable process standpoint, it was inferred that the natural dye from Chinese gall can be applied as commercial natural dyestuff with high market potential.

In a related work, indicaxanthin has been used in conventional and ultrasonic dyeing of modified acrylic fibers [61]. Remarkable improvement in dye uptake and fair to good fastness properties have been reported in sonicator dyeing of fibers with orange-yellow natural dye.

In this regard, Ali and El-Mohamedy [62] have made an attempt to extract the colorant of red prickly pear plant, *Opuntia lasiacantha* Pfeiffer. The extracted red dye, betalain (Figure 8), along with ferrous sulfate, copper sulfate, potassium dichromate and tannic acid mordants, was used in wool dyeing. The fabrics dyed with betalain showed high bacteria inhibition activity and good fastness properties.

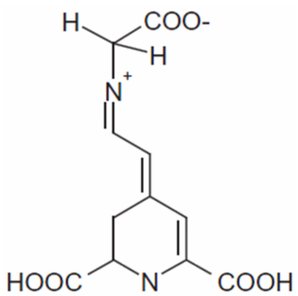
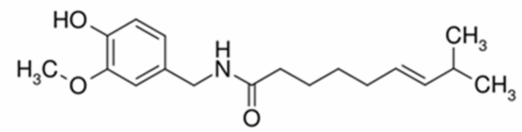


Figure 8: Betalain pigment structure extracted from *Opuntia lasiacantha* Pfeiffer fruits.

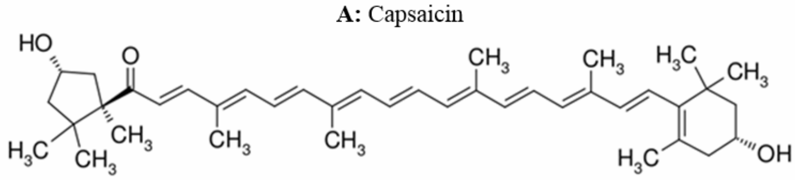
Fruit shells of tropical evergreen tree, *Garcinia mangostana Linn*, contain various and useful natural components such as mangosteen that is a red pigment. Chairat, et al., [63] have extracted mangosteen using an aqueous acetic acid solution. The extract solution with dark red color was used in coloration of silk and cotton with Al, Fe, Ca, Zn, and a natural mordant. Generally, color depth, wash and light fastness of dyed fibers were considerably improved through mordants incorporation and the best results were obtained in post-mordanting with Fe and Ca.

Recently, *Emlica officialis G.* fruit, a rich resource of natural tannins, has been used as natural mordants. It contains mainly two hydrolysable tannins which give gallic acid and ellagic acid upon hydrolysis. Prabhu, et al., [64] have used the ethanol isolated tannins, individually or in mixture with copper sulfate, for enhancement of dyeing yield and color fastness properties in natural coloration of cotton and silk. Four natural dyes used were turmeric *(Curcuma longa L.)*, pomegranate rind *(Punica granatum L.)*, henna *(Lawsonia inermis L.)*, and Indian madder *(Rubia cordifolia L.)*, with which different shades were obtained with good color fastness and color strength. Antibacterial test was also carried out to evaluate the bactericidal property of *Emlica officialis G.* on cotton and silk and, it was found that natural mordant imparted good antibacterial activity which was stable up to 20 wash cycles.

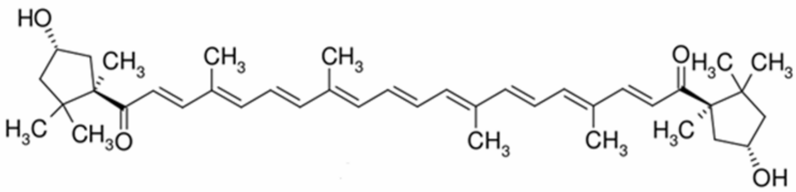
In a similar work, Kulkarni, et al., [65] have tried to produce different shades of yellow color using the colorant extracted from chili skin. Oleoresin is an oil soluble extract from the fruits of chili, *Capsicum Annum Linn*, and is primarily used as a coloring and/or flavoring agent in food products. It is composed of capsaicin, the main flavoring compound giving pungency in higher concentrations, and capsanthin and capsorubin [66] (Figure 9). The yellow pigment of *Capsicum annum*, that mainly contains oleoresin, was extracted and used with mordants for coloration of cotton. Good light and rub fastness were reported for the fabrics dyed with the green chili extract [65].



A: Capsaicin



B: Capsanthin



C: Capsorubin

Figure 9: Chemical structure of oleoresin ingredients; Capsaicin, Capsanthin and Capsorubin (IUPAC name: **A:** (E)-8-Methyl-non-6-enoic acid 4-hydroxy-3-methoxy-benzylamide **B:** (2E,4E,6E,8E,10E,12E,14E,16E,18E)-19-((R)-4-Hydroxy-2,6,6-trimethyl-cyclohex-1-enyl)-1-((1R,4S)-4-hydroxy-1,2,2-trimethylcyclopentyl)-4,8,13,17-tetramethyl-nonadeca-2,4,6,8,10,12,14,16,18-nonaen-1-one **C:** (2E,4E,6E,8E,10E,12E,14E,16E,18E)-1,20-Bis-((1R,4S)-4-hydroxy-1,2,2-trimethyl-cyclopentyl)-4,8,13,17-tetramethyl-icosa-2,4,6,8,10,12,14,16,18-nonaene-1,20-dione.

Also, natural colorant extracted from banana peel, *Musa cavendish*, through the alkaline extraction was applied with iron mordant in coloration of cotton [67]. In addition to good color imparted, dyed cotton exhibited two additional properties; i.e., excellent antibacterial activity and high UV protection property.

In a most recent published paper, orange peel, as a new natural dyestuff resource with strong ultraviolet absorbance was used to dye wool fibers [68]. All the dyed wool fabrics demonstrated good color fastness to washing with soap, and to rubbing and acceptable color fastness to light. The authors have reported that the abundantly available agricultural by-product, orange peel, has great potential to be utilized as a natural textile dyestuff that could also impart the textiles remarkable UV-protection property.

