IOSR Journal of Polymer and Textile Engineering (IOSR-JPTE)
e-ISSN: 2348-019X, p-ISSN: 2348-0181, Volume 2, Issue 3 (May - Jun. 2015), PP 74-78
www.iosrjournals.org

A Review on the Physico-Chemical Studies of Dyeing Progress and Dyeing Kinetics Using Natural Dyes

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Abstract: *It is felt very 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 fibres combinations to manipulate the processes of natural dyeing efficiently in order to get maximum color yield in economical way. A review on the effects of dye extraction medium, optimum concentrations of dye source material, extraction time, dyeing time, mordant concentration and methods of mordanting on silk dyed with natural dyes has presented in the article.*
Keywords*: Dyes, natural fibers, physico- chemical, silk textiles, etc.*

I. 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 prehistoric times. Natural dyes may have a wide range of shades 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, after many decades, 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.

Source-wise, the natural dyes have variable chemical compositions, which are influenced by a number of physical and chemical factors, besides the compositions of vegetal part of the plant from where the natural dye extracts are obtained. This again depends on conditions and place of growing, harvesting period, extraction methods and conditions, application method and technological process followed. Many workers have reported some of the most significant experimental results on laboratory trials regarding dyeing conditions, techniques and achievable best parameters for extraction and application of natural dyes, including observations on variation in extraction parameters, such as extraction temperature [3], extraction time, extraction solvent, vegetal material and liquor ratio, type of media or solvent used and also on observations of varying mordanting agent and technique as well as dyeing parameter. The effects of dye extraction medium, optimum concentrations of dye source material, extraction time, dyeing time, mordant concentration and methods of mordanting on silk dyed with natural dyes has been reported by Grover et al [4]. The acidic media exhibited maximum percent absorption for jatropha, lantana, hamelia and euphorbia dyes, while kilmora and walnut showed good results in alkaline medium. The result obtained from different experiments leads to the optimization of a standard recipe for a particular dye-mordant-fibre combination. Srivastava et al [5] studied the optimum dyeing technique for dyeing wool by determining the optimum wavelength, dye material concentration, extraction time, dyeing time, pH, concentration of mordant, etc. They also concluded that colouring of wool and silk textiles with tea extracts, which have highest affinity for both wool and silk at pH- 2 - 4 in presence and absence of either of the ferrous sulphate and aluminiumsulphate as mordants. Optimization of dyeing process variables for wool with natural dyes obtained from turmeric has been studied and reported by Agarwal et al.[6]. Bansal and Sood[7] studied the optimum conditions for development of vegetable dye on cotton from Eupatorium leaves. The optimization of dyeing of wool by Rhododendron arboretum as a natural dye source was reported by sati et al.[8]. Rose et al.[9].Maulik and Bhowmik[10], Siddiqui et al . [11] studied the effect of process variables on dyeing with selective natural dyes. The standardization and optimization of dyeing conditions are essential for effectively colouring any textile in a particular shade in techno-economic way to produce maximum color yield. Gupta et al.[l2] studied and reported the kinetics and thermodynamics of dyeing with Juglone for different fibres. The isotherm for wool, human hair, silk, nylon and polyester was linear, indicating a partition mechanism of dyeing.

The study observed the slope of isotherms was found to increase with the increasing temperature in all the cases. ΔH and ΔS values were positive for all these dyeing. The apparent diffusion coefficient was highest for wool and lowest for silk. Gulrajani et al.[l3] studied the kinetics of annatto and reported that it has high affinity for both nylon and polyester fibres. The process of dyeing is endothermic as the dye uptake increases at higher temperature. Mahaleet al.[l4] investigated the conditions of extraction and application of African mariegold on silk yarn. The optimum conditions were found to be 60 min dye extraction time, 30min mordanting, 30min dyeing using mixtures of 5% potash alum, l% potassium dichromate and l% copper sulphate as mordants. Also studied the dyeing absorption isotherm, heat of dyeing, free energy and entropy of dyeing for red and jackfruit wood[l5]. Study with red sandal wood revealed that this dyeing process follows a linear nernst absorption isotherm for jute; while study on jackfruit wood revealed that These studies are notably important for understanding the dyeing theory and physico- chemical dyeing parameters of different natural dye-fibre combinations. These are also important for practically manipulating dyeing conditions to get maximum color yield from particular natural dyes.

