Eco-dyeing of silk with *Tridaxprocumbens* using natural mordants.

Sudhakar R
Apparel Technology & Management, Bangalore University, Bangalore - 560 001, India.

Abstract: In the present study, natural dye was extracted from the dried leaves of *Tridaxprocumbens* and was assessed for its dyeing and colourfastness properties on silk. The process conditions for the dye extraction and dyeing of silk was optimized. Gall nut (*Quercus infectoria*), dried fruits of amla (*Phyllanthusemblica*), dried fruits of bahera (*Terminaliabellerica*), myrobalan (*Terminalia chebula*), tamarind seed coat (*Tamarindus indica*) and laboratory grade tannic acid were used as natural mordants. The influence of potash alum as a mordant along with natural mordants on colour strength and colour fastness was also assessed. The shades of all the dyed samples were observed to be in the yellow-red quadrant of the colour space diagram. The relative colour strength of all the mordant dyed samples increased considerably indicating a positive influence of the mordants on the colour strength of the dyed samples. Combination of natural mordant with potash alum resulted in slightly increased K/S values as compared to natural mordant alone indicating better colour strength. All the dyed samples exhibited good fastness ratings with respect to washing, rubbing and perspiration irrespective of the mordants used. However the light fastness was found to be considerably influenced by the mordants, myrobalan showing better light fastness as compared to other mordants and unmordanted sample. The present study demonstrates the potential of *Tridaxprocumbens* as a natural dye to obtain different shades on silk with acceptable colour fastness.

Key words:Colour fastness, Eri silk, Mordants, Natural dye, *Tridaxprocumbens*,

I. INTRODUCTION

Natural dyes are colorants that are obtained from natural sources without chemical processing. For thousands of years human beings have used natural colours for a variety of purposes. With the advent of synthetic colorants, the use of natural colorants saw a drastic decline, the main reasons being that, with synthetic colorants, it was possible to obtain a wider range of colours and better colour fastness at a reasonable price [1]. However, in recent years there is an increasing demand for natural colorants because of the rising interest in preserving ecosystems, attributable to the fact that they are more eco-friendly and are more quickly biodegraded than the synthetic ones [2]. Studies conducted on the colour characteristics and fastness properties of natural dyes show that the colours obtained are soft and varied, and afew of the dyes have wash fastness ratings similar to those of acid dyes on wool and silk [3-7].In spite of their inferior fastness, natural dyes are more acceptable to environmentally conscious people around the world [8].However natural dyeingposes some problems such as low dye exhaustion and poor fastness. In order to overcome these, most of the dyeing processes are carried out by adding metal-based mordants [9]. These mordants comprise heavy metal ions such as copper, iron, chromium etc., which remain unexhausted in the residual dyebath causing environmental problems [10-11].Over the past few years natural biomordants viz., *Acacia catechu* [9],*Tamarindusindica* seed coat [12], *Emblicaofficinalis* dried fruit [13], *Terminaliachebula*[14],rind of *Punicagranatum*[15], galls of *Quercusinfectoria*[16]have been employed for dyeing various textile substrates in lieu of harmful metallic mordants. As there is a growing need for the non-toxic method of coloration for health sensitive applications such as children's garments as well as intimate clothing, an attempt is made in the present study to utilize natural mordants viz., gall nut, amla, bahera, myrobalan, tamarind seed coat and tannic acid in place of metallic mordantsfor dyeing silk with *Tridaxprocumbens*.

*Tridaxprocumbens*Linn (compositae) is a common grass found in tropical areas of all countries, growing primarily during rainy season. It is distributed throughout lndiaupto 2400 m above sealevel and in all hot countries. It is a common weed in India, present along with economically important crops.The leaves are also cooked as a vegetable and they are also eaten by cattle. It habitats waste places, road sides and hedges throughout India [17]. It is commonly used in Indian traditional medicine as anticoagulant, antifungal and in dysentery [18].The leaf juice possesses antiseptic activity and it is also used to treat cuts, burns and wounds. Apigenin (fig. 1) andquercetin(fig. 2) are the major flavonoids reported to be present in *Tridax*procumbens [19-20].

The use of natural dyes would be more beneficial with regard to environmental impact and sustainability, especially if dyes are extracted from renewable sources such as leaves as in the present case.

The present study was aimed to establish the extraction and dyeing process for silk fabrics using the leaves of *Tridaxprocumbens*, as well as to determine the influence of natural mordants on the fastness properties and colour values with the aim of reducing the toxic effects of synthetic mordantson the environment.

