Eco-Friendly of Textiles Dyeing and Printing with Natural Dyes

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Abstract : This article reports the studies available on the ultra violet (UV) protection property of natural dyes; antibacterial and deodorizing properties of natural dyes; application of natural dyes for textile printing; effect of different mordants and mordanting method; ultrasonic method of natural dyeing. Dyeing with natural dyes using padding techniques; thermodynamics and kinetics of dyeing with natural dyes have also been discussed.

Keywords: natural dyes, dyeing, printing, textiles, mordant, eco-friendly

1. Introduction

Natural dyes are known for their use in colouring of food substrate, leather, wood as well as natural fibers like wool, silk, cotton and flax as major areas of application since ancient times. Natural dyes may have a wide range of shades, and can be obtained from various parts of plants including roots, bark, leaves, flowers, and fruit [1]. Since the advent of widely available and cheaper synthetic dyes in 1856 having moderate to excellent colour fastness properties, the use of natural dyes having poor to moderate wash and light fastness has declined to a great extent. However, recently there has been revival of the growing interest on the application of natural dyes on natural fibers due to worldwide environmental consciousness [2]. Although this ancient art of dyeing with natural dyeing with natural dyes withstood the ravages of time, a rapid decline in natural dyeing continued due to the wide available of synthetic dyes at an economical price. However, even after a century, the use of natural dyes never erodes completely and they are still being used. Thus, natural dyeing of different textiles and leathers has been continued mainly in the decentralized sector for specialty products along with the use of synthetic dyes in the large scale sector for general textiles owing to the specific advantages and limitations of both natural dyes and synthetic dyes.

The use of non-toxic and eco-friendly natural dyes on textiles has become a matter of significant importance because of the increased environmental awareness in order to avoid some hazardous synthetic dyes. However, worldwide the use of natural dyes for the colouration of textiles has mainly been confined to craftsman, small scale dyers and printers as well as small scale exporters and producers dealing with high valued eco-friendly textile production and sales [2-4]. Recently, a number of commercial dyers and small textile export houses have started looking at the possibilities of using natural dyes for regular basis dyeing and printing of textiles to overcome environmental pollution caused by the synthetic dyes [5]. Natural dyes produce very uncommon, soothing and soft shades as compared to synthetic dyes. On the other hand, synthetic dyes are widely available at an economical price and produce a wide variety of colours; these dyes however produce skin allergy, toxic wastes and other harmfulness to human body.

There are a small number of companies that are known to produce natural dyes commercially. For example, de la Robbia, which began in 1992 in Milan, produces water extracts of natural dyes such as weld, chlorophyll, logwood, and cochineal under the Eco-Tex certifying system, and supplies the textile industry. In USA, Allegro Natural Dyes produces natural dyes under the Ecolour label for textile industry [6]. Aware of the Toxic Substance Act and the Environmental Protection Agency, they claim to have developed a mordant using a non-toxic aluminium formulation and biodegradable auxiliary substance. In Germany, Livos Pflanzenchemie Forschungs and Entwicklungs GmbH marked numerous natural products. In France, Bleu de Pastel sold an extract of woad leaves. Rubia Pigmenta Naturalia is The Netherlands company, which manufactures and sells vegetable dyes. There are several small textile companies using natural dyes. India is still a major producer of most natural dyed textiles [4].

For successful commercial use of natural dyes, the appropriate and standardized dyeing techniques need to be adopted without scarifying required quality of dyed textiles materials. Therefore, to obtain newer shades with acceptable colour fastness behaviour and reproducible colour yield, appropriate scientific techniques or producers need to be derived from scientific studies on dyeing methods, dyeing process variable, dyeing kinetics and compatibility of selective natural dyes. A need has also been felt to reinvestigate and rebuild the traditional processes of natural dyeing to control each treatment and pre-dyeing process (preparation, mordanting) and dyeing process variables for producing uncommon shades with balanced colour fastness and eco-performing textiles.

Production of synthetic dyes is dependent on petrochemical source, and some of synthetic dyes contain toxic or carcinogenic amines which are not eco-friendly [7]. Moreover, the global consumption of textiles is estimated at around 30 million tonnes, which is expected to grow at the rate of 3% per annum. The colouration of this huge quantity of textiles needs around 700,000 tonnes of dyes which causes release of a vast amount of unused and unfixed synthetic colourants into the environment [2].

In this article, we review the studies carried out so far on the application of natural dyes on textiles to understand the science of natural dyeing as well as to focus the ultra-violet (UV) protection properties, antibacterial and deodorizing properties, application of natural dyes for textile printing.

