Dyeing of jute fabric with tesu extract: Part 1 — Effects of different mordants and dyeing process variables

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Selective pre-mordanting (single and double) and natural dyeing of 6% H₂O₂ (50%) bleached jute fabric have been carried out using myrobolan (harda) and metallic salts (patash alum and aluminium sulphate) as mordants and aqueous extract of tesu (palash flower petals) as dyeing agent under varying dyeing condition to optimize the dyeing process variables. It is found that the 20 % myrobolan followed by 20 % aluminium sulphate in sequence is a most potential double pre-mordanting system rather than using them as single mordant separately, considering the results of important textile-related properties and colour yield. Effects of dyeing process variables (time, temperature, pH, MLR, mordant conc., dye conc., and salt conc.) on surface colour strength have been evaluated to optimize dyeing conditions. Colour fastness to washing, rubbing and light, in general, and dyeing pH sensitivity, in particular, for selective fibre-mordants-dye systems have also been assessed and it is found that dyeing at pH 11 for the system offers overall good colour yield and colour fastness properties. Improvement in wash and light fastness is also achieved with suitable chemical post-treatment.

Keywords: Dyeing, Jute, Metallic Salt, Mordanting, Myrobolan, Natural dyes, Tesu

1 Introduction

Off late, organic awareness and environmental consciousness of consumers have revived the interest on natural dyed textiles from natural fibres. As consumers understand the value for money, organic and non-toxic ecofriendly textiles dyed with natural dyes, this has created a new brand icon in the market and therefore, textiles using ecofriendly natural dyes on natural fibres are in further revival stage. Most of the natural dyes are proved to be non-toxic and ecofriendly with some exceptions. Jute is coarser variety of agro-renewable and biodegradable multicellular natural fibre, chemically being lignocellulosic in nature. Being coarser, it is mainly used for low priced bulk packaging, i.e. sacking. But, now a days, it is also being used as wall decorative and furnishing fabrics, besides many other diversified jute products including home textiles and geotextiles. Jute is chemically composed of cellulose, hemi-cellulose and lignin as major constituents and accepts wide classes of dyes. Hence, unlike cotton, the dyeing characteristics of jute for any natural dyes are different owing to the differences in its chemical composition, chemical functionality pattern and differences in fine as well as gross structure from cotton.

Despite the common disadvantages for application of natural dyes on textiles such as practical difficulty in its two stage application (mordanting and dyeing), reproducibility, non-uniformity of shades and poor to moderate colour fastness, their special technical advantages include its uncommon and soothing shades, agro-renewable nature and ecofriendliness with few exceptions. However, except few discrete reports, there is lack of detailed scientific information on the exact chemistry of colour component of natural dyes, physical chemistry of dyeing, standardized dyeing process conditions and optimised dye extraction method for different natural dyes. Moreover, insufficient scientific information and knowledge possess further challenge to the textile chemists and dyers to achieve uniform and optimum colour yield. The availability of required scientific information and reports in literature for dyeing of silk and wool fibres with natural dyes are much abundant, but the same for jute is rather scanty and sporadic. Recently, some studies have been reported for

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the application of few natural dyes (jackfruit wood, red sandal wood and their mixture with other natural dyes) on jute and cotton. With this milieu, as a continuation of our earlier work on this aspect, it is thought appropriate to study the effects of selective single and double mordants on textile related mechanical properties as well as effects of dyeing process variables on colour yield and colour fastness of jute fabrics using aqueous extract of natural dye obtained from tesu (palash flower petals).

2 Materials and Methods

2.1 Materials

6% H$_2$O$_2$ (50%) bleached, plain weave jute fabric (215 tex warp, 285 tex weft, 64 ends/dm, 58 picks/dm, 320 g/m$^2$ fabric area density and 0.74 mm fabric thickness), obtained from M/s Golster Mills, Bowreah, Howrah, was used for the study. Laboratory reagents (LR) grade aluminium sulphate & potassium aluminium sulphate and commercial grade acetic acid, common salt, sodium carbonate, sodium hydroxide & non-ionic soap obtained from local suppliers were used. A natural mordanting assistant myrobolan (harda, botanically known as _Terminalia chebula_) powder was also used as one of the mordant assistants for the present study.

As natural dye, petals of tesu (_Butea monosperma frondosa_) were used to obtain creamish orange / yellowish orange / ochre brown / reddish brown colour shades using their aqueous extract with varying mordant for dyeing of jute fabrics. Structure of the main colour component of tesu extract has been identified earlier$^{14}$ as butein (Scheme 1), a chalcone of yellowish red / orange colour.

2.2 Methods

2.2.1 Extraction of Colour Component and Optimisation of Extraction Conditions

Tesu powder, prepared after grinding the sun dried petals of palash flower, was taken in hot water for extraction of its colour component under varying condition, such as MLR 1:5 - 1:40, pH 4-12, time period 15-120 min and temperature 30-100°C, in order to optimize the conditions of colour component extraction. Absorbance (optical density) at a particular maximum absorbance wavelength ($\lambda_{\text{max}}$, 490nm) for the aqueous extract of the tesu was estimated using Hitachi-U-2000 UV-Vis absorbance spectrophotometer. The wavelength scan of UV-Vis spectrum (Fig. 1) of coloured solution of aqueous extract of tesu indicates the characterized peaks and troughs at 300-600 nm out of the total scan at 190-1100 nm. The usual pH of aqueous extract of tesu flower petals was found to be 6; which, as per necessity, was either acidified with acetic acid or alkalfied with NaOH solution for altering or varying the pH.