In another research work exploiting high energy of gamma rays, impacts of different absorbed doses (20, 25, 30, 35 and 40 kGy) of gamma radiations on both natural dye powder and silk fabric were investigated [69]. Natural colorants from pomegranate (*Punica granatum L.*) rind powder was extracted, characterized and utilized in dyeing of silk fabric. Aqueous, alkali and alcohol solubilized extracts were obtained from un-irradiated and irradiated pomegranate rind powders. Aluminum, copper, chromium and tannic acid (2–10% each) were used as mordants in dyeing. It was found that treatment of pomegranate peel colorant and silk fabric with gamma irradiation has greatly improved the extraction and dyeing process and required less material to liquor ratio, dyeing time, dyeing temperature and salt concentration. Moreover, color strength and fastness properties of silk fabrics dyed with irradiated pomegranate peel were noticeably enhanced through gamma ray treatments. The most effective dose for colorant extraction from pomegranate peel, surface alteration of silk fabric and enhancing silk fabric dye uptake ability was 40 kGy. In general, it was concluded that pomegranate (*Punica granatum L.*) peel is a suitable source of colorant for textile industry.

Relevantly, an ecological approach has been presented by valorization of outer green shell of almond fruit as a novel natural dye using chemical solvent free extraction, small amount of metallic mordant, alternative natural mordants, auxiliary free dyeing and washing [70]. The use of a smaller amount of metallic mordant (0.2 g/L - 1 %) and less plant (1 g plant/3 g fabric) than in most current natural dye applications is a major advantage of dyeing with almond shell extract. The authors reported that the use of biomordants is very efficient as an alternative method that may contribute to reduce the reliance on toxic metal salt mordants.

4.5. Dyes/antimicrobials from bark

Barks of various plants as natural colorant resources have been studied in some researches (Table 6). For instance, the colorant extracted from the tree bark has been used by Vinod, et al., [71] for dyeing of textile fibers. In this work, pigment from the bark of *Macaranga peltata* was extracted by methanol and ellagic acid was identified as the main coloring agent using spectral techniques. This yellow pigment along with tannic acid and potash alum $[Al(NH_4)(SO_4)_2.12H_2O]$ mordants were applied in silk dyeing, and good fastness properties were reported as the result.

Another example could be the application of barberry plant, Berberis aristata DC, that its bark has been used as natural colorant resource in textile dyeing [72]. Silk was dyed with aqueous extract of barberry bark and post-mordanted with combination of various mordants. According to the results, several conclusions were drawn: I) optimal dyeing condition for silk using this natural dye was simultaneous mordanting because of the highest dye uptake during this method; II) alum must be used together with other mordants owing to its synergistic role in dye uptake, and III) a variety of shades could be produced on silk using combination of mordants.

Similar attempt has been made to use *Sticta coronata* lichen as an abundant resource of natural dye in New Zealand in ultrasonic assisted coloration of silk [73]. During this dyeing process, a natural mordant, Catechu, *Acacia catechu*, along with alum were used, while dye uptake was improved notably assisting ultrasound energy. The natural dye had good affinity towards silk and imparted brilliant lilac color had good fastness properties.

Table 6: Characteristics of dyes/antimicrobials extracted from bark of the plants and their application properties

No.	Source *(Used part)*	Botanic name	Chemical base	Shade on substrate	Substrate Applied	Properties achieved	Dye fastness[a]	Extraction method	Reference
1	Chandada *(bark)*	*Macaranga Peltata*	---	Yellow, Red	Silk	Color	---	Soxhlet extraction with methanol	[63]
2	Barberry *(bark)*	*Berberis aristata DC.*	---	Wide range of shades	Silk with metallic mordants	Color	Wash *(P)* Light *(VG)* Rub *(VG)*	Aqueous extraction	[64]
3	---	*Sticta coronate lichen*	---	Lilac (pale purple)	Silk with metallic, natural mordants	Color	---	Ultrasonic extraction with aqueous acetone	[65]
4	Jackfruit *(wood)*	*Artocarpus heterophyllus Lam*	Flavanol (Hydroxy-flavone) (Morol)	Yellow to golden brown	Cotton, jute with metallic, natural mordants	Color	Wash *(VG)* Light *(M)* Rub *(G)*	Aqueous extraction	[66]
5	Jackfruit *(wood)* Manjistha/Madder *(root)* Red sandalwood Babool *(Bark, wood)*	*Artocarpus heterophyllus L., Rubia cordifolia, Pterocarpus santalinius, Acacia arabica*	Morol Putpurin, Manjistin, Pupuroxanthin, Pseudo-purpurin, Nordamncanthal, Santalin A, Santalin B, Deoxyxantalin, Flavonoid, Cretonoid, Polyphenol	Variety of shades	Jute with natural, metallic mordants	Color	Wash *(VG)* Light *(M)* Rub *(VG)*	Aqueous extraction	[67]

[a] Excellent *(E)*, Very good *(VG)*, Good *(G)*, Medium *(M)*, Poor *(P)*

Recently, Jackfruit wood, *Artocarpus heterophyllus* Lam, extract has been used in natural dyeing of cellulosic fibers with myrobolan bio-mordant (Figure 10), aluminum , iron, tin and ethylene di-amine tetra-acetic acid (EDTA) as mordants by Samanta , et al., [74]. The main coloring component of jackfruit is morol, a typical flavanol that imparted yellow to golden brown shade with various mordants on cotton and jute fibers. Good color fastness properties were reported for both dyed fibers.

Similarly, binary mixtures of jackfruit extract and other natural dyes including manjistha, red sandal wood, marigold, sappan wood, and babool were used in coloration of jute fibers with myrobolan and aluminum mordants [75]. The compatibility of mixture of the dyes was also examined. After dyeing, additional treatment with various chemicals was performed to enhance wash and light fastness.

4.6. Dyes/antimicrobials from flower

Flowers of various plants have been considered as the natural dyes/antimicrobials resource (Table 7). For example, the dyeing potential of pigments from marigold flowers, *Tagetes patula* L., flavonoids has been evaluated by Guinot, et al., [76]. Highest extraction efficiency was obtained using aqueous ethanol solvent and the extracts were used with mordants for dyeing of wool. According to the results, marigold flower was introduced as a potential dyeing material.