2. Application of natural dyes on textile

2.1 Ultra-violet (UV) protection properties with natural dyes

Recently, consumers have become increasingly aware of the need for sun protection, which is related to the incidence of sun-induced skin damage and its relationship with an increased exposure to UV light. Ultraviolet radiation (UVR) can lead to acute and chronic reactions and damage, such as acceleration of skin ageing [8]. An overdose of UV can cause various skin, eye, and even DNA damage [9]. The UVR band consists of three regions: the UV-A band (320–400 nm), the UV-B band (290–320 nm), and the UV-C band (200–290 nm) [10]. The highest energy region, the UV-C band, is absorbed completely by oxygen and ozone in the upper atmosphere. Of the total solar UV radiation reaching the earth's surface, 6% is in the UV-B region and 94% in the UV-A region [11]. UV-A causes little visible reaction on the skin but has been shown to decrease the immunological response of skin cells. Among the three radiations, UV-B is the most responsible for the development of skin cancers [12]. Therefore, the transmittance of UVR, including UV-A and UV-B, through the fabrics was evaluated in research.

There are many factors involved in the development of skin cancers and cumulative UV exposure of a patient is an important variable. Besides avoiding the sun, the most frequently recommended form of UV protection is the use of suitable clothes, hats, and sunscreens [13]. The protective properties of suitable clothes depend on fibre composition (natural or synthetic fibres), fabric construction (porosity, weight, and thickness), and the wet processing history of the fabric (using dyes, UV absorbers, and other finishing chemicals) [12-14] From extensive literature surveying, it can be observed that there are some recent reports for application of natural dyes on UV protective textile materials. Sarkar [12] evaluated of UV protection for different structures of cotton fabrics (plain, twill and sateen weave) dyed with madder, cochineal and indigo at different conditions and reported that dyeing of cotton fabrics with natural colourants increases the ultraviolet protective abilities of the fabrics and can be considered as an effective protection against ultraviolet rays. The Ultraviolet Protection Factor (UPF) is further enhanced with colourant of dark hues and with high concentration of the colourant in the fabric. Kim [9] studied the dyeing characteristics and UV protection property of green tea dyed on cotton fabric by using chitosan mordanting condition. The results show that chitosan mordanting can effectively increase the UV protection property of both UV-A and UV-B of green tea dyed cotton fabrics. Chitosan mordanted undyed cotton and chitosan unmordanted dyed cotton did not show an increase in UV protection property. Feng et al. [10] reported the ultraviolet protective properties of the cotton and silk fabrics dyed by *Rheum and Lithospermum erythrorhizon*. Experiment results revealed that the fabrics dyed with natural dyes had good ultraviolet protective properties. They could absorb about 80% of the ultraviolet rays. Wang et al. [14] researched the dyeing and ultraviolet protection of silk fabric using vegetable dyes extracted from Flos Sophorae. It was found that the aqueous solution of this vegetable dye has excellent thermal stability in acid conditions. The optimum extraction conditions were obtained for Flos Sophorae: extraction temperature of 100°C, extraction time of 60 min, and material, and material to liquor ratio of 1:10. The UPF and T $(UVA)_{AV}$ values for the silk fabric dyed by the optimum dye solution were found to meet the Chinese Standard (UPF = 69 > 30, T $(UVA)_{AV}$ = 1.07% < 5%). According to the standard, the silk fabric can claim to be a “UV-Productive product”. Grifoni et al. [15] studied the UV protection properties of flax and hemp fabrics dyed with weld, dyer’s woad, logwood lipstick tree, madder, brasil wood, and cochineal as natural dyes. Experiment results revealed that natural dyes could confer good UV protection. Weld-dyed fabric gave the highest protection level. Mongkholrattanasit et al. [16] reported the UV protection properties of silk fabric dyed with eucalyptus leaf extract. Experiment results observed that with an increase in the dye concentration.