Table 1 shows the various conditions used for the aqueous extraction of colour component and the respective absorbance values. Optimum values are identified by observed maximum absorbance value (for corresponding maximum colour yield) and are shown by bold letters in Table 1. Unless otherwise mentioned, in usual cases, the aqueous extract of dye liquor from tesu was prepared following the optimized conditions of extraction, i.e. time 60 min, temperature 90°C, MLR 1:20 and pH 11. For the study on each extraction variable, other variables were kept constant (bold values in Table 1), selected on the basis of maximum absorbance.

2.2.2 Purification of Colour Component

Pre-cut and sun-dried branches of tesu were initially crushed to powder form in a mechanical pulverizer cum grinder and then subjected to aqueous extraction under the above-mentioned optimum extraction conditions. To obtain purified dyes, aqueous extract of tesu powder was double filtered
and then gradually concentrated by evaporation technique with slow heating (60°C) in water bath to semi-dry solid mass. This was further solvent extracted using standard soxhlet extraction of the semi-dry mass of the dye wrapped in filter paper, using 50:50 ethyl alcohol : toluene mixture for 10 cycles nearly for 2h at 70°C. The solvent extracted component of tesu was then evaporated to dry mass of the purified colour component under low temperature drying in vacuum oven. The dry residue was then washed in methyl alcohol followed by washing with acetone and final drying in air to obtain the dry powder of the purified colour component of tesu.

### 2.2.3 Mordanting of Jute Fabrics with Myrobolan

The myrobolan powder was soaked in water (1:10 volume) for overnight (12h) at room temperature to obtain the swelled myrobolan gel. This gel was then mixed with a known volume of water and heated at 80°C for 30 min. The solution was then cooled and filtered in a 60 mesh nylon cloth and the filtrate was used as final mordanting solution (10-40%) —an additional mordanting assistant using MLR of 1:20.

Pre-wetted conventional H₂O₂ bleached jute fabrics were separately treated with the myrobolan (harda) solution in separate bath initially at 40-50°C and then the temperature was raised to 80°C. The mordanting was continued for 30 min. After the harda mordanting, fabric samples were dried in air without washing to make them ready for either subsequent dyeing or for subsequent second mordanting.

### 2.2.4 Mordanting of Jute Fabrics with Metallic Salts

Mill bleached (using H₂O₂) jute fabrics with or without initial first mordanting with harda were further mordanted prior to dyeing using 10-40% aqueous solution of aluminium sulphate [Al₂(SO₄)₃] and potash alum [KAl(SO₄)₂] at 80°C for 30 min using material-to-liquor ratio of 1:20. After this mordanting, the fabric samples were finally dried in air without washing to make them ready for subsequent dyeing. In all, 12 single mordanted [harda, Al₂(SO₄)₃ and KAl(SO₄)₂] samples and 8 double mordanted [harda+Al₂(SO₄)₃ and harda + KAl(SO₄)₂] samples were used for the further study.

### 2.2.5 Dyeing of Pre-mordanted Jute Fabrics

Bleached and pre-mordanted (single and double) jute fabrics were dyed using the aqueous colouring extract of tesu under varying condition. For general study of dyeing behaviour using different mordants, the normal dyeing condition [dye – liquor conc. of aqueous extract of tesu, 30%; mordant conc., 20%; MLR , 1:20; common salt , 10 gpl; pH , 11.0 (with requisite amount of NaOH); dyeing temperature, 100°C and dyeing time, 60 min] were used.

To study the effects of dyeing process variables on colour yield for optimizing the dyeing conditions, the parameters were varied taking dyeing time 30-120 min, dyeing temperature 60-100°C, material-to-liquor ratio 1:10 - 1:50, conc. of mordants 10-40%, conc. of aqueous extract of tesu (as natural dye) 10-60 %, common salt conc. 5-20 g/L and pH 4-12. The selective mordant systems used for the study of dyeing process variables were double pre-mordanting

<table>
<thead>
<tr>
<th>Extraction variable</th>
<th>Absorbance at λmax, 490nm</th>
</tr>
</thead>
<tbody>
<tr>
<td>Time, min</td>
<td></td>
</tr>
<tr>
<td>15</td>
<td>2.426</td>
</tr>
<tr>
<td>30</td>
<td>2.457</td>
</tr>
<tr>
<td>45</td>
<td>2.458</td>
</tr>
<tr>
<td><strong>60</strong></td>
<td><strong>2.462</strong></td>
</tr>
<tr>
<td>75</td>
<td>2.460</td>
</tr>
<tr>
<td>90</td>
<td>2.427</td>
</tr>
<tr>
<td>120</td>
<td>2.213</td>
</tr>
<tr>
<td>Temperature, °C</td>
<td></td>
</tr>
<tr>
<td>30 (RT)</td>
<td>2.345</td>
</tr>
<tr>
<td>45</td>
<td>2.356</td>
</tr>
<tr>
<td>60</td>
<td>2.391</td>
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<td>75</td>
<td>2.434</td>
</tr>
<tr>
<td><strong>90</strong></td>
<td><strong>2.462</strong></td>
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<tr>
<td>100</td>
<td>2.358</td>
</tr>
<tr>
<td>Material-to-liquor</td>
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<tr>
<td>ratio</td>
<td></td>
</tr>
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<td>2.435</td>
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<td>1:10</td>
<td>2.443</td>
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<td>1:15</td>
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<td>2.460</td>
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<tr>
<td>1:30</td>
<td>2.457</td>
</tr>
<tr>
<td>1:40</td>
<td>2.431</td>
</tr>
<tr>
<td>pH</td>
<td></td>
</tr>
<tr>
<td>4.0</td>
<td>1.918</td>
</tr>
<tr>
<td>6.0</td>
<td>2.016</td>
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<tr>
<td>7.0</td>
<td>2.319</td>
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<tr>
<td>8.0</td>
<td>2.324</td>
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<td>9.0</td>
<td>2.339</td>
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<tr>
<td>10.0</td>
<td>2.422</td>
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<tr>
<td><strong>11.0</strong></td>
<td><strong>2.462</strong></td>
</tr>
<tr>
<td>12.0</td>
<td>2.220</td>
</tr>
</tbody>
</table>