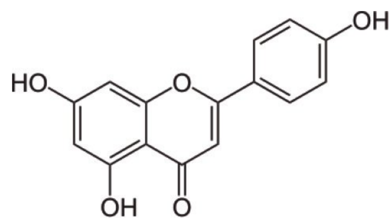


Figure 1.Apigenin

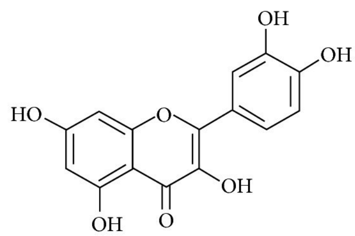


Figure 2.Quercetin

II. EXPERIMENTAL WORK:

2.1 Fabric: Degummed eri silk fabric with the following specifications was used for dyeing- ends / cm- 32; picks / cm- 28; and weight in grams / m^2-90.

2.2 Chemicals and Auxiliaries: Sodium carbonate and acetic acid were used for the maintenance of solution pH.Potassium aluminiumsulphatewas used in combination with natural mordants. All the chemicals used were of laboratory grade. Non-ionic detergent was used for soaping after dyeing.

*2.3 Dye:*The fresh leaves of *Tridaxprocumbens*were collected from the field, washed under running water and dried at room temperature. The dried leaves were used for extraction of the dye.

*2.4 Extraction of dyes:*The dried leaves were extracted in aqueous mediumby boiling 2 g of the material separately for 60 minutes at material to liquor ratio 1:50 with pH of the solution being maintained from 5 to 10. The dye extraction giving the maximum K/S value on silk was considered as optimum pH for extraction.

The extraction time of the dye was optimized by boiling 2 g of the material separately for 30, 45, 60, 90, 120, and 150 minutes, keeping material to liquor ratio 1:50. The dye extraction giving the maximum K/S value on silk was considered as optimum time of extraction.

For the determination of the quantity of colouring matter, 10 grams of the material was extracted at optimum conditions and the resultant solution was filtered, evaporated till dry and the solid material obtained was weighed. The total colouring matter obtained was 15% on the dry weight. The crude colouring matter obtained was used for dyeing without further purification.

*2.5 Natural Mordants and mordanting:*Gall nut (*Quercusinfectoria*), dried fruits of Amla (*Phyllanthusemblica*), dried fruits of Bahera (*Terminaliabellerica*), Myrobalan (*Terminalia chebula*), Tamarind seed coat (*Tamarindus indica*) and laboratory gradeTannic acid were used as natural mordants.The water extracts without further purification were used for mordanting the silk fabric.

Two sets of pre-mordanting was carried out in an aqueous solution of the natural mordant at 4% concentration for 30 minutes at 70^0C (liquor ratio 1:30). One set of premordanted samples were again treated with 4% Alum for 30 minutes at 70^0C (liquor ratio 1:30). All the mordanted fabrics were then separately dyed.

*2.6 Optimizing Dyeing pH and Temperature:*Silk fabric samples were dyed at varying pH conditions (pH 4 to pH 10) with equal concentrations (5%) of the extracts at 90^0C for 30 minutes to examine the effect of pH on dyeing and the K/S values of the samples were determined. Silk fabric samples were dyed with equal concentrations (5%) of the extracts at range of temperatures 40-90^{0}C for 30 minutes and then the K/S values of the dyed samples were measured to examine effect of temperature on dyeing.

*2.7 Dyeing Process:*Dyeing was carried out in an open bath beaker dyeing machine equipped with programmable control of temperature and time. The silk fabric was entered into the dye bath containing 5% dye solution (owm), pH 5 at 40^{0}C and the temperature was gradually raised to 90^{0}C in 15 minutes (liquor ratio 1:40) and the dyeing was continued for 45 minutes. At the end of dyeing, the dyed samples were rinsed, soaped with non-ionic soap at 60^{0}C, rinsed with water, squeezed and dried.

*2.8 Colour Measurement:*The reflectance of the dyed samples were measured using a Gretag Macbeth Color eye 7000A spectrophotometer interfaced to a personal computer, under illuminant D_{65} using 10^0 standard observer with specular component excluded and UV-component included, from which the corresponding K/S values and CIE L a* b* C and h co-ordinates were calculated at the appropriate λ_{max} of each sample. Each fabric sample was folded twice to realize a total of four thicknesses of the fabric. Four readings were made on each sample at different sites and the results were averaged.