2.2 Antibacterial and deodorizing properties of natural dyes

Textile materials and garment are susceptible to microbial attack, as these provide large surface area and absorb moisture required for microbial growth [17]. Natural fibres have protein (keratin) and cellulose, etc., which provide basic requirements such as moisture, oxygen, nutrients and temperature for bacterial growth and multiplication. This often results in objectionable odour dermal infection, product deterioration allergic responses and often related diseases [18]. The control of microorganisms (e.g. bacteria, mildews molds, yeasts and viruses) on textile fabrics extends to diverse areas such as hospitals, the environment and everyday households [19]. Although known for a long time for dyeing as well as medicinal properties, the structures and protective properties of natural dyes have been recognized only in the recent past. Many of the plants used for dye extraction are classified as medicinal, and some of these have recently been shown to possess remarkable antimicrobial activity [20]. A number of studies have indicated that plants synthesize aromatic substances such as alkaloids, terpenoids, and phenolic compounds as their secondary metabolites [21]. These compounds are antimicrobial and are produced by the plants in response to an attack by a pathogen; therefore their function is that of defense mechanism for plants against microorganisms. These compounds are of great importance as the substances have been successful in protecting plants from bacterial attack since time immemorial, without causing the bacteria to develop resistance to them [21].

The use of natural products and natural dyes for antimicrobial finishing of textile materials has been widely reported. Han and Yang [22] studied the antimicrobial activity of wool fabric dyed with curcumin and found that the antimicrobial ability of curcumin finished wool is semidurable, more durable to home laundering than to light exposure. The inhibition rates against *Staphylococus aureus* (*S. aureus*) and *Escherichia coli (E. coli)* were 45% and 30% respectively, after 30 cycles of home laundering. The inhibition rate against *S. aureus* was almost 80% after 5-AFU (AATCC Fading Unit) light exposure but started to decrease substantially after 10-AFU. The *E. coli* inhibition against light was poor. After 5-AFU exposure, the fabric retained only 30% of its original antimicrobial ability. Dahl et al. [23] also studied photokilling of bacteria by the natural dye curcumin and revealed that Gram-negative bacteria displayed greater resistance to curcumin phototoxicity relative to Gram-positive bacteria. Oxygen was required for curcumin phototoxicity. The crude methanolic extracts of stem root, leaves, fruit, seeds of *Artocarpus heterophyllus* [24] and their subsequent partitioning with petrol, dichloromethane, ethyl acetone and butanol fractions exhibited a broad spectrum of antibacterial activity. The butanol fractions of the root bark and fruit were found to be the most active. None of the fraction was active against the fungi test. Mariegold [25] showed negative test against microbiological control for *E. coli* and *Salimonella*. Singh et al. [20] tested *acacia catechu*, *kerria lacca*, *quercus infectoria*, *rubia cordifora*, and *rumex maritimus* against pathogens like *E. coli*, *Bacillus subtilis*, *Klebsiella pneumoniae*, *Proteus vulgaris*, and *Pseudomonas aeruginosa*. Minimum inhibitory concentration was found to vary from 5 μg to 40 μg. Gupta et al. [26] studied antimicrobial property of eleven natural dyes (acacia, indigo, lac, kamala, pomegranate, gall nuts, cutch, myrobolan, himalayan rhubarb, Indian madder and golden dock) against three types of Gram-negative bacteria (*E. coli*, *Klebsiella pneumoniae* and *Proteus vulgaris*. Seven out of the eleven natural dyes (lac, kamala, pomegranate, gall nuts, cutch, myrobolan and himalayan rhubarb) showed activity against one or more of the three bacteria studied. Kamala, pomegranate and gall nuts were found to be efficient biocides after dyeing on cotton, particularly gall nuts. The latter was highly effective against *E. coli* and *Proteus vulgaris*, reducing the number of colonies by 99%. The dyes examined exhibited good wash fastness. Ke et al. [27] reported the colour evaluation and antibacterial property of wool fabric dyed with root of *Rhizoma coptidis* (berberine). Results indicated that wool fabrics dyed with mordant, or at higher temperature, or in alkaline solution processed deeper shades and darker colour. And the wool fabric showed good antibacterial property after dyeing with *R. coptidis* extracts. Chen and Chang [28] studied the antimicrobial activity of cotton fabric pretreated microwave plasma and dyed with onion skin and onion pulp extraction. The best inhibition zone of anti *S. aureus* is found to be 1.1-0.8 cm by 10 min plasma-treated grafting time of onion skin extraction and 0.7-0.5 cm by 30 min plasma-treated grafting time of onion pulp extraction. The samples with 10 and 30 min plasma-treated grafting of both onion skin and onion pulp show anti *S. aureus* ability even after 5 times test washing, but both the samples lost their anti *S. aureus* property with 60 min grafting. The FTIR-ATR spectrum of dyed cotton fabric shows flavonoids function peak at 1624 cm^{-1} of onion skin that provide cotton fabric brown colour with wash fastness rating 4. Perumal et al. [29] reported the antibacterial activity of pigment from *Sclerotinia sp*. and its use in dyeing cotton. Dev et al. [30] studied the dyeing and antimicrobial characteristics of chitosan treated wool fabrics with henna dye. The microbial reduction percentage values for chitosan treated samples against both *E. coli* and *S. aureus* show that the microbial reduction is better against both bacteria. The fabrics dyed with henna alone without chitosan application do exhibit significant antimicrobial activity. The combined antimicrobial effect of chitosan and natural dyes is very good and can be used to develop clothes for protecting against common infection. Joshi et al. [31] also studied a comprehensive review on natural product based bioactive agents such as chitosan, natural dyes, neem extract and other herbal products for antimicrobial finishing of textile substrate.