Similarly, pretreated silk fibers were eco-friendly colored with flower extract [77]. A bright reddish brown pigment, containing quercetin was used for dyeing of fibers after extraction by soaking the dried and ground flower sample, *Delonix regia,* in distilled water.

Catechu, *Acacia catechu*, extract with light brown pigment that contains mainly tannin, was also used in wool coloration [78]. Different shades were obtained on wool fabrics and the fastness properties were sufficiently good for practical and large scale dyeing.

Dyeing potential of flowers of ornamental flowering tree, *Spathodea campanulata*, was evaluated by Kumaresan, et al., [79]. Extraction of colorant was performed separately both in boiling water and ethanol. Cotton was dyed individually as well as in binary combination of natural/metallic mordants. The extraction with ethanol could produce brown shades. Generally, in almost all mordant-dye-fiber systems, color fastness was quite higher than wash, rub, and perspiration fastnesses. It was concluded that, this natural dye can be used as a green source in coloration of textiles.

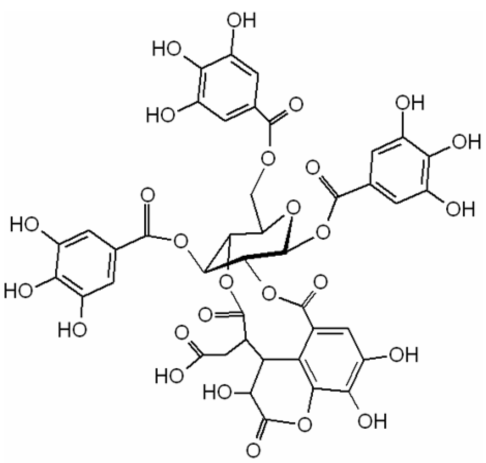


Figure 10: Chemical structure of chebulinic acid, the main component of myrobolan natural mordant.

Samanta, et al., [80] have used aqueous extract of flower petals of tesu flowering tree, *buteamonosperma frondosa*, in coloration of jute fibers in alkaline medium to obtain various shades like creamish orange, yellowish orange, ochre brown, and reddish brown. The main coloring component in the yellowish red/orange extract was butein (Figure 11). Good color fastness and color yield was reported for optimized dyeing condition (dye-fiber-mordant system) and further improvements in light fastness were obtained by applying proper post-chemical treatment.

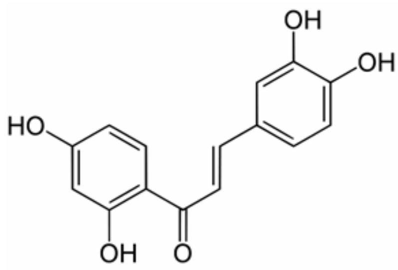


Figure 11: Chemical structure of butein extracted from flower petals of tesu flowering tree (IUPAC name: (E)-1-(2,4-dihydroxyphenyl)-3-(3,4-dihydroxyphenyl)prop-2-en-1-one).

The antimicrobial activity of five natural dyes, *Acacia catechu, Kerria lacca, Quercus infectoria, Rubia cordifolia* and *Rumex maritimus*, on wool fibers has been reported by Singh, et al., [81]. The anti-microbial activity of *Catechu* has also been proven by Ibrahim khan, et al., [82]. Moreover, tannin-rich extract of oak has been used as an antibacterial agent for textiles by Gupta and Laha [83]. In this work, the extract of *Quercus infectoria* plant in combination with alum, copper and ferrous mordants have been used in natural antibacterial treatment of cotton fabrics. Good antibacterial properties were imparted to cotton against both Gram-positive and Gram-negative bacteria.

The methanol extracts of *Ricinus communis*, *Senna auriculata* and *Euphorbia hirta* were tested for their antimicrobial efficacy on various denim fabrics directly by using dip method [84]. Results showed that the fabrics had good antibacterial activity.

Table 7: Characteristics of dyes/antimicrobials extracted from flower of the plants and their application properties

No.	Source *(Local name)*	Botanic name	Chemical base	Shade on substrate	Substrate Applied	Properties achieved	Dye fastness [a]	Extraction method	Reference
1	Marigold flower	*Tagetes patula* L.	Flavonoid	---	Woven wool with mordant	Development of new natural Coloring matters	---	Extraction with water-ethanol mixture	[68]
2	Assamese *(Radha sura)*	*Delonix regia*	Quercetin	Bright reddish brown	Silk with mordant	Eco-friendly textile pretreatment that does not utilize metal mordanting	Light *(M)*	Extraction with water	[69]
3	Catechu *(Khoyar)*	*Acacia catechu* W.	Tannin	Light brown	Wool with mordant	Production of different colors	Light *(G)*	---	[70]
4	African tulip tree	*Spathodea campanulata*	---	Various shades	Cotton with metallic natural mordants	Color	Wash *(E)* Light *(M)* Rub *(E)* Perspiration *(E-G)*	Aqueous extraction	[71]
5	Tesu	*Buteamonosperma frondosa*	Butein	Yellow, Orange, Brown	Jute with metallic, natural mordants	Color	Wash *(G)* Light *(G)* Rub *(G)*	Aqueous extraction	[72]
6	Plants *(Khoyar,---, Mese mazısı, Majathi, ---)*	*Acacia catechu, Kerria lacca, Quercus infectoria, Rubia cordifolia, Rumex maritimus*	---	---	Wool	Antimicrobial	---	---	[73]
7	Oak *(Mese mazısı)*	*Quercus infectoria*	---	---	Cotton with mordants	Antimicrobial	---	---	[75]
8	Herb Leaves, Stem and Flower	*Ricinus communis, Senna auriculata, Euphorbia hirta*	---	---	Denim Fabric	Antibacterial	---	Methanol extraction	[76]
9	Herb, Sappanwood	---	---	---	Wool with mordant	anti-ultraviolet effect	Rub *(G)*	Dissolution and extraction	[77]
10	Saffron	*Crocus sativus L.*	Anthocyanidin	Yellow to brown	Wool with metallic mordants	Color	Wash *(G)* Light *(G)*	Aqueous extraction	[78]

[a] Excellent *(E)*, Very good *(VG)*, Good *(G)*, Medium *(M)*, Poor *(P)*

The natural colorant from sappan was also used for coloration of wool fibers after treatment with the protease and transglutaminase [84]. The pigment of sappan was extracted by dissolving the herb in distilled water, allowed to boil in a beaker that had been kept over water bath for quick extraction. Then, it was used for dyeing of wool fibers with ferrous sulfate, potassium alum, and tannic acid mordants. Good anti-ultraviolet effect was imparted to the fibers while modification with protease led to some decrease in wet rub fastness. Transglutaminase had almost no influence on rub fastness, while the treated fibers showed good dry rub fastness.