*Values in bold indicate the optimum values considered while studying the effect of other extraction parameters.*
with 10-40% harda (1st mordant) followed by 10-40% aluminium sulphate (2nd mordant) applied in sequence on jute. After the dyeing is over, the dyed jute fabric samples were repeatedly washed with hot and cold water and then dried in air. Finally, the dyed samples were subjected to soaping with 2 g/L soap solution at 60°C for 15 min, followed by repeated water wash and drying under sun. Unless otherwise stated, after optimization of the dyeing process variables, all other experiments of dyeing with tesu extract were carried out at optimum dyeing conditions observed.

2.2.6 Surface Colour Strength, Dyeing Uniformity and other Colour Interaction Parameters

$K/S$ values of dyed jute fabrics were determined by measuring surface reflectance of the dyed samples using a computer-aided Macbeth 2020 plus reflectance spectrophotometer. Following Kubelka Munk equation was used with the help of relevant Color-Lab plus colour measurement and matching software for calculating the $K/S$ value:

$$K/S_{\lambda_{\text{max}}} = \frac{(1 - R_{\lambda_{\text{max}}})}{2R_{\lambda_{\text{max}}}}$$

where $K$ is the coefficient of absorption; $S$, the coefficient of scattering; and $R_{\lambda_{\text{max}}}$, reflectance at maximum absorbance wavelength of $\lambda_{\text{max}}$.

Also, coefficient of variation (CV%) of $K/S$ values was determined from the 10 point $K/S$ data taken at 10 different points of the corresponding dyed fabric samples indicating the dye uniformity. The CV% of $K/S$ values was determined using the standard equations.

Total colour difference ($\Delta E$), lightness/darkness ($\Delta L^*$), redness/greenness ($\Delta a^*$), blueness/yellowness ($\Delta b^*$), change in chroma ($\Delta C^*$), and change in hue ($\Delta H_{ab}$) values were measured before and after dyeing to compare the shade depth and colour differences of each dyed sample against particular undyed (bleached/mordanted) standard sample.

General metamerism index (MI) was calculated employing following equation:

$$MI = \frac{\sum (AR_x)^2}{X^2} + \frac{\sum (AR_y)^2}{Y^2} + \frac{\sum (AR_z)^2}{Z^2}$$

where $\bar{x}$, $\bar{y}$, and $\bar{z}$ are the standard CIE observer functions for three primary colours, namely red, blue and green respectively; and $X$, $Y$ and $Z$ are the CIE tristimulus values of the corresponding samples.

Brightness index (BI) was calculated as per ISO-2470-1977 method using the following relationship:

$$\text{Brightness index} = \frac{\text{Reflectance value of sample at 457 nm}}{\text{Reflectance value of standard diffuser (white tile) at 457 nm} \times 100}$$

2.2.7 Measurement of Colour Difference Index

A newer colour interaction parameter called colour difference index (CDI) has been postulated earlier which indicates the combined effects of different known individual colour difference parameters between any two samples when dyed with varying shade under different conditions of dyeing. Hence, this index has also been used in the present work to understand the combined effects of different dyeing variables by a single parameter. For the application of same concentration of dye between two sets of dyeing under varying condition, only the magnitudes of the respective $\Delta E$, $\Delta C$, $\Delta H$ and MI values (irrespective of their sign and direction) may be considered to calculate CDI values using the following empirical relationship:

$$\text{Colour difference index (CDI)} = [((\Delta E \times \Delta H)/ (\Delta C \times MI))]$$

2.2.8 Evaluation of Colour Fastness

Colour fastness to washing of the dyed fabric samples was determined as per IS: 764-1984 method using a Sasmira launder-O-meter following ISO-3 wash fastness evaluation method. The wash fastness rating was assessed using grey scale as per ISO -05-AO2 (loss of shade depth) and ISO –105-AO3 (extent of staining) and the same was cross-checked by measuring the loss of depth of colour and staining using Macbeth 2020 plus computer aided colour measurement system attached with relevant software.

Colour fastness to rubbing (dry and wet) was assessed as per IS: 766-1984 method using a motorized semi-automatic digital crockmeter and using grey scale as per ISO-105-AO3 (extent of staining).

Colour fastness to exposure to UV light was determined as per IS: 2454-1984 method. The half of the samples (10cm × 1cm) was exposed to UV light in a Shirley MBTF Microscal fade-O-meter (having 500 watt Philips mercury bulb with tungsten filament lamp simulating natural day light) along with the eight (1-grade) blue wool standards (BS 1006: BOI: 1978). The fading of each sample was observed and compared...
against the fading of blue wool standards (1–8) to assess the relative rating of light fastness grade from 1 to 8.