The colours are given in CIE Lab coordinates, L corresponding to the lightness (100 = White, 0 = Black), a* to the red-green coordinate (positive sign=red, negative sign=green) and b* to the yellow-blue co-ordinate (positive sign = yellow, negative sign = blue). C is a measure of chroma (saturation) and represents the distance from the neutral axis. h is a measure of hue and is represented as an angle ranging from 0 degrees to 360 degrees. Angles that range from 0 to 90 degrees are reds, oranges and yellows. 90 to 180 degrees are yellows, yellow-greens and greens. 180 to 270 degrees are greens, cyan and blue. 270 to 360 degrees are blue, purples, magentas and return to reds.

KubelkaMunk(K/S) function is given by:

$$K / S = \frac{(1 - R)^2}{2R}$$

Where *K* is the absorption coefficient, *S* is the scattering coefficient for a colourant at a specific wavelength and *R* is the reflectance at complete opacity. The K/S value at $\lambda_{max}$is directly proportional to theconcentration of dye on the substrate.

The colour difference and relative colour strength between the mordant dyed samples and unmordanted dyed sampleswere also obtained using following relationships.

Colour difference (ΔE)= $\sqrt{(\Delta L)^2 + (\Delta a)^2 + (\ldots}$

Relative Colour strength (%)= $\frac{K/S \text{ of mordanted sample}}{K/S \text{ of unmordanted sample}}\ x$

2.9 ColourFastness determination[21]*:*Colour fastness to washing was assessed as per AATCC Test Method 61-1996 Test No. 2A. Evaluation of colour fastness to crocking was accomplished by AATCC Test Method 8-1996 using a Crockmeter. Colour fastness to perspiration was determined using AATCC Test Method 15-1997 using a Perspirometer. Colour fastness to light was evaluated as per AATCC Test Method 16-2004 option 5.

III. RESULTS AND DISCUSSION

3.1 Dye extraction: Extracts obtained under varying conditions of pH and duration was used for dyeing silk fabrics separately and the colour strength (K/S) of the dyed samples was measured. Fig. 3 shows the effect of pH used for extraction on the colour strength of the samples. Maximum colour strength was obtained for the samples dyed with the extract obtained at pH 7. Fig. 4 shows the influence of time on extraction on the colour strength of the samples. Darker shades were obtained for the sample dyed with the extract obtained at one hour duration.

3.2 Dyeing: The colouring matter extracted at optimum conditions was used for dyeing silk. Fig. 5 shows the effect of pH of the dye bath on the dye uptake expressed as colour strength (K/S value). Maximum colour strength was obtained when the dye bath was maintained at pH 5. Increase in pH of the dyebath resulted in a gradual decrease of the colour strength. At higher pH values the conjugate structure of the natural dyes might be destroyed because of oxidation, thus resulting in lower colour strength of the dyed silk silk fabric [22]. Fig. 6 shows the plot of colour strength against the dye bath temperature. Higher K/S value was observed for the sample dyed at 90^0C indicating better dye uptake under these conditions.