In a related work, Mortazavi, et al., [86] introduced saffron, *Crocus sativus L.*, as a potent natural dye resource. They used saffron flower petals and sepals extract, a rich resource of anthocyanidin natural dye, in wool dyeing. Different metallic mordants such as tin, copper, aluminum, iron, and chromium together with oxalic and tartaric acid were used to obtain a variety of shades as well as to enhance fastness properties of natural dyed wool. The color of fresh petals and sepals was light purple, but the shade range obtained on wool varied from yellow to brown with mordants. Wash and light fastnesses of wool fibers were excellent with iron and chromium, moderate with copper and aluminum, and poor with tin mordants.

4.7. Dyes/antimicrobials from other parts of plants

Natural extracts of other parts of plants have also important dyeing/antimicrobial potential and their application on textile has been reported (Table 8).

For instance, ultrasonic assisting extraction of natural dye from *Sargentodoxa cuneata* and subsequent application on wool fabric was studied by Xinsheng, et al., [87]. The authors have reported deeper color of fabrics dyed with ultrasonically extracted dye. In a similar work, alum mordanted wool was dyed in 50% *Reseda luteola* L. (weld), 20% *Rhamnus petiolaris* Boiss (buckthorn) and 50% *Datisca cannabina* L. (bastard hemp) dye-baths [88-89]. Extraction of yellow pigments containing mainly flavonoid and anthraquinone was performed using hydrogen chloride/methanol/water mixture. The authors suggested reverse-phase high-performance liquid chromatography as an efficient characterization technique for analyzing and identification of natural dyes in historical textiles.

Dahlia variabilis plant has been similarly used for dyeing of wool [90]. The flavonoids – anthocyanins rich colorant was extracted by ultrasound and was used for dyeing along with alum and milk of tartar. Good fastness properties along with the intrinsic potential of this dye to replace phenolphthalein as a pH indicator were reported.

On the other hand, selected natural dyes from madder (red), weld (yellow) and woad (blue)- extracts of *Rubia tinctoria*, *Reseda luteola L.* and *Isatis tinctoria*, respectively- were applied on cotton using mordants [91]. It was concluded that using UV absorbers or antioxidants improves the light fastness of dyed fabrics.

In a related research, Raja and Thilaghavati [92] have used the inherent antibacterial characteristic of some natural dyes in bi-functional environmentally friendly treatment of wool. Four natural dyes, with in vitro antimicrobial activity against both gram-positive and gram-negative bacteria, were chosen. The effect of mordant and protease enzyme on the bactericidal properties of natural dyes was examined. Results demonstrated higher dye uptake and antibacterial property in enzyme treated wool which was ascribed to enhanced dye affinity and diffusion due to hydrolysis of outer layer of wool with enzyme. Moreover, although the color fastness properties of dyed wool were increased upon mordant treatment, none of mordant treated natural dyed fibers showed antibacterial activity. Such behavior was attributed to the blocking of bio-active ingredients in natural dye through possible complex formation of metal ions with those components [92].

Table 8: Characteristics of dyes/antibacterial extracted from other parts of plants and their application properties.

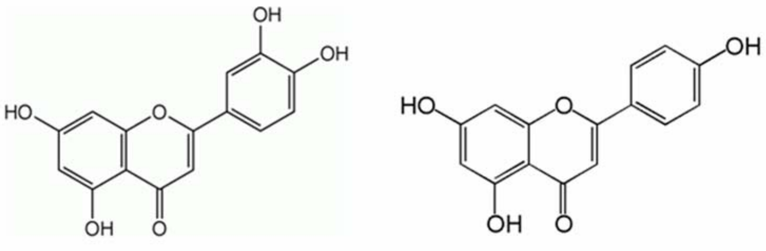
No.	Source *(Used part)*	Botanic name	Chemical base	Shade on substrate	Substrate Applied	Properties achieved	Dye fastness[a]	Extraction method	Reference
1	Vegetable	*Sargentodoxa cuneata*	---	---	Wool with mordant	Color	---	Ultrasonic extraction technique	[79]
2	Weld, Buckthorn, Bastard hemp	*Rhamnus petiolaris* Boiss, *Datisca cannabina* L., *Reseda luteola* L.	Flavonoid and Anthraquinone	Yellow	Wool with mordant	Color	---	Extraction of with hydrogen chloride/ methanol/ water mixture	[80]
3	Plant	*Dahlia variabilis*	Flavonoids - Anthocyanins	---	Wool with mordant	Good potential to replace phenolphthalein as an indicator	Light *(G)*	Ultra sound assisted extraction	[82]
4	Madder *(seed)*, weld and woad	*Reseda luteola* L., *Isatis tinctoria, Rubia tinctoria*	---	Red Yellow Blue	Cotton with mordant	Color	Light *(P)*	---	[83]
5	Silver oak Flame of forest Tanner's senna Wattle	*Grevillea robusta, Spathodea campanulata, Cassia auriculata, Acacia decurrens*	Tannin --- --- Tannin	---	Wool with enzyme and mordants	Color, antimicrobial	---	Aqueous extraction	[84]
6	Yarrow *(flower)*	*Achillea millefolium*	Flavonoids, luteolin and apigenin	---	Wool with metallic mordants	Color	Wash *(G)* Light *(G)*	Aqueous extraction	[85]

[a] Excellent *(E)*, Very good *(VG)*, Good *(G)*, Medium *(M)*, Poor *(P)*

Kiumarsi, et al., [93] have recently optimized the dyeing process of wool by yarrow, *Achillea millefolium*, using Taguchi statistical method. *Achillea millefolium* is a pharmaceutical plant known as a powerful healing herb containing flavoniods such as luteolin and apigenin (Figure 12).