3 Results and Discussion

3.1 Effect of Different Mordants on Mechanical Properties

Table 2 shows that for the treatment with higher percentage of mordant above 20% concentration, either in single or double mordanting system, there is a noticeable loss in tensile strength. Amongst harda, potassium aluminium sulphate (alum) and aluminium sulphate single mordanting at comparable dosages, there is relatively higher strength loss in case of aluminium sulphate. However, for harda treatment followed by aluminium sulphate treatment (upto 10-20%), the strength loss is not too high, being acceptable.

3.2 Effect of Different Mordants on Colour Yield and Colour Fastness

Table 3 shows the effect of different single and double mordants on surface colour strength (K/S value), colour difference (ΔE) and CV% of K/S value (uniformity of the shade) for jute fabric dyed with 30% (on weight of solid source material of tesu powder) tesu extract. Amongst all the single mordants and two types of double pre-mordanting systems studied, the mordanting with 20% harda + 20% aluminium sulphate shows the maximum surface colour depth (K/S). This mordant has already been found best suited for dyeing of jute and chemically modified jute with red sandal wood and jackfruit wood.

Table 4 shows the effects of mordants on colour fastness behaviour. There are almost no differences in colour fastness to washing, light and rubbing, irrespective of per cent application of three mordants used. However, there are some clear differences in colour fastness ratings when atleast 20% harda and 20% aluminium sulphate double pre-mordanting system is used. Between the two types of double pre-mordanting systems, 20% harda followed by 20% aluminium sulphate shows better colour fastness rating.

Dye uniformity (CV% of K/S values) is found to be below 5% (Table 3) which is acceptable as per earlier report. This may be due to possible synergistic effect of these two types of mordanting agents applied, which perhaps facilitate more strong complex formation among the fibre-mordants-dye system in presence of both myrobolan (harda) containing chebulinic acid residue and aluminium sulphate containing mordantable metal ions acting in combination.

3.3 Effect of Dyeing Process Variables

Effect of different dyeing process variables have been studied to optimize the dyeing conditions for maximum and uniform colour yield for 20% harda + 20% Al2(SO4)3 mordanted and dyed fabrics. The results are shown in Table 5. It is found that when all other variables are kept fixed, with the increase in time of dyeing (45–120 min), K/S value slowly increases up to 60 min and then starts decreasing on further increase in dyeing time. This may be explained by the possibility of achieving dyeing equilibrium at 60 min, depending on the rate of dye diffusion (being slowest step among transportation, absorption, diffusion, and fixation of colourant molecule from tesu extract). There may be some desorption / breaking of dye-fibre-mordant complex which leads to the dropping trend above 60 min of dyeing time.

On increasing the dyeing temperature (60°-100°C), the surface colour strength (K/S values) increases measurably from 60°C to 90°C and then drops down. Increase in temperature of dyeing inevitably supplies
more energy for transportation of dye molecule thus facilitating the higher rate of dye sorption and diffusion up to 90°C before the desorption starts at relatively higher temperature (100°C). Hence, it is found that at 90°C there is maximum dyes absorption from tesu extract, giving highest $K/S$ values. However, for dyeing at 100°C for 60 min or more the dyes possibly start showing desorption and the colour yield is reduced.