There are only few reports [32-34] on the dyeing of natural fibers for the improvement of deodorizing property with natural dyes. Hwang et al. [32] reported deodorizing property of cotton, silk, and wool fabrics dyed with gardenia, coffee sludge, *Casia tora*. L., and pomegranate extracts. The deodorizing performance of fabrics dyed with various natural colourants extracts was in the range of 50-99%. The deodorizing performance increased in the order of gardenia < *Casia tora*. L. < coffee sludge < pomegranate. Especially the deodorizing performance of all fabrics dyed with pomegranate was found to be highest 99%. Lee [33] studied deodorizing property of cotton, silk, and wool fabrics dyed with coffee sludge (*Coffee Arabica* L.) extract. It was found that fabrics dyed with the *Coffee Arabica* L. extract showed good deodorization performance. Lee et al. [34] also studied deodorizing and dyeing properties of cotton, silk, and wool fabrics dyed with *Amur Corktree, Dryopteris crassirhizoma*, *Chrysanthemum boreale, Artemisia* extracts. The results revealed that deodorizing performance of fabrics dyed with various natural colourant extracts between 34% and 99%. It is worth nothing that the use of natural colourant notably enhanced the deodorizing performance. Wool fabrics showed the highest performance increase at 98-99%, followed by silk and cotton.

2.3 Application of natural dyes for textile printing

There are four reports [35-38] on the printing of textile materials with different natural dyes. Rekaby et al. [35] studied the printing of natural fabrics (wool, silk, cotton and flax) with natural dyes from alkanet and rhubarb by using pigment-printing technique. Results show that the highest *K/S* value was obtained by using Meypro gum as a thickener. The *K/S* increases rapidly as the concentration of the natural dye powder in the printing paste increases from 10 to 40 g/kg printing paste. Moreover, results show that the printed goods, which were fixed via steaming, have relatively higher colour strength than their corresponding samples fixed via thermodynamics. The best results were obtained by using metal mordants at a concentration of 20 g/kg printing paste. The colour fastness results were ranging between very good and excellent. Karolia and Buch [36] studied the resist printed natural dyed textile of *Ajarkh*. Hebeish et al. [37]

evaluated the reactive cyclodextrin in cotton printing with *Lawsonia Intermis Linn lythracease* (henna) as natural dye. Hakeim et al. [38] studied the pretreated cotton fabric by chitosan was printed with natural colouring matter, curcumin. The colour yield of the prints increased by increasing the molecular weight of chitosan. The stiffness results of the printed cotton fabric pretreated with low molecular weight chitosan showed better performance. The rubbing fastness of the prints was good.

2.4 Thermodynamics and kinetic of dyeing with natural dyes

It is felt essential to develop a knowledge base on dye chemistry and effects of dyeing process variables as well as rate of dyeing and chemical kinetics of dyeing for different natural dyes and fibers combinations to manipulate the processes of natural dyeing efficiently in order to get maximum colour yield in economical way [2].