The aqueous extract was used for dyeing of wool with aluminum, tin, and copper mordants. Interesting variety of shades from pale yellow to dark green was obtained with very good wash and light fastness for all dyed samples. The process was claimed to be economical due to saving time and energy, and consequently could be commercialized.

In another interesting study for identification and development of new natural dye sources, wood wastes generated from processing of heartwood of some plants were used in dyeing of cotton fabric, wood viscose and bamboo viscose textiles [94].



A: Luteolin **B:** Apigenin

Figure 12: Chemical structure of luteolin and apigenin extracted from *Achillea millefolium* plant (IUPAC name A: 5,7-Dihydroxy-2-(4-hydroxyphenyl)-4H-1-benzopyran-4-one; B: 2-(3,4-Dihydroxyphenyl)- 5,7-dihydroxy-4-chromenone.

Accordingly, natural dyes were extracted from padouk and movingui sawdust using alkaline and aqueous media and used in mordant free dyeing process. Yellow and red color hue was obtained with movingui and padouk extracts, respectively. Moreover, dyeing with alkaline extracts gave better results than the aqueous extracts. Generally, cotton fabrics presented better dyeing ability than bamboo and wood viscose textiles. Considering the fact that the chemical composition was more or less similar for the whole samples and degree of polymerization for man-made fibers was lower, the observed differences were attributed to lower crystallinity of cotton fabrics resulted from former textile process. Overall, based on the results, it was inferred that padouk and movingui extracts could be used to dye cellulosic textile materials as an attractive alternative for synthetic dyes regularly used in the industry.

5. Conclusions

As a result of the increasing awareness of the high risk impact associated with the application of synthetic colorants used for textiles, the application of natural or green extracts of the plants as colorants or antimicrobials has been recently increased considerably. These dyes are extracted from different parts of the plants such as bark, leaf, root, seed, fruit, and flower that contain coloring materials like tannin, flavonoids, quinonoids, etc.

There have been undeniable developments in extraction techniques of natural dyes/antimicrobials such as the invention of ultrasonic, enzyme assisted ultrasonic, and microwave assisted extraction methods where they prove high extraction efficiency over the conventional methods. Review of the relevant papers revealed that most of natural dyes have inherently antimicrobial properties and could consequently possess high medicinal activity. While these dyes from natural sources are eco-friendlier than synthetic ones, the level of mordants used for better fastness purpose is normally higher than the official permitted concentration and consequently, possess environmental risks. Metallic mordants are being progressively replaced either by cleaner/greener alternatives such as natural tannins from plants as bio-mordants or by cleaner technologies such as plasma, microwave, ultrasonic, etc. with higher yield over conventional methods.

The results of this literature review showed that the non-standardized time-consuming extraction methods, non-reproducibility of colors on textiles using natural dyes, and poor fastness properties are the main problems ahead of the dyers and finishers to use these natural products in textile industry and consequently, are the fields that must be explored by the researchers.

Real effort has been put to include all the relevant referred publications on the application of natural dyes/antimicrobials on green treatments of textiles in this review. However, the limitation of our resources and the sheer number of publications in this field may have prevented the comprehensiveness of this report. Our sincere apologies are extended to any and all authors whose works are not included in this review.

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6. References

1. P. Guinot, A. Roge, A. Gargadennec, M. Garcia, D. Dupont, E. Lecoeur, L. Candelier, C. Andary, Dyeing plants screening: an approach to combine past heritage and present development, *Color Technol.,* 122(2006), 93-101.
2. H. R. Rajabi, O. Khani, M. Shamsipur, V. Vatanpour, High-performance pure and Fe^{3+}-ion doped ZnS quantum dots as green nanophotocatalysts for the removal of malachite green under UV-light irradiation, *J. Hazard. Mater.*, 250-251(2013), 370-378.
3. M. B. Kasiri, A. R. Khataee, Photooxidative decolorization of two organic dyes with different chemical structures by UV/H_2O_2 process: Experimental design, *Desalination*, 270(2011), 151-159.
4. A. K. Samanta, A. Konar, Dyeing of textiles with natural dyes, in: E.P.A. Kumbasar, (Ed.), Natural Dyes, InTech, Croatia, 3rd chapter, 2011.
5. M. Shamsipur, H. R. Rajabi, O. Khani, Pure and Fe^{3+}-doped ZnS quantum dots as novel and efficient nanophotocatalysts: Synthesis, characterization and use for decolorization of Victoria blue R, *Mater. Sci. Semicond. Process,* 16(2013), 1154-1161.
6. M. B. Kasiri, A. Nemati Babaylou, H. Zandkarimi, Photo-Oxidative Stability of a Series of Red Acrylic Paints, *Prog. Color Colorants Coat.* 7(2014), 177-185.
7. M. Sadeghi-Kiakhani, S. Safapour, Eco-friendly dyeing of treated wool fabrics with reactive dyes using chitosanpoly(propylene imine)dendreimer hybrid, Clean Techn. Environ. Policy, DOI 10.1007/s10098-014-0855-z.
8. T. L. Dawson, Biosynthesis and synthesis of natural colours, *Color Technol.,* 125(2009), 61-73.
9. M. O. Bulut, E. Akar, Ecological dyeing with some plant pulps on woolen yarn and cationized cotton fabric, *J. Clean. Prod.,* 32(2012), 1-9.
10. V. Sivakumar, J. Vijaeeswarri, J. L. Anna, Effective natural dye extraction from different plant materials using ultrasound, *Ind. Crops Prod.,* 33(2011), 116-122.
11. M. B. Kasiri, S. Safapour, Natural Dyes and Antimicrobials for Textiles, in: E. Lichtfouse, J. Schwarzbauer, D. Robert, (Eds.), Green Materials for Energy, Products and Depollution, 6th Chapter, Springer Science+Business Media, Dordrecht, 2013.
12. D. J. Hill, Is there a future for natural dyes? *Rev Prog Coloration,* 27(1997), 18-25.
13. M. B. Kasiri, S. Safapour, Natural dyes and antimicrobials for green treatment of textiles, Environ. *Chem. Lett.,* DOI 10.1007/s10311-013-0426-2.
14. T. L. Dawson, It must be green: meeting society’s environmental concerns, *Color Technol.,* 124(2008), 67-78.
15. A. K. Prusty, T. Das, A. Nayak, N. B. Das, Colourimetric analysis and antimicrobial study of natural dyes and dyed silk, *J. Clean. Prod.,* 18(2010), 1750-1756.
16. A. Shams-Nateri, Reusing wastewater of madder natural dye for wool dyeing, *J. Clean. Prod.,* 19(2011), 775-781.
17. H. Ahmadi, S. Safapour, A review on the UV-Protective finish of natural textiles using UV-absorbers, *J. Stud. Color. World,* 2014, In Press.
18. K. Farizadeh, M. Montazer, M. E. Yazdanshenas, A. Rashidi, R. Mohammad Ali Malek, Extraction, identification and sorption studies of dyes from madder on wool, *J. Appl. Polym. Sci.,* 113(2009), 3799-3808.
19. I. K. Hong, H. Jeon, S. B. Lee, Extraction of natural dye from Gardenia and chromaticity analysis according to chi parameter, *J. Ind. Eng. Chem.* (2014), http://dx.doi.org/10.1016/j.jiec.2014.10.004
20. A. Arora, M. L. Gulrajani, D. Gupta, Identification and characterization of Ratanjot (*Arnebia nobilis* Reichb.f.), *Nat. Prod. Radiance,* 8(2009), 142-145.
21. N. S. E. Ahmed, R. M. El-Shishtawy, The use of new technologies in coloration of textile fibers, *J. Mater. Sci.,* 45(2010), 1143-1153.
22. M. M. Kamel, R. M. El-Shishtawy, B.M. Yussef, H. Mashaly, Ultrasonic assisted dyeing III. Dyeing of wool with lac as a natural dye, *Dyes Pigments.,* 65(2005), 103-110.
23. H. C. Tiwari, P. Singh, P. K. Mishra, P.