<table>
<thead>
<tr>
<th>Mordant and conc.</th>
<th>Dyeing Shade</th>
<th>$K/S$ value at $\lambda_{max}$</th>
<th>$\Delta E$</th>
<th>CV% of $K/S$ value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nil (control)</td>
<td>a White</td>
<td>0.81</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>b Light creamish orange</td>
<td>2.82</td>
<td>2.77</td>
<td>4.57</td>
</tr>
<tr>
<td>Harda</td>
<td>10% a Lt cream colour with green tone</td>
<td>3.12</td>
<td>3.36</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>b Light ocher brown</td>
<td>4.88</td>
<td>6.74</td>
<td>4.64</td>
</tr>
<tr>
<td></td>
<td>20% a Cream colour with green tone</td>
<td>3.23</td>
<td>5.29</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>b Ocher brown</td>
<td>5.12</td>
<td>6.92</td>
<td>4.21</td>
</tr>
<tr>
<td></td>
<td>30% a Greenish cream</td>
<td>3.65</td>
<td>4.63</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>b Ocher brown</td>
<td>5.68</td>
<td>7.13</td>
<td>4.78</td>
</tr>
<tr>
<td></td>
<td>40% a Greenish cream</td>
<td>3.71</td>
<td>5.04</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>b Ocher brown</td>
<td>5.96</td>
<td>8.72</td>
<td>4.92</td>
</tr>
<tr>
<td>KAl(SO$_4$)$_2$</td>
<td>10% a Almost no change (White)</td>
<td>0.85</td>
<td>4.08</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>b Light yellowish orange</td>
<td>2.84</td>
<td>5.86</td>
<td>4.12</td>
</tr>
<tr>
<td></td>
<td>20% a Almost no change (White)</td>
<td>0.88</td>
<td>4.03</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>b Yellowish orange</td>
<td>2.91</td>
<td>6.12</td>
<td>4.86</td>
</tr>
<tr>
<td></td>
<td>30% a Almost no change (White)</td>
<td>1.02</td>
<td>4.97</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>b Yellowish orange</td>
<td>2.98</td>
<td>6.95</td>
<td>4.33</td>
</tr>
<tr>
<td></td>
<td>40% a Almost no change (White)</td>
<td>1.08</td>
<td>3.89</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>b Deep yellowish orange</td>
<td>3.43</td>
<td>7.42</td>
<td>5.11</td>
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<tr>
<td>Al$_2$(SO$_4$)$_3$</td>
<td>10% a Almost no change (White)</td>
<td>0.82</td>
<td>4.31</td>
<td>-</td>
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<tr>
<td></td>
<td>b Light creamish orange</td>
<td>2.91</td>
<td>6.38</td>
<td>4.22</td>
</tr>
<tr>
<td></td>
<td>20% a Almost no change (White)</td>
<td>0.85</td>
<td>4.57</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>b Creamish orange</td>
<td>3.00</td>
<td>6.67</td>
<td>4.37</td>
</tr>
<tr>
<td></td>
<td>30% a Almost no change (White)</td>
<td>0.87</td>
<td>4.02</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>b Creamish orange</td>
<td>3.06</td>
<td>6.21</td>
<td>4.59</td>
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<tr>
<td></td>
<td>40% a Almost no change (White)</td>
<td>0.92</td>
<td>4.94</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>b Deep creamish orange</td>
<td>3.54</td>
<td>7.56</td>
<td>4.91</td>
</tr>
<tr>
<td>Harda + KAl(SO$_4$)$_2$</td>
<td>10% + 10% a Light yellowish green</td>
<td>3.79</td>
<td>5.83</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>b Light ocher brown</td>
<td>5.21</td>
<td>7.28</td>
<td>4.78</td>
</tr>
<tr>
<td></td>
<td>20% + 20% a Yellowish green</td>
<td>3.84</td>
<td>5.76</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>b Ocher brown</td>
<td>7.28</td>
<td>8.59</td>
<td>5.87</td>
</tr>
<tr>
<td></td>
<td>30% + 30% a Yellowish green</td>
<td>3.88</td>
<td>6.81</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>b Deep ocher brown</td>
<td>7.05</td>
<td>8.67</td>
<td>5.24</td>
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<tr>
<td></td>
<td>40% + 40% a Yellowish green</td>
<td>3.96</td>
<td>5.84</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>b Deep ocher brown</td>
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<td>9.80</td>
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<td>Harda + Al$_2$(SO$_4$)$_3$</td>
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<td>3.86</td>
<td>5.72</td>
<td>-</td>
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<tr>
<td></td>
<td>b Light reddish brown</td>
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<td>7.35</td>
<td>4.14</td>
</tr>
<tr>
<td></td>
<td>20% + 20% a Light yellowish green</td>
<td>3.95</td>
<td>5.79</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>b Reddish brown</td>
<td>7.71</td>
<td>10.73</td>
<td>4.87</td>
</tr>
<tr>
<td></td>
<td>30% + 30% a Yellowish green</td>
<td>3.99</td>
<td>5.77</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>b Deep reddish brown</td>
<td>6.02</td>
<td>9.75</td>
<td>4.58</td>
</tr>
<tr>
<td></td>
<td>40% + 40% a Yellowish green</td>
<td>3.93</td>
<td>6.47</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>b Deep reddish brown</td>
<td>8.66</td>
<td>11.74</td>
<td>5.99</td>
</tr>
</tbody>
</table>

a—Without dyeing and b—Dyeing with 30% tesu extract at pH 11, $\Delta E$ – Total colour difference.
Table 5 also shows varying degree of dye uptake and corresponding up and down (crests and troughs) trends for $K/S$ value with the variation in $pH$ values from 4 to 12. With the increase in $pH$ from 4 to 6, an initial dropping trend is observed and further increase in $pH$ from 6 to 10 shows the increase in $K/S$ value up to $pH$ 10. However, on further increase in $pH$ from 10, the $K/S$ value starts decreasing. So, $pH$ 10 may be considered as optimum value. But the dye uniformity and colour fastness properties (Table 6) are found to be relatively much better at $pH$ 11. Hence $pH$ 11, though the $K/S$ value is to some extent lower than that obtained at $pH$ 10, is preferred and considered to be a better optimum condition of dyeing for the overall balanced results.

Keeping other variables constant, with the variation in material-to-liquor ratio (MLR) from 1:10 to 1:50 (Table 5), initially the $K/S$ value sharply increases...
upto MLR of 1:20 and then there is a slow increase upto MLR 1:30, after which the value drops down with the further increase in MLR. Hence, the optimum MLR is considered to be either 1:20 or 1:30. However, the difference in $K/S$ values observed between MLR 1:20 and MLR 1:30 is marginal, and the cost of dyeing is found to be much lower for MLR 1:20 and much higher for 1:30. Hence, MLR 1:20 gives a better balance between colour yield and cost of dyeing, considering it as optimum dyeing conditions.