Das et al. [39] studied the diffusion coefficient, rate of dyeing and effect of variation in salt and pH of wool and silk with annato (*Bixa orellana*). Wool dyed at 90°C appears to have more time of half dyeing (*t ½)* than silk dyed at the same temperature, thereby indicating that the wool has lower rate of dyeing as compared to silk. The diffusion coefficient of annato on silk higher than on wool. The exhaustion of annato to wool and silk is found to be maximum at dye bath pH ~ 4.5. The exhaustion of the dye increases with the increase in dye bath pH. For pre- and post- salt application, the ability of the salt to incorporate colouring component present in annato in the structure of wool and silk fiber follow a common trend: ferrous sulfate > aluminium sulfate > magnesium sulfate, as dividend from the *K/S* values of the respective dyes substrate. Samanta et al. [40] has studied the dyeing adsorption isotherm, heat of dyeing, free energy and entropy of dyeing for jackfruit wood. Study with jackfruit wood revealed that this dyeing process follows a Nernst adsorption isotherm, except jute-$FeSO_4$-jackfruit combination of natural dyeing, where it follows Langmuir adsorption isotherm. Farizadeh et al. [41] reported the extraction, identification and sorption of dyes from madder on wool fabric. The results show that adsorption dependent on the pH and temperature, and the adsorption isotherm of madder on wool is a Langmuir type. The adsorption of madder on wool is an exothermic process. Gupta and Gulrajani [42] studied and reported the kinetics and thermodynamics of dyeing with lawsone (2-hydroxy-1, 4-naphthoquinone) on wool, hair, silk, tussah, nylon and polyester fibers. Diffusion coefficient of silk and nylon showed the highest rate of dyeing followed by wool, tussah and hair fibers. Polyester dyed at 130°C had the slowest rate of dye uptake. The linear isotherm of dyeing for all fibers conformed to the partition mechanism of dyeing corresponding to the solid-solution model observed in dyeing hydrophobic fibers with disperse dyes. The standard affinity for all fibers increased with increase in temperature. Kongkachuichay et al. [43] studied thermodynamics of adsorption of laccaic acid on silk. The adsorption isotherm obtained was identified to be Langmuir type. When the temperature increased, the partition ratio and standard affinity decreased drastically. The values of heat of dyeing and entropy of dyeing were -13 kcal/mol and -0.03 kcal/mol/K, respectively. Chairat et al. [44] also studied an adsorption and kinetic of lac dyeing on silk and found that the experimental data fitted well to the Langmuir and Freundrich isotherms with a high correlation coefficient (R^2). The pseudo second-order kinetic model was indicated with the activation energy of 47.5 kJ/mol. The values of the enthalpy (ΔH) and entropy of activation (ΔS) were 44.7 kJ/mol and -175.7 J/mol K, respectively. The free energy of activation (ΔG) at 30°C was 97.9 kJ/mol. Chairat et al. [45] reported adsorption and kinetic of lac dyeing on cotton under dyeing conditions of pH 3.0, liquor ration 1:100 and an initial dye concentration 480 ± 10 mg/L. It was found that the adsorption kinetics of lac dyeing on cotton with pH control was found to follow the pseudo second-order kinetic model with an activation energy of 42.4 kJ/mol. Dyeing properties of silk fabric with berberine in terms of the thermodynamic and kinetic factors, including standard affinity, enthalpy change, entropy change, dyeing rate, diffusion coefficient and activation energy of diffusion, as reported by Ke et al. [27]. The results show that the adsorption isotherm of berberine on silk fabric belongs to Langmuir type. The analysis of dyeing thermodynamics shows that the adsorption of berberine on silk fabric is an exothermic process. When the fabric is dyed at higher temperature, the lower affinity and less dye uptake are obtained; however, the higher temperature increases the initial dyeing rate and diffusion coefficient. Vinod et. al. [46] studied and reported the kinetic and adsorption of dyeing with natural colourant from the bark of *Macaranga peltala* for silk yarn. The adsorption studies of *Macaranga peltala* for silk yarn revealed that the process fits well with the Langmuir isotherm model. The experiment results found that the adsorption was exothermic and spontaneous in nature, and exhibited first-order kinetics. The rate of adsorption increases as the disrupting effect of the added electrolyte cation increases and follows the order: $Al^{3+} > Ca^{2+} > Na^{+}$. Aydin et al. [47] investigated of the adsorption isotherm used for wool dyeing by aqueous extraction of Cehri fruit (*Fructus Rhamni Petiolari*) and reported that the adsorption isotherm can identified to be Nernst type. Septhum et al. [48] studied an adsorption of alum-morin dyeing onto silk yarn and revealed that the pseudo second-order kinetic model was indicated for alum-morin dyeing (simultaneous mordanting) of silk at pH 4.0 with ac activation energy (E_a) of 45.26 kJ/mol. The value of the enthalpy of activation (ΔH) for alum-morin dyeing on silk at pH 4.0 was 31.29 kJ/mol. Also the free energy ($\Delta G°$) and entropy changes ($\Delta S°$) for alum-morin on silk were -17.73 kJ/mol and -45.7 kJ/mol K, respectively consistent with a spontaneous and exothermic adsorption process.