Srivastava, Evaluation of various techniques for extraction of natural colorants from pomegranate rind– Ultrasound and enzyme assisted extraction, Indian *J. Fibre Text. Res.,* 35(2010), 272-276.

24. M. Dabiri, S. Salimi, A. Ghassempour, A. Rassouli, M. Talebi, Optimization of microwave-assisted extraction for alizarin and purpurin in Rubiaceae plants and its comparison with conventional extraction methods, *J. Sep. Sci.,* 28(2005), 387-396.
25. K. Sinha, P. D. Saha, S. Datta, Extraction of natural dye from petals of Flame of forest (*Butea monosperma*) flower: Process optimization using response surface methodology (RSM), *Dyes Pigments*., 94(2012), 212-216.
26. F. Nourmohammadian, M. Davoudzadeh Gholami, An Investigation of the Dyeability of Acrylic Fiber Via Microwave Irradiation, *Prog. Color Colorants Coat.,* 1(2008), 57-63.
27. D. Gupta, S. Kumari, M. Gulrajani, Dyeing studies with hydroxyanthraquinones extracted from Indian madder. Part 1: Dyeing of nylon with purpurin, *Color Technol.,* 117(2001), 328-332.
28. D. Gupta, M. Gulrajani, S. Kumari, Light fastness of naturally occurring anthraquinone dyes on nylon, *Color Technol.,* 120(2004), 205-212.
29. M. Montazer, M. Parvinzadeh, Effect of ammonia on madder-dyed natural protein fiber, *J. Appl. Polym. Sci.,* 93(2004), 2704-2710.
30. M. Montazer, M. Parvinzadeh, A. Kiumarsi, Colorimetric properties of wool dyed with natural dyes after treatment with ammonia, *Color Technol.,* 120(2004), 161-166.
31. M. Feiz, H. Norouzi, Dyeing studies of wool fibers with madder (*Rubia Tinctorum*) and effect of different mordants and mordanting procedures on color characteristics of dyed samples, *Fiber. Polym.,* 15(2014), 2504-2514.
32. V. P. Papageorgiou, A. N. Assimopoulou, E. A. Couladouros, D. Hepworth, K. C. Nicolaou, The chemistry and biology of alkannin, shikonin, and related naphthazarin natural products, *Angew. Chem. Int. Ed.,* 38(3)(1999), 270-300.
33. A. Arora, D. Gupta, D. Rastogi, M. L. Gulrajanii, Kinetics and thermodynamics of dye extracted from *Arnebia nobilis* Rech.f on wool, *Indian J. Fibre Text. Res.,* 37(2012), 178-182.
34. E. W. C. Chan, Y. Lim, S. Wong, K. Lim, S. Tan, F. Lianto, Effects of different drying methods on the antioxidant properties of leaves and tea of ginger species, *Food Chem.,* 113(1)(2009), 166-172.
35. S. Han, Y. Yang, Antimicrobial activity of wool fabric treated with curcumin, *Dyes Pigments.,* 64(2005), 157-161.
36. A. V. Popoola, Comparative fastness assessment performance of cellulosic fibers dyed using natural colorants, *J. Appl. Polym. Sci.,* 77(2000), 752-755.
37. T. Kavitha, R. Padmashwini, A. Swarna, V.R. Giri Dev, R. Neelakandan, Effect of chitosan treatment on the properties of turmeric dyed cotton yarn, *Indian J. Fibre Text. Res.,* 32(2007), 53-56.
38. L. Ammayappan, J. J. Moses, Study of antimicrobial activity of aloevera, chitosan, and curcumin on cotton, wool, and rabbit hair, *Fiber. Polym.,* 10(2009), 161-166.
39. G. Ke, W. Yu, W. Xu, Color evaluation of wool fabric dyed with *Rhizoma coptidis* extract, *J. Appl. Polym. Sci.,* 101(2006), 3376-3380.
40. Y. Son, B. Kim, K. Ravikumar, T. Kim, Berberine finishing for developing antimicrobial nylon 66 fibers: % Exhaustion, colorimetric analysis, antimicrobial study, and empirical modeling, *J. Appl. Polym. Sci.,* 103(2007), 1175-1182.
41. D. Das, S.R. Maulik, S.C. Bhattacharya, Colouration of wool and silk with *Rheum emodi*, Indian J. Fibre Text. Res., 33(2008), 163–170.
42. D. Das, S. R. Maulik, S.C. Bhattacharya, Dyeing of wool and silk with *Bixa Orellana*, *Indian J. Fibre Text. Res.,* 32(2007), 366-372.
43. M. P. Sathianarayanan, B. M. Chaudhari, N. V. Bhat, Development of durable antibacterial agent from ban-ajwain seed (*Thymus serpyllum*) for cotton fabric, *Indian J. Fibre Text. Res.,* 36(2011), 234-241.
44. S. Jafari, H. Izadan, A. Khoddami, M. Zarrebini, Investigation into the Dyeing of Soybean Fibers with Natural Dyes and their