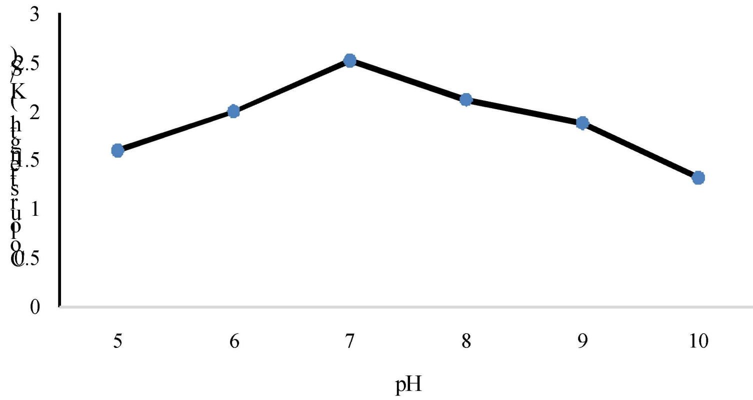


Figure 3. Effect of pH on dye Extraction

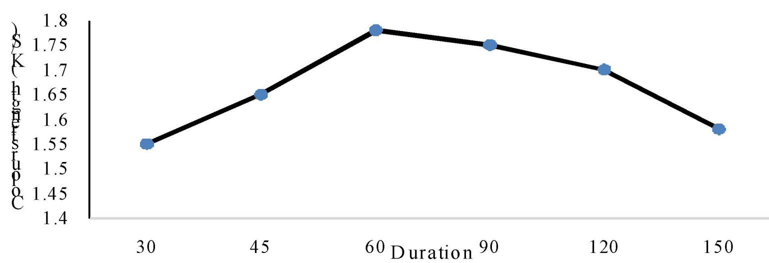


Figure 4. Effect of duration on dye Extraction

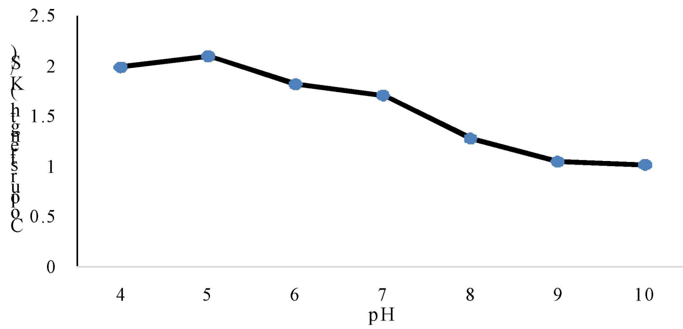


Figure 5.Effect of dye bath pH on colour strength on silk fabric.

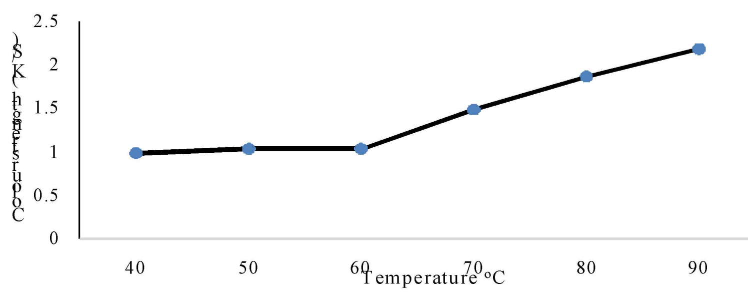
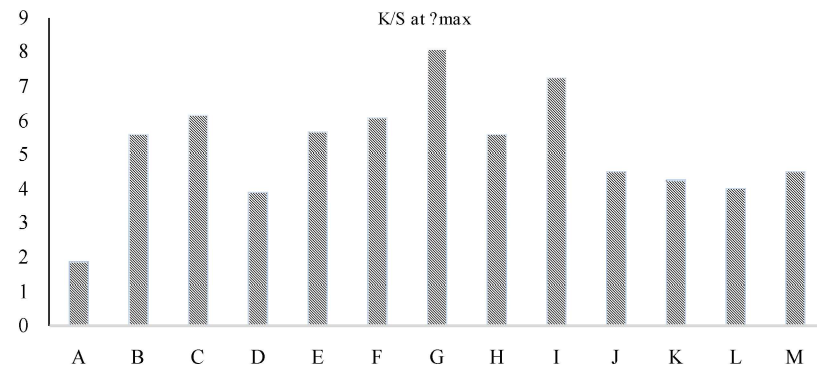
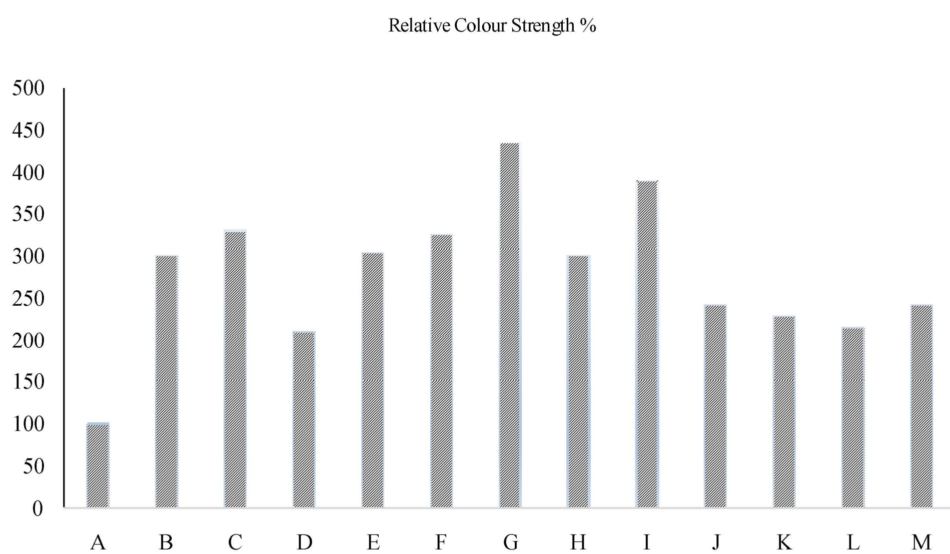


Figure 6. Effect of dyebath temperature on colour strength of silk fabric



A=Unmordanted, B= Gallnut 10%, C= Gallnut 10% + 4% Alum, D= Amla 10%, E= Amla 10% + 4% Alum, F= Bahera 10%, G= Bahera10% + 4% Alum, H= Myrobalan 10%, I=Myrobalan 10% + 4% Alum, J= Tamarind seed 10%, K= Tamarind seed10% + 4% Alum, L= Tannic acid 10%, M= Tannic acid 10% + 4% Alum