2.5 Ultrasonic method of natural dyeing

Customer's demand for ecofriendly textile and ecofriendly dyes led to the revival of natural dyes for textiles, with the newer energy efficient dyeing process and more reproducible shade developing process [2]. Vankar et al. [49] studied and reported ecofriendly sonicator dyeing of cotton with *Rubia cordifolia* Linn. using biomordant. Use of biomordant replaces metal mordants, thus making natural dyeing more ecofriendly. Vankar et al. [50] also studied conventional and ultrasonic methods of dyeing cotton fabric with aqueous extract of *Eclip alba*. The effects of dyeing show higher colour strength values obtained by the latter. Kamel et al. [51-52] dyed wool and cationised cotton with lac as a natural dye using both conventional and ultrasonic techniques. Ultrasonic provide effectiveness in dye-uptake of cationised cotton and wool fabrics with lac dye, and enhanced effect after was about 66.5% for cationised cotton and 41-47% for wool. Ecofriendly ultrasonic textile dyeing with natural dyes such as *Acacia catechu* and *Tectona grandis* show better and faster dye uptake after enzyme pretreatment on cotton fabric, and results of dyeing are better than metal mordanted fabric, as reported by Vankar and Shanker [53]. Vankar and Tiwari [54] studied the sonicator and conventional dyeing of cotton fabric from *Melastoma malabathricum* and found that the percentage dye uptake of sonicator dyeing technique shows higher than the conventional dyeing. Vankar et al. [55-56] also studied sonicator dyeing of cotton, wool and silk with *Mahonia napaulensis DC* and *Symplocos spicata*. Vankar et al. [57] reported the sonicator dyeing of cotton with the leaves extract *Acer pectinatum* Wallich and the results show that the net enhancement of dye uptake due to metal mordanting and sonication has been found to range from 25% to 60% in the case of cotton compared to the controlled samples at 19-55%. Sivakumar et al. [58] also reported ultrasound energy dyeing of leather with beetroot for industrial and also found to be beneficial in natural dyeing of leather with improved rate of exhaustion. The ultrasonic technique has been used to dyeing on wool fabric with *Sargentodoxa cuneata* as natural dye, as studied by Xinsheng et al. [59].

2.6 Different mordants and mordanting methods

Mordanting is the treatment of textile fabric with metallic salts or other complex forming agents which bind the natural mordantable dyes onto the textile fibers. Mordanting can be achieved by either pre-mordanting, simultaneously mordanting or post-mordanting [2]. Different types and selective mordants or their combination can be applied on the textile fabrics to obtain varying colour or shade, to increase the dye uptake and improve the colour fastness behaviour of any natural dye [2]. Extensive work has been reported [60-73] in this area study.