Antimicrobial Properties, *Prog. Color Colorants Coat.* 7(2014), 95-104.
45. S. Ali, T. Hussain, R. Nawaz, Optimization of alkaline extraction of natural dye from Henna leaves and its dyeing on cotton by exhaust method, *J. Clean. Prod.,* 17(2009), 61-66.
46. V. R. Giri Dev, J. Venugopal, S. Sudha, G. Deepika, S. Ramakrishna, Dyeing and antimicrobial characteristics of chitosan treated wool fabrics with henna dye, *Carbohydr. Polym.,* 75(2009), 646-650.
47. R. Mongkholrattanasit, J. Krystufek, J. Wiener, Dyeing and fastness properties of natural dyes extracted from Ecalyptus leaves using padding techniques, *Fiber. Polym.,* 11(3)(2010), 346-350.
48. R. Mongkholrattanasit, J. Kryštůfek, J. Wiener, M. Viková, Dyeing, fastness, and UV protection properties of silk and wool fabrics dyed with eucalyptus leaf extract by the exhaustion process, *Fibers Text. East Eur.,* 19(2011), 94-99.
49. H. T. Deo, B. K. Desai, Dyeing of cotton and jute with tea as a natural dye, *JSDC,* 115(1999), 224-227.
50. E. Syamili, B. Elayarajah, Kulanthaivelu, R. Rajendran, B. Venkatrajah, P. Ajith Kumar, Antimicrobial cotton finish using green tea leaf extracts interacted with copper, *Asian J. Textile,* 2(2012), 6-12.
51. A. S. M. Raja, G. Thilagavathi, Dyes from the leaves of deciduous plants with a high tannin content for wool, Color Technol., 124(2008), 285–289.
52. P.S. Vankar, R. Shanker, S. Dixit, D. Mahanta, S.C.Tiwari, Sonicator dyeing of natural polymers with *Symplocos spicata* by metal chelation, *Fiber. Polym.,* 9(2008), 121-127.
53. A. A. Khan, N. Iqbal, S. Adeel, M. Azeem, F. Batool, I.A. Bhatti, Extraction of natural dye from red calico leaves: Gamma ray assisted improvements in colour strength and fastness properties, *Dyes Pigments.,* 103(2014), 50-54.
54. W. Haddar, I. Elksibi, N. Meksi, M. Farouk Mhenni, Valorization of the leaves of fennel (*Foeniculum vulgare*) as naturaldyes fixed on modified cotton: A dyeing process optimization based on a response surface methodology, *Ind. Crop. Prod.,* 52(2014), 588-596.
55. R. Rajendran, C. Balakumar, J. Kalaivani, R. Sivakumar, Dyeing and antimicrobial properties of cotton fabrics finished with *Punica Granatum* extracts, *JTATM,* 7(2011), 1-12.
56. A. Çalıs, G. Yuvalı Çelik, H. Katırcıoglu, Antimicrobial effect of natural dyes on some pathogenic bacteria, Afr. J. Biotechnol., 8(2)(2009), 291–293.
57. D. Das, S.C. Bhattacharya, S.R. Maulik, Dyeing of wool and silk with *Punica granatum, Indian J. Fibre Text. Res.,* 31(2006), 559-564.
58. A. Guesmi, N. Ben Hamadi, L. Ladhari, F. Sakli, Dyeing properties and colour fastness of wool dyed with indicaxanthin natural dye, *Ind. Crops Prod.,* 37(2012), 493-499.
59. A. Guesmi, N. Ladhari, N. Ben Hamadi, M. Msaddek, F. Sakli, First application of chlorophyll-a as biomordant: sonicator dyeing of wool with betanin dye, *J. Clean. Prod.,* 39(2013), 97-104.
60. B. Zhang, L. Wang, L. Luo, M. W. K., Natural dye extracted from Chinese gall - the application of color and antibacterial activity to wool fabric, *J. Clean. Prod.,* 80(2014), 204-210.
61. A. Guesmi, N. Ben hamadi, N. Ladhari, F. Sakli, Sonicator dyeing of modified acrylic fabrics with indicaxanthin natural dye, *Ind. Crops Prod.,* 42(2013), 63-69.
62. N. F. Ali, R. S. R. El-Mohamedy, Eco-friendly and protective natural dye from red prickly pear (*Opuntia Lasiacantha* Pfeiffer) plant, J. *Saudi Chem. Soc.,* 15(2011), 257-261.
63. M. Chairat, J. B. Bremner, K. Chantrapromma, Dyeing of cotton and silk yarn with the extracted dye from the fruit hulls of mangosteen, *Garcinia mangostana* linn, *Fiber. Polym.,* 8(6)(2007), 613-619.
64. K. H. Prabhu, M. D. Teli, N. G. Waghmare, Eco-friendly dyeing using natural mordant extracted from *Emblica officinalis* G. fruit on cotton and silk fabrics with antibacterial activity, *Fiber. Polym.,* 12(2011), 753-759.
65. S. S. Kulkarni, U. M. Bodake, G. R. Pathade, Extraction of natural dye from chili (*Capsicum Annum*) for textile coloration, Universal *J. Environ. Res. Technol.,* 1(2011), 58-63.
66. A. Pérez-Gálvez, H.D. Martin, H. Sies, W.

Stahl, Incorporation of carotenoids from paprika oleoresin into human chylomicrons, *Br. J. Nutr.*, 89(2003), 787-793.