II. Color Interaction Parameters & dye Compatibility

Newer shades can be achieved by applying mixture of compatible natural dyes or by using sequential dyeing technique with two or three natural dyes. For the use of mixture of natural dyes, the dyers must know whether the natural dyes are compatible with each other or not. Reports are available on dyeing of different textiles with selective mixtures of natural dyes. According to one report[l6] it was observed that the use of a mixture of turmeric and madder on cotton in case of simultaneous mordanting shows a synergistic effect in color development than that for single dye application; 50:50 ratios of turmeric and madder gives the best results. For the combined dye application, it is observed that in case of simultaneous mordanting method, turmeric when combined with either madder or red sandal wood gives better color strength, while my robolan shows the reverse trend. Such studies using mixture of natural dyes on jute or cotton or any other textiles are rare and scanty. Several studies have been reported on compatibility of binary and tertiary mixture of synthetic dyes; however such studies on natural dyes are still rare and sporadic. Sakata & Kataryama[l7], Patel et al. [18] and Sudhaker & Gowda [l9] analyzed the different color parameters of silk fabric dyed with dry the Indian subcontinent madder by L 8, a 8 and b 8 values. The color of jute [20] dyed with aqueous extract of tea was investigated on computer aided color measuring system in terms of K /S and CIE Lab color difference values. CIE L 8 C 8 H 8 values were studied for cotton and woollen textiles dyed with yellow natural dye and it was found that the H 8 values for cotton and wool. This indicates the predominance of yellow hue with different mordants and high E values, Iron(II)sulphate mordant with high C 8 and H 8 values shows change in shade towards grey to black, as studied by Tastsaroni & Eleftheriadis[21]. Mishra et al .[22]studied dyeing of silk fabric for reddish shade with naturaldye of different concentrations and then measured the K/S values. Sarkar and seal [23] studied the influence of different mordanting systems on color strength and colorfastness of flax fabrics dyed with selective natural colorant. Results obtained were then analyzed and correlated with color strength and related color interaction parameters. Recently, Samanta et al .[24-26] have studied the compatibility of binary combinations of jackfruit wood with manjistha, red sandal wood, mariegold, sappan wood and babool applied on jute fabric. In this work, a newer and simple method of assessing relative compatibility rating (RCR) of pairs of natural dyes has been proposed with compatibility rating of 0-5 scale, (0 indicates completely non-compatible and 5 indicates highest order of compatibility), based on determination of newer color difference index (CDI) values. In a very recent report on the application of natural dyes even on synthetic fibres[27] , 100% nylon fabric does not pick up dyes such as henna and turmeric, but if it is pre-treated for 5 min with heat or / and acetic acid it readily picks up color. Gupta et al .[28] reported few spectral curves confirming the behavior of purpurin as a mordantable dye and showing the color change with the mordant due to the characteristic of metal - dye interactions. Thus, for producing variety of shades and variations in L 8, C 8, H 8 values, the use of different mordant-natural dye combination and mixture of natural dyes are the two important routes.

III. Effects of Chemical Pretreatments

The primary objective in chemical modification of a polymer/ fibre material is normally to preserve its original chemical structure and related favorable properties. Such chemical modifications may involve incorporation of new functional groups with or without large scale chain rupture or chain degradation. This however involves addition of some newly built-in property criteria, favoring development of the modified material for improved dye ability with or without affecting some other property criteria. There are many reports on the dyeing of unmodified and modified jute [29-32] and cotton [33] textiles with different classes of synthetic dyes. However, such studies are sporadic and scanty with natural dyes. There are only few reports available on the chemical modifications of cotton textiles for the improvement of their dye ability with natural dyes [33-35]. There is only one report available on chemical modification of jute substrate with acrylamide monomer using K-