Ali et al. [74] studies the effect of pre-mordanting and post-mordanting with aluminium sulfate, ferrous sulfate under different and reported that pre-mordanting and post-mordanting with ferrous sulfate, there was huge change in hue and a great deal of decrease in the chroma or purity of colour. Also, alum and iron did not result in any appreciable increase in fastness properties [74]. Wool and silk fabrics have been dyed with colourant extracted from *Rheum emodi* in the absence and presence of magnesium sulfate, aluminium sulfate and ferrous sulfate mordants for producing shades of different colours, ranging from yellow to olive green as reported by Bhattacharya et al. [75]. Bhattacharya et al.. [76] reported the methods of dyeing silk with some natural colourants derived from arjun bark (*Terminilia-Arjuna*), babul bark (*Acacia arabica*) and pomegranate rind (*Punica granatum*) and also mordanted with various mordants viz. tannic acid, copper sulfate, stannous chloride, ferrous sulfate and aluminium sulfate by using different mordanting techniques (pre, post and meta mordanting). It was found that dyed samples were compared to the un-mordanted dyed samples which indicate that mordanting improves colour strength, brightness as well as fastness properties of the dyed silk fabric. Shin and Cho [77-78] studied the wool and cotton fabrics treated with different mordant types and also various mordanting techniques were used to dye with American fleabane colourant. It was found that the dye affinity of cotton fiber lower than wool and the *K/S* value of pre-mordanting was higher than simultaneous mordanting or post-mordanting. Ameican fleabane produced mainly yellowish colour on mordants and mordanting method. Pruthi et al. [79] optimized the dyeing of silk with barberry bark dye (*Berberis aristata* Dc.) using mordant combination and found that the dyed samples possess very good to excellent fastness. Chairat et al. [80] extracted colour dye component from the dried fruit hulls of mangosteen (*Garcinia mangostana* Linn) was used for dyeing of cotton and silk yarn. The results showed that the dyeing of cotton using the post-mordanting method with ferrous sulfate and calcium hydroxide not only provided better depth of shade but also provided better wash fastness and light fastness than with mordants (alum; zinc tetrafluoroborate) or without a mordant. Deo and Desai [81] reported the dyeing of cotton and jute with an aqueous extract of tea and shows that dyeing was carried out with and without metal salts as mordants, using three different dyeing methods: pre-mordanting, meta-mordanting and post-mordanting. The resulting wash and light fastness of the dyed fabrics were good to excellent. The exhaust dyeing process of wool yarn, cotton fabric and polyamide 6, 6 with immediate use of the ash-tree bark (*Fraxinus excelsior* L.) extract as a dyebath and direct addition of $FeSO_4.7H_2O$ stock solution as a meta-mordant process showed good shade reproducibility and satisfying levelness of the dyed material as reported by Bechtold et al. [82]. Vankar and Shanker [83] studied dyeing of silk fabric with flower of *Delonix regia* extract and using a biomordant and enzyme. A bright reddish brown hue colour was observed when 30% owf *Delonix* extract was used on the pretreated silk material and also found that dyed fabric showed resistance to fading when mordanting with an enzyme or biomordant. Osman et al. [84] evaluated the effect of levelness by using different variables, including: three different natural fabrics; namely, wool, silk and cotton dyed with yellow natural dye from onion skins under the effect of different mordants, and also three different natural dyes; namely, onion skins, turmeric and madder applied on wool fabric samples under the effect of different mordants. The results show that dyed samples with the highest colour strength (*K/S*) have the highest unlevelness and the lowest colour difference values (i.e. the highest light fastness). Hou et al. [85] studied dyeing and mordanting methods properties to wool fabrics of catechu dye purified by micro-filtration membrane. The results show that the liquor of catechu dye is stable at pH values of 3-7 and its colour changes to a deeper brown-red when its pH value is above 8. The dyed wool fabric has good colour fastness to washing, alkaline perspiration and dry rubbing. Shanker and Vankar [86] reported the use of aqueous extract of Gulzuba flower (*Hibiscus mutabilis*) with metallic salts yield shades with good fastness properties for cotton, wool and silk dyed fabric. Cotton and silk fabrics were dyed with *Cassia tora L.* extract at 90°C for 60 min with pre-mordant of various metal salts as mordants as studied by Lee and Kim [87]. It was found that the *K/S* of cotton fabrics increased in the order of the dyeing using $FeSO_4 > CuSO_4 > ZnSO_4 > MnSO_4 \approx Al_2(SO_4)_3 > NiSO_4 >$ none, however, the *K/S* of silk fabrics increased in the order of the dyeing using $FeSO_4 > CuSO_4 > ZnSO_4 \approx Al_2(SO_4)_3 > MnSO_4 \approx NiSO_4 >$ none. Mordants $FeSO_4$ and $CuSO_4$ for cotton fabric, $FeSO_4$ and $CuSO_4$ and $Al_2(SO_4)_3$ for silk fabric were found to give good light fastness. El-Shishtawy et al. [88] studied dyeing of modified acrylic fibers with curcumin and madder natural dyes by using absence and presence alum and ferrous sulfate as mordant. It can be seen that acrylic samples dyed with madder in the absence and presence mordant show the beige to brownish shade. However, lemon yellow to brownish- yellow was found in acrylic dyed with curcumin. Cotton and silk fabrics dyed with African marigold flower (*Tagetes Ereecta* L.) extract by using iron and copper sulfate as a mordant have a good light fastness and also good to excellent wash fastness as studied by Jothi [89]. Lokhande and Dorugade [90] also studied and reported the effect of nylon fabric was dyed with three natural dyes derived from onion (*Allium cepa*), lac (*Laccifer Lacca*) and turmeric (*Curcuma longa*) using various mordants by two different techniques. (viz. open bath and high temperature high pressure (HTHP) dyeing methods). HTHP dyeing has been found to give better results as compared to the open bath dyeing. Good wash fastness was obtained with all three natural dyes.

2.7 Dyeing with natural dyes using padding techniques

The *padding technologies* are particularly advantageous to dyeing with the low-affinity products, because the dye affinity to fiber by padding is unnecessary (in phase of the dye deposition on the fabric) [91]. The dye bath is cloth "padded": mechanically applied by the rapid passage through the small padding trough, the intensive squeezing between expression rollers follows immediately. The process of padding is continuous and very rapid. It depends on the arrangement of the following dye fixation if the total procedure is continuous or semi continuous.