Table 5 shows varying degree of up and down trends (crests and troughs) in $K/S$ values with the increase in mordant concentrations from 10% to 20% which is followed by a subsequent decreasing and rising trends up to 40% each of myrobolan and Al$_2$(SO$_4$)$_3$. Thus, considering the dyeing cost and colour yield, the optimum mordant concentration may be considered as 20%; though application of 40% each of the two double mordants gives little bit higher colour yield and $K/S$ values, but it assumes higher dye non-uniformity showing CV% of $K/S$ value nearly 6 (Table 3). Moreover, there is noticeable loss in fabric tenacity (Table 2) for mordanting with more than 20% of each of the said double mordanting agents. So, for this double pre-mordanting system, the optimum concentration of mordants should be 20% for both the first (harda) and second (aluminium sulphate) mordants. This may be attributed to the possible reduction in activation energy required for the absorption and fixation of tesu as natural dye on fibre surface by forming a dye-fibre-mordant complex. Above this concentration, the excess amount of these two mordants do not work favourable until their concentration reaches upto 40%, facilitating the formation of more amount of such complex.

There is a slow and sharp increase in $K/S$ value with the increase in dye conc. upto 30% (on the basis of weight % of dried solid tesu flower petals), above which it almost levels off reaching probably the first saturation level at around 30% dye. Hence, 30% dye conc. is taken as the optimum value.

Also the optimum concentration of common salt for tesu dyed jute fabrics is 10 g/L, as the $K/S$ values are found to be maximum at this salt conc. The addition of an electrolyte (common salt) to the dyeing liquor expectedly increases the exhaustion of the dye. Common salt is dissolved completely in the aqueous tesu extract are: 60 min dyeing time, 90°C temperature, 1:20 MLR, 11 pH, 20% mordant conc., 30% dye conc. (weight percentage of dried colour source material) and 10 g/L common salt conc.

3.4 Colour Strength and Related Colour Interaction Parameters

Table 5 also shows the effects of different process variables on $K/S$ values along with other colour interaction parameters, including total colour difference

<table>
<thead>
<tr>
<th>Dyeing conditions</th>
<th>$K/S$ at $\lambda_{max}$</th>
<th>$\Delta E$</th>
<th>$\Delta L$</th>
<th>$\Delta a$</th>
<th>$\Delta b$</th>
<th>$\Delta C$</th>
<th>$\Delta H$</th>
<th>BI</th>
<th>MI</th>
<th>CDI</th>
<th>CDI$<em>{max}$-CDI$</em>{min}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dye conc., %</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>10</td>
<td>4.56</td>
<td>9.63</td>
<td>3.46</td>
<td>3.51</td>
<td>8.27</td>
<td>4.58</td>
<td>15.2</td>
<td></td>
<td></td>
<td></td>
<td>3.47</td>
</tr>
<tr>
<td>20</td>
<td>5.00</td>
<td>14.97</td>
<td>5.48</td>
<td>5.09</td>
<td>12.97</td>
<td>13.49</td>
<td>11.15</td>
<td>5.24</td>
<td></td>
<td></td>
<td>0.73</td>
</tr>
<tr>
<td>30</td>
<td>7.63</td>
<td>14.98</td>
<td>6.82</td>
<td>3.43</td>
<td>12.89</td>
<td>13.21</td>
<td>1.86</td>
<td>10.31</td>
<td>4.92</td>
<td>0.43</td>
<td></td>
</tr>
<tr>
<td>40</td>
<td>5.96</td>
<td>16.71</td>
<td>6.17</td>
<td>5.01</td>
<td>14.7</td>
<td>15.28</td>
<td>2.76</td>
<td>8.42</td>
<td>5.89</td>
<td>0.51</td>
<td>0.44</td>
</tr>
<tr>
<td>50</td>
<td>6.03</td>
<td>15.57</td>
<td>6.89</td>
<td>3.58</td>
<td>13.49</td>
<td>13.82</td>
<td>1.92</td>
<td>9.51</td>
<td>5.12</td>
<td>0.42</td>
<td></td>
</tr>
<tr>
<td>60</td>
<td>7.17</td>
<td>17.18</td>
<td>5.94</td>
<td>6.04</td>
<td>14.94</td>
<td>15.75</td>
<td>-3.40</td>
<td>6.35</td>
<td>6.71</td>
<td>0.55</td>
<td></td>
</tr>
<tr>
<td>Salt conc., g/L</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>5.39</td>
<td>10.16</td>
<td>4.88</td>
<td>1.84</td>
<td>8.72</td>
<td>8.80</td>
<td>-1.42</td>
<td>11.06</td>
<td>3.21</td>
<td>0.51</td>
<td></td>
</tr>
<tr>
<td>10</td>
<td>7.63</td>
<td>13.62</td>
<td>5.08</td>
<td>4.58</td>
<td>11.77</td>
<td>12.21</td>
<td>-3.24</td>
<td>7.12</td>
<td>4.98</td>
<td>0.72</td>
<td>0.36</td>
</tr>
<tr>
<td>15</td>
<td>5.80</td>
<td>11.64</td>
<td>5.58</td>
<td>1.66</td>
<td>10.08</td>
<td>10.14</td>
<td>-1.26</td>
<td>10.19</td>
<td>3.48</td>
<td>0.41</td>
<td></td>
</tr>
<tr>
<td>20</td>
<td>5.46</td>
<td>9.62</td>
<td>5.35</td>
<td>1.16</td>
<td>7.91</td>
<td>7.94</td>
<td>-0.93</td>
<td>11.86</td>
<td>3.08</td>
<td>0.36</td>
<td></td>
</tr>
</tbody>
</table>