2 S-208 initiation and Cetyltrimethyl ammonium bromide (CTAB) for investigating their effects on subsequent dyeing with natural dyes like turmeric and madder. Treatment of textile materials with Chitosan[36] obtained from the waste products of crab and prawn fishing is a possible route for the chemical modification of textile polymers to develop a variety of effects in textile dyeing and finishing applications. Bandhpoadhyay et al .[37] reported that the pre-treatment of cotton textiles with chitosan can reduce the amount of electrolyte addition during dyeing with reactive dyes and enhance fixation, but such studies using natural dyes are yet not reported. Sorption behavior of shellac, a natural thermosetting resin, on wool [36] fibers and enzymatic degradation of shellac-modified wool fibres has also been investigated with the use of methanol as solvent, the amount of sorbed shellac is found to be maximum (about 0.03g/g of wool), which is decreased with the increase in molecular weight of the alcohol from methanol to t-butanol. Shin et al.[38] studied the antimicrobial finishing of polypropylene nonwoven fabric by treatment with chitosan which has gained improved dye ability as well. Effect of low temperature plasma treatment [39] on colouring of wool and nylon-6 fabrics increases the dye uptake and saturated dye exhaustion for acid dye, despite the increased electro-negativity of the fibre surface. Pascual & Julia[40] reported that the pre-treatment with hydrogen peroxide improves the effectiveness of chitosan, Alkali pre-treatment followed by chitosan application increases the rate of dyeing and alters the physico-chemical parameters of dyeing kinetics and causes a reduction in hydrophobicity and shrinkage. However, a high concentration of chitosan can produce a rough hand and uneven dyeing, increasing the viscosity as well as resistance to shrink. Davarpanah et al .[41]studied and reported the surface modification of silk fibre using anhydrides to graft the polysaccharide chitosan and to improve dyeing ability of the grafted silk. The physical properties show acceptable changes, regardless of weight gain scanning electron microscopy (SEM) analysis shows the presence of foreign materials firmly attached to the surface of silk. FTIR spectroscopy provided evidence that the chitosan is grafted onto the acylated silk through the formation of new covalent bonds. The dyeing of the chitosan grafted acylated silk fibre indicated the higher dye ability in comparison to the acylated and degummed silk samples.
However, specific chemical modification is required to suit different purposes and the same has many odd effects too, i.e., increasing yellowing, variation in shades and colorfastness properties. These effects are not always favorable and hence one can use this judiciously with specific knowledge and study.

IV. Colour Fastness Properties of Natural dyed

Textiles color fastness is the resistance of a material to change any of its color characteristics or extent of transfer of its colourants to adjacent white materials in touch. The color fastness is usually rated either by loss of depth of color in original sample or it is also expressed by staining scale, i.e., the accompanying white material gets tinted or stained by the color of the original fabric. However, among all types of color fastness, light fastness, wash fastness and rub fastness are considered generally for any textiles; perspiration fastness is considered specifically for apparels only.

Light Fastness

Light fastness of many natural dyes, particularly which are extracted from flower petals are found to be poor to medium. So, an extensive work has been carried out to improve the light fastness properties of different natural dyed textiles. Cook [42] has reported a comprehensive review on different attempts made for improving color fastness properties of dyes on different textile fibres by different means. The study includes tannin-related after-treatments for improving the wash fastness and light fastness of mordantable dyes on cotton; some of these treatments might be applicable to specific natural dyes. Most of the natural dyes have poor light stability as compared to the best available synthetic dyes, and hence the colours in museum textile are often different from their original colors. The relative light stability of a range of dyes was reviewed by Padfield & Landi [43] who also studied changes in qualitative fashion. These changes in color were studied quantitatively by Duff et al .[44], who expressed the changes in terms of the Munsell scale and also in CIE color parameters. Wool dyed with nine natural dyes was exposed in Microscal MBTF fading lamp. The fastness ratings were similar to those observed by Padfield & Landi [43] in day light fading. After rating by the blue wool standards for light fastness (LF) rating, yellow dyes (old fustic, Persian berries) showed poor light fastness (l-2) ; reds l(cochineal (tin mordant)], alizarin (alum and tin mordant) and lac (tin mordant) showed better light fastness (3-4); indigo showed light fastness rating of 3-4 or 5-6 depending on the mordant used; and logwood black (chrome mordant) showed light fastness rating of 4-5 or 6-7 using other mordants. Gupta [45, 46] reported the effects of chemical structure of natural dyes on light fastness and other colorfastness properties. A large proportion of natural dyes are of course, mordant dyes. There is strong influence of nature, type and concentration of mordants on wash and light fastness grades. The influences of different mordants were found to play important role in fading of l8-yellow natural dyes [47]. Wool dyed with different natural dyes specimens was exposed to a xenon arc lamp for assessing its light fastness up to 8 AATCC fading units equivalent to BS-8B blue wool standards. The corresponding color change after exposure to xenon arc lamp was also assessed in each case. Turmeric, fustic and mariegold dyes faded significantly more than any of the other yellow dyes. However, the use of tin and alum mordants causes significantly more fading than that with the use of chrome, iron, or copper mordant. Thus, the type of mordant is found to be more important than the dye itself in determining the light fastness of natural colored textiles. Oda[48,49] reported effects of various additives on the photo fading of carthamin in cellulose acetate film. The rate of photo fading was remarkably suppressed in the presence of nickel hydroxyl arylsulphonates, while the addition of UV absorbers afforded little retardation in the rate of fading. cristea and vilarim[50] , Lee et al .[51], Micheal & Zaher [52] and Gupta et al .[53] made various attempts to improve the light fastness of different fabrics dyed with natural dyes. Samanta et al.[26] have recently reported the improvement in light fastness to one to half unit for natural dyed jute textiles by application of l% benztriazole in specific conditions. The corresponding mechanism of action of benztriazole on jute has also been studied and reported by them.