The dye bath by padding is about one order higher than by the common dyeing from the "long bath" (the so-called *exhaustion methods*), in which the dyestuff *exhausts* on the fiber in consequence to its affinity to the fiber. The higher padding bath concentration results in more rapid dye diffusion in fiber during the next fixation operation. Much smaller bath volume (related to the fiber unit) causes the higher dye exploitation (see also Agarwal and Patel) [92].

In the case of natural dyes, the dye fixation is based on the reaction with the salts of complex-forming metals-mordants in the same or next bath-or the textile can be *pre-metalized* with mordant (this *pre-mordanting* is carried out from the long bath-the large non-effectiveness is mentioned above. Therefore, we also experimented with pad-dry principle at this operation) [91].

In semi continuous dyeing technology, several methods of dye fixation are known. The following two principles are important for our purpose:

(a) *fixation by drying*, the so-called *pad-dry* method, the process is rapid but requires a reliably functional drying device (an excellently even -drying effect breadth-ways and cross-ways in the fabric is necessary, otherwise it may result in colour depreciation and unevenness),

(b) *fixation by batching* of the padded goods at room or slightly increased temperature, now known as the *pad-batch* method. The padded and rolled goods are wrapped up in an airtight plastic sheet so that no selvedge drying occurs during storage, which lasts 8–24 hours.

After both dye fixation methods water rinsing follows repeatedly.

Most research on natural dyes has been focused on the fundamental aspects of natural dyes by exhaustion process. Little attention has been give to the continuous process such as pad-batch and pad-dry techniques [16, 91-94]. Mongkholrattanasit et al. [91] reported that silk and wool fabrics dyed with the water extract of eucalyptus leaves in the presence of the $FeSO_4$ mordant in the same padding bath show a color range of a brown grey shade to a dark grey shade. The yield (exploitation) of the coloring component of eucalyptus leave is surprisingly good in wool fabric (about 68%–52% from the lowest to the highest concentrations), and this corresponds to the medium deep brown-grey shades in the concentrations of more than 20 g/L eucalyptus leaf extract [91]. In the silk fabric, the exploitation is less favorable and the decline with the changing to deeper shades is more distinct (about 22% to 15% exploitation) [91]. Agarwal and Patel [92] investigated the wool fabric dyed with natural dye extracted from the skin of babool bark with and without mordants using padding techniques. The results show that the dye liquor ratio is done using two padding techniques, namely cold pad-batch and pad-dry-steam techniques. Mongkholrattanasit et al. [93] has studied the dyeing and fastness properties of natural dyes extracted from eucalyptus leaves using padding techniques. It is reported that a natural dye extracted from eucalyptus leaves was applied to a silk and wool fabric by the use of two padding techniques, namely the pad-batch and pad-dry techniques under different conditions. Silk and wool fabrics dyed in a solution composed of eucalyptus extract from leaves showed a shade of pale yellow to brown. The exception was when the fabric was dyed with ferrous mordant, resulting in a shade of dark grayish-brown. The fastness properties ranged from good to excellent, while light fastness was fair to good. Mongkholrattanasit et al. [94] studied the properties of wool and cotton fabrics dyed with eucalyptus, tannin and flavonoids and reported that wool fabrics dyed with eucalyptus leaf extract, quercetin, rutin and tannin show higher colour strength than cotton fabrics. Tannins are considered as a main material in dyeing processes not only because of the shade similarities of eucalyptus leaves and tannin dyed on wool and cotton fabric but also because of the *K/S* and absorption spectra similarities of eucalyptus leaf extracts and tannin solutions.

The application of natural dyes on textile by pad-batch and pad-dry technique of dyeing can be considered as an affective eco-option because it gives extremely good results with substantial minimization of processing cost. In case of pad-dry technique, the average hot air consumption is considerably high whereas no hot air is being consumed in cold pad-batch process which leads to energy conservation. However, the time employed for the fixation of natural dye is very long in cold pad-batch technique. So, these techniques can be considered as best suitable for small scale industries or cottage dyeing of natural dyes.

3 Conclusions

Natural dyes cannot be used as simple alternatives to synthetic dyes and pigments. They do, however, have the potential for application, in specified areas, to reduce the consumption of some of the more highly polluting synthetic dyes. They also have the potential to replace some of the toxic, sensitizing and carcinogenic dyes and intermediates. It has been found that natural dyes are used in antibacterial, deodorizing, UV-protection and also food products.

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