*Bleached jute fabric undyed and ^ Only mordanted undyed; $\Delta E$ – Total colour difference, $\Delta L$ – lightness/darkness difference, $\Delta a$ – greenness/redness difference, $\Delta b$ – blueness/yellowness, $\Delta H$ – change in hue, $\Delta C$ change in chroma, BI – brightness index, MI – metamerism index and CDI – colour difference index.
(ΔE), changes in hue (ΔH), changes in chroma (ΔC),
general metamerism index (MI), brightness index (BI)
and colour difference index (CDI) values. It is
interesting to observe that among the dyeing
conditions (time, temperature, pH, MLR, mordant,
dye conc. and salt conc.) varied, the most important
and predominating variables are identified as pH of
the dye bath as indicated by wide dispersion of CDI
values. Dispersion of CDI for the variation in pH
from 4 to 12 is 2.47, while for all other dyeing process
variables, the dispersion of CDI is << 0.50; for
variation of time and mordant concentration, it is
nearly 0.06-0.07; for variation in dye concentration it is
0.35-0.36, and for variation in dye temperature it is
0.44. The order of increasing CDI values therefore
appears to be as follows:

Mordant < Time < Temperature < MLR < Salt conc.</nearly 0.06-0.07; for variation in dye concentration it is
0.35-0.36, and for variation in dye temperature it is
0.44. The order of increasing CDI values therefore
appears to be as follows:

Mordant < Time < Temperature < MLR < Salt conc.
<< Dye conc. <<< pH
Therefore, for uniform dyeing using tesu extract for
jute or any other cellulosic textiles, special care are to
be taken for control of pH of the dye bath.
The data for ΔE, ΔL, Δa and Δb indicate the
differences in colour yield or surface colour strength
for varying dyeing condition in each case, as
compared to standard un-dyed pre-mordanted jute
fabric. The higher range of ΔE values (> 15) is
observed for the variation in pH and dye
concentration, indicating that these two are the major
controlling parameters responsible for uniform
dyeing.
Changes in hue for all the cases are found to be
negative (Table 5), indicating that there is no major
change in predominating hue, except showing some
hypsochromic shift in colour/tone. However, the
maximum negative ΔH value is observed in case of
the variation in pH from 10 to 11 which further
indicates the high pH sensitivity of colour yield for
this particular natural dye. Tesu contains butein
(3,4,2',4' tetra-hydroxy chalcone) of reddish yellow
colour having o-di-hydroxy and meta-di-hydroxy
phenol structure jointed together. Thus, due to
phenolic acidity of di-hydroxy phenolic structures in
the dye molecules of tesu, it is highly potent to
ionization in aqueous medium in presence of alkali.
Hence, in presence of alkali at pH 10-11, the
ionization of single dye molecules of tesu is presumed
to be maximum to give higher dye transportation,
absorption and diffusion rendering higher colour
yield.
Appreciable reduction in brightness index is
observed for tesu dyed jute fabric, irrespective of the
dyeing conditions used which is also reported earlier
by Samanta et al. for jute fabric dyed with most of
the natural dyes. However, interestingly it may be
noted that at lower pH of 4-8 and lower concentration
of the dye (10-30%), the reduction in brightness index
is much lower than that observed in other conditions.
Brightness index of dyed products depends on
reflectance value of dye and its orientation along the
fibre axis after fixation. Hence, it may be assumed
that the dye molecules of tesu being asymmetrical, the
single colourant molecules of tesu could not be
properly oriented along the fibre axis. Expectedly, the
reduction in brightness index is found to be higher
when the application of dye concentration is higher.
The general metamerism index indicates the
metameric effect on the tesu dyed jute product for
different conditions of dyeing. In all these cases, the
observed MI varies from 2.7 to 6.71 (Table 5) and
data are not much widely dispersed within a particular
condition being varied, but varies to a noticeable
degree from one condition to other, indicating its
potent metamerism from one varying condition to
other.

3.5 Colour Fastness
Table 6 shows the colour fastness data for washing,
rubbing and UV-light exposure for jute fabric dyed

<table>
<thead>
<tr>
<th>pH</th>
<th>Shade</th>
<th>K/S value</th>
<th>CV% of K/S value</th>
<th>Washing</th>
<th>Colour fastness to Light</th>
<th>Rubbing</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>None 2% CTAB 2% Cetrimide 2% Sandofix-HCF None 1% Benztriazole (None)</td>
<td>LOD ST LOD ST LOD ST LOD ST LOD ST LOD ST LOD ST LOD ST LOD ST LOD ST LOD ST LOD ST</td>
<td></td>
</tr>
<tr>
<td>10</td>
<td>Reddish brown</td>
<td>8.10</td>
<td>5.23</td>
<td>2 2-3        4 3-4        3-4 4-5        4 3-4        2 3 4 2</td>
<td></td>
<td></td>
</tr>
<tr>
<td>11</td>
<td>Reddish brown</td>
<td>7.47</td>
<td>4.87</td>
<td>2 3 3 4 4 4 4 4 5 4 4 2-3 3-4 4-5 3-4</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

CTAB – n-cetyl-trimethyl ammonium bromide, LOD – Loss of depth of shade, and ST – Staining of adjacent white bleached jute.
with aqueous extract of tesu as natural dye. The
colour fastness results in general indicate its moderate
colour fastness properties. Cetyl tri-methyl
ammonium bromide (CTAB), cetrimide and sandofix-
HCF, all being quaternary ammonium compound of
different molecular weights and sizes, act as known
cationic dye fixers to improve wash fastness
properties of mordantable anionic natural dyes.
Comparing the chemical structures of the myrobolan
containing chebulinic acid residue and the natural
dye, it is revealed that the constituent components of
these two natural materials (harda and tesu) contain
mordantable – OH groups and – C=O group in closer
position to each other in tesu, and – OH groups and –
COOH groups in harda (acidic in nature) can
participate easily in complex formation with metal
ions on one hand as well as free such groups may
form anions. At alkaline pH, tesu and myrobolan both
may form anions by abstraction of proton with the
help of – OH – ion from alkali. Thereafter, simply the
cationic dye fixers and dye anions of chebulinic acid
of myrobolan may combine to form precipitated
bigger size giant complex either inside the fibre or on
the surface of the dyed fibres to improve their wash
fastness properties.