Wash Fastness Duff et al.[54] studied the light fastness and wash fastness [44,45] under standard condition (5opc) and also at 2opc with a washing formulation used in conservation work for restoration of old textiles. Some dyes undergo marked changes in hue on washing due to the presence of even small amounts of alkali in washing mixtures, highlighting the necessity to know the pH of alkaline solutions used for the cleaning of textiles dyed with natural dyes. As a general rule, natural dyes show moderate wash fastness on wool, as assessed by the ISO II test. Logwood and indigo dyes are found to be much faster. Duff et al.[54] studied the wash fastness of dyeing from native Scottish and imported dyes. In the ISO II test, the fastness of the indigo and logwood was superior to that of the native natural dyeing, such as Persian berries and water-lily root respectively. But in comparison of native and imported yellow, reds, red/purples, greens and browns, there was little difference between the two groups. Samanta et al.[26] recently reported that the treatment with 2% CTAB or Sandofix-HCF improves the wash fastness to nearly l unit.

Rub Fastness

In general, rub fastness of most of the natural dyes is found to be moderate to good and does not require any after-treatment. Samantaet al .[26, 55] reported that the jackfruit wood, Manjistha, red sandal wood, babool and mariegold have good rub fastness on jute and cotton fabrics. Sarkar[23] also reported good rub fastness for mariegold on cotton, silk and wool. It also has good rub fastness property. It also reported that the cutch and ratanjot show moderate to good dry rub fastness but the wet rub fastness is found to be average. Samanta et al.[56] studied the dye color strength related parameters and compatibility for dyeing cotton fabrics with binary mixture of jackfruit wood and other nature dyes. However, it must be remembered that the color fastness of natural dyes not only depends on chemical nature and type of natural colorants, but also on chemical nature and type of mordants being used. So, a dyer must know the use of proper combinations of fibre-mordant to achieve best color fastness. More research is required on exploring about the use of natural after- treatment agents to improve both wash and light fastness of natural dyes.

V. Conclusions

It has still not become possible mainly due to the essential two dependant factors (dye and mordant) and color development mechanism of natural colors applied to textiles whether pre-mordanted or simultaneous mordanted or post-mordanted, the color depends not only on the natural colorant but also on the mordant and mordanting assistants used. The prediction of match by controlling these two variables simultaneously is not possible by the latest technology known so far. However, for specific pre- mordanting method, color matching database for natural dyeing of any particular textile material and match prediction are possible with computerized program. These studies will help to popularize the use of natural dyes by solving some of its problems relative to application methods, reproducibility and colorfastness. Improving the computerized color matching for use of synthetic dyes has now become a regular practice in most of the textile industry (except jute industry).

Acknowledgement

I would like to express my heartfelt appreciation to the authority of Primeasia University , We also extend my deepest thank to my family, my friends and for all those helm me in moral and other supports.

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