67. S. M. Salah, Antibacterial activity and ultraviolet (UV) protection property of some Egyptian cotton fabrics treated with aqueous extract from banana peel, *Afr. J. Agric. Res.,* 6(20)(2011), 4746-4752.
68. X. Hou, X. Chen, Y. Cheng, H. Xu, L. Chen, Y. Yang, Dyeing and UV-protection properties of water extracts from orange peel, *J. Clean. Prod.,* 52(2013), 410-419.
69. M. Ajmal, S. Adeel, M. Azeem, M. Zuber, N. Akhtar, N Iqbal, Modulation of pomegranate peel colourant characteristics for textiledyeing using high energy radiations, *Ind. Crop. Prod.,* 58(2014), 188-193.
70. Ö. E. Ismal, L. Yıldırım, E. Özdogan, Use of almond shell extracts plus biomordants as effective textile dye, *J. Clean. Prod.,* 70(2014), 61-67.
71. K. N. Vinod, Puttaswamy, K. N. N. Gowda, R. Sudhakar, Natural colorant from the bark of *Macaranga peltata*: kinetic and adsorption studies on silk, *Color Technol.,* 126(2010), 48-53.
72. N. Pruthi, G. D. Chawla, S. Yadav, Dyeing of silk with barberry bark dye using mordant combination, *Nat. Prod. Radiance,* 7(1)(2008), 40-44.
73. H. F. Mansour, S. Heffernan, Environmental aspects on dyeing silk fabric with sticta coronata lichen using ultrasonic energy and mild mordants, *Clean. Techn. Environ. Policy,* 13(2011), 207-213.
74. A. K. Samanta, P. Agarwal, S. Datta, Dyeing of jute and cotton fabrics using Jackfruit wood extract: Part I– Effects of mordanting and dyeing process variables on colour yield and colour fastness properties*, Indian J. Fibre Text.* Res., 32(2007), 466-476.
75. A. K. Samanta, P. Agarwal, S. Datta, Dyeing of jute with binary mixture of Jackfruit wood and other natural dyes– Study on colour performance and dye compatibility, *Indian J. Fibre Text. Res.,* 33(2008), 171-180.
76. P. Guinot, A. Gargadennec, G. Valette, A. Fruchier, C. Andary, Primary flavonoids in marigold dye: extraction, structure and involvement in the dyeing process, *Phytochem. Anal.,* 19(2008), 46-51.
77. P. S. Vankar, R. Shanker, Eco-friendly pretreatment of silk fabric for dyeing with Delonix regia extract, *Color Technol.,* 125(2009), 155-160.
78. S. D. Bhattacharya, A. K. Shah, Metal ion effect on dyeing of wool fabric with catechu, *JSDC,* 116(2000), 10-12.
79. M Kumaresan, P. N. Palanisamy, P. E. Kumar, Application of eco-friendly natural dye on cotton using combination of mordants*, Indian J. Fibre Text. Res.,* 37(2012), 194-198.
80. A. K. Samanta, A. Konar, S. Chakraborti, Dyeing of jute fabric with tesu extract: Part 1– Effects of different mordants and dyeing process variables, *Indian J. Fibre Text. Res.,* 36(2011), 63-73.
81. R. Singh, A. Jain, S. Panwar, D. Gupta, S. K. Khare, Antimicrobial activity of some natural dyes, *Dyes Pigments.,* 66(2005), 99-102.
82. M. Ibrahim Khan, A. Ahmad, S. Ahmad Khan, M. Yusuf, M. Shahid, N. Manzoor, F. Mohammad, Assessment of antimicrobial activity of *Catechu* and its dyed substrate*, J. Clean. Prod.,* 19(2011), 1385-1394.
83. D. Gupta, A. Laha, Antimicrobial activity of cotton fabric treated with *Quercus infectoria* extract, *Indian J. Fibre Text. Res.,* 32(2007), 88-92.
84. M. Sumithra, N. V. Raaja, Antibacterial efficacy analysis of *Ricinus communis*, *Senna auriculata* and *Euphorbia hirta* extract treated on the four variant of denim fabric against *Escherichia coli* and *Staphylococcus aureus, J. Textile Sci. Eng.,* 2(2012), 1-4.
85. R. Zhang, Z. Cai, Study on the natural dyeing of wool modified with enzyme, *Fiber. Polym.,* 12(2011), 478-483.
86. S. M. Mortazavi, M. Kamali Moghaddam, S Safi, R. Salehi, Saffron petals, a by-product for dyeing of wool fibers, *Prog. Color. Colorants. Coat.,* 5(2012), 75-84.
87. X. Xinsheng, W. Lu, J. Shunhua, Z. Qicheng, Extraction of coloring matter from *Sargentodoxa cuneata* by ultrasonic technique and its application on wool fabric, *Indian J. Fibre Text. Res.,* 33(2008), 426-430.
88. O. Deveoglu, E. Torgan, R. Karadag, High-

performance liquid chromatography of some natural dyes: analysis of plant extracts and dyed textiles, *Color Technol.,* 128(2012), 133-138.

89. M. Mirjalili, K. Nazarpoor, L. Karimi, Eco-friendly dyeing of wool using natural dye from weld as co-partner with synthetic dye, *J. Clean. Prod.,* 19(2011), 1045-1051.
90. P. K. Mishra, K. Singh, K. K. Gupta, H. Tiwari, P. Srivastave, Extraction of natural dye from *Dahlia variabilis* using ultrasound, *Indian J. Fibre Text. Res.,* 37(2012), 83-86.
91. D. Cristea, G. Vilarem, Improving light fastness of natural dyes on cotton yarn, Dyes Pigm., 70(2006), 238–245.
92. A.S.M. Raja, G. Thilagavathi, Influence of enzyme and mordant treatments on the antimicrobial efficiency of natural dyes on wool materials, *Asian J. Textile,* 1(2011), 138-144.
93. A. Kiumarsi, R. Abomahboub, S. M. Rashedi, M. Parvinzadeh, *Achillea Millefolium*, a New source of natural dye for wool dyeing, *Prog. Color. Colorants Coat.,* 2(2009), 87-93.
94. J. S. Tchinda, A. Pétrissans, S. Molina, M. K. Ndikontar, S. Mounguengui, S. Dumarcay, P. Gérardin, Study of the feasibility of a natural dye on cellulosic textile supportsby red padouk (*Pterocarpus soyauxii*) and yellow movingui (*Distemonanthus benthamianus*) extracts, *Ind. Crop. Prod.,* 60(2014), 291-297.