Formation of complexes between dye, myrobolan
and cationic dye fixer cannot be ignored. Myrobolan
as the mordanting assistant, containing numerous
phenolic hydroxyl groups with few – COOH groups
in chebulinic acid residue, may thus form complexes
with cationic dye-fixers to improve the wash fastness.
However, it is also known that, in case of natural dyes
having few phenolic –OH and few –COOH groups in
it, there will be higher degree of improvement in wash
fastness than the natural dyes having no phenolic –
OH group and carboxylic group and this will be
predominant if the dye contains both of them in it17.
The colourant of tesu contain butein with phenolic –
OH group (both ortho and meta) as well as ketone
group linked CH=CH joining two di-hydroxy phenol
structure and myrobolan contains – COOH group in
it. Hence, the effect of such cationic dye fixing agent
is predominating for the combination of tesu and
harda in the present study.

Data in Table 6 also indicates that the colour
fastness to wash, light and rubbing for application of
selected natural dye at higher alkaline pH, with or
without treatment with dye fixing agents, is always
found to be better, because higher alkali concentration
(pH 11) causes better ionization or dye anion and
harda anion formation (preventing aggregation of dye
molecules) to participate in complex formation among
tesu, harda and dye fixers. This ionization capacity of
tesu also hastens the dye transportation as well as
absorption and diffusion of single dye molecules
throughout the surface of the fabric, showing better dye
uniformity expectedly at higher pH. It is reported
earlier that alkaline pH of the dye bath improves the
colour fastness of jute dyed with red sandal wood26,
and there is even changes in thermal transitions of
some natural dyes when subjected to heating
(equivalent to dye bath temperature or higher)27. Light
fastness of dyed textiles depends on constitution of
dyes28, UV-absorbance nature27 of dyes, fineness of
yarn, textile structure and even fibre type29. However,
both light fastness and wash fastness depend on nature
of pre-treatment (alkaline/acidic) on the fabric before
dyeing30.

Table 6 shows the light fastness of the sample
studied which can only be improved after the treatment
with 1% benztrizole by nearly one grade or so. The
overall poor light fastness observed for colour
component of tesu (butein) applied on jute is not only
due to the chemical structure of butein, but is partly
due to the susceptibility of jute fibre to known photo-
yellowing tendency on exposure to UV light and
consequent fading of the natural colour of the jute
fibre, which can also partly be protected or reduced by
the action of benzetriazole, as UV-absorber18,26. It is
observed from wavelength scan of UV-Vis spectrum of
tesu extract in aqueous medium (pH 6) (Fig. 1) that in
the UV region (190-380nm), there is hardly any
absorbance peak. On the contrary, in this zone it gives
negative absorbance and hence, there is no preferential
UV absorption by tesu colourant. Hence, light fastness
is not to be affected at all in UV-light exposure.

Wet and dry rub fastness of jute fabric dyed with
tesu (Table 6) are in between 3-4 (wet) and 4-5 (dry)
for dyeing at pH 11; and 3 (wet) and 4 (dry) for dyeing
at pH 10. This indicates that there is no need for the
special aftertreatment for its further improvement.
Medium to good rub fastness properties in all these
cases indicate that there are almost no superficial
unfixed natural dyes left on the fibre surface after
soaping and washing and expectedly this natural dye is
penetrated well inside the fibre voids and probably
might have been fixed well by coordinated complex
formation with the mordants and mordanting assistant
and also by hydrogen bonding with the help of –OH
and –COOH groups of jute fibre.
4 Conclusion

4.1 The optimized conditions for aqueous extraction of colour component from tesu are 60 min extraction time, 90°C temperature, 1:20 MLR and 11 pH.

4.2 The optimized conditions of dyeing of bleached jute substrates with tesu are: 60 min dyeing time; 90°C dyeing temperature; 1:20 MLR; 11 pH; 20% (owf) mordant concentration for each of myrobolan (harda) and aluminium sulphate and 10 gpl common salt conc. for this fibre-mordants-dye systems studied.

4.3 Interpretation from colour difference index values clearly reveals that pH of dyebath and dye concentrations are the two most important dyeing process variables amongst all other parameters of dyeing process variables in this case.

4.4 Wash fastness rating is improved by nearly one unit higher on specific post-treatment with quaternary ammonium compounds as dye fixer, while light fastness can be improved by one unit higher by the benzetriazole post-treatment under the specific condition.

4.5 Dyeing carried out at pH 11 shows better dye uniformity and wash fastness than that obtained at pH 10, though the observed colour strength at pH 10 is maximum for the specific double pre-mordanted jute fabric dyed with tesu, indicating its pH sensitivity on colour yield and colour fastness properties.

Acknowledgement

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