Figure 7. Effect of mordants on Colour strength of the dyed silk fabrics



A=Unmordanted, B= Gallnut 10%, C= Gallnut 10% + 4% Alum, D= Amla 10%, E= Amla 10% + 4% Alum, F= Bahera 10%, G= Bahera10% + 4% Alum, H= Myrobalan 10%, I=Myrobalan 10% + 4% Alum, J= Tamarind seed 10%, K= Tamarind seed10% + 4% Alum, L= Tannic acid 10%, M= Tannic acid 10% + 4% Alum

Figure 8. Effect of mordants on the Relative Colour strength of the dyed silk fabrics

*3.3 Colour co-ordinates:*Colour co-ordinates of silk samples dyed with *Tridaxprocum*bens extract are given in Table 1. The leaf extract of *T. procumbens* produced yellowish shades, with hue angles ranging between 55 and 86 for all the samples. It is observed that all the colour coordinates are positive with respect to red-green (a*), and yellow-blue (b*) and therefore, all ofthem lie in the yellow-red quadrant of the colour space diagram.Gallnut and Amla when combined with alum as mordants showed a shift in hue angles recording 16.96 and 17.45 respectively. All the mordanted samples showed a decrease in L values indicating darker shades. Combination of natural mordant with alum resulted in slightly increased L values as compared to natural mordant alone indicating a lighter shade but with improved K/S values. All the mordantedsamples showed considerable increase in colour strength.The mordant activity sequence was Bahera+Alum>Myrobalan+Alum>Gallnut+Alum>Bahera>Amla+Alum>Gallnut>Myrobalan>Tamarind seed coat>Tannic acid+Alum>Tamarind seed coat+Alum>Tannic acid>Alum>Unmordanted.

The relative colour strength of the dyed samples is shown in fig. 7. The relative colour strength of all the dyed samples exceeded 200%, the maximum being observed in case of sample mordanted with Bahera + alum (435%). Fig. 8 shows the colour difference of all the mordant dyed samples. The colour difference (ΔE) ranged from 6.08 to 17.73, the maximum recorded in samples mordanted with tamarind seed coat. Tamarind seed coat, Bahera and myrobalan recorded higher ΔE values compared to the other mordants used.

Table 1. Colorimetric data (L*, a*, b*, C*, h*, ΔE, K/S values) of unmordanted and mordanted dyed samples.

Mordant	L	a	b	C	h	ΔE	K/S at λ_{max}	Relative colour strength (%)
Unmordanted	66.39	0.86	11.24	11.28	85.59	-	1.86	100
Gallnut 10%	59.83	1.43	16	16.06	84.89	8.12	5.6	301
Gallnut 10% + 4% Alum	64.27	0.84	16.94	16.96	16.96	6.08	6.14	330
Amla 10%	61.11	1.82	15.24	15.35	83.16	6.69	3.90	210
Amla 10% + 4% Alum	61.46	2.03	17.33	17.33	17.45	7.92	5.66	304
Bahera 10%	55.93	1.85	22.16	22.24	85.21	15.15	6.07	326
Bahera10% + 4% Alum	55.21	2.1	24.7	24.8	85.1	17.54	8.09	435
Myrobalan 10%	56.75	1.2	19.81	19.84	86.48	12.90	5.6	301
Myrobalan 10% + 4% Alum	58.04	1.01	25.37	25.39	87.7	16.41	7.25	390
Tamarind seed 10%	51.18	9.61	13.8	16.8	55.1	17.73	4.5	242
Tamarind seed10% + 4% Alum	53	9.16	13.47	16.29	55.79	15.91	4.26	229
Tannic acid 10%	59.41	1.02	16.28	16.31	86.41	8.61	4	215
Tannic acid 10% + 4% Alum	60.19	1,51	15.69	15.77	84.48	7.65	4.5	242

Table 2.Washing fastness, rubbing fastness, perspiration fastness and light fastness of the dyed samples.

Mordant	**Washing**		**Rubbing**		**Perspiration**		**Light**
	CC	**CS**	**Dry**	**Wet**	**CC**	**CS**	**CC**
Unmordanted	4	4-5	4-5	4-5	4	4-5	2
Gallnut 10%	4	4-5	4-5	4	4	4-5	4
Gallnut 10% + 4% Alum	4	4-5	4-5	4	4	4-5	3
Amla 10%	4-5	4-5	5	4-5	4	4-5	3
Amla 10% + 4% Alum	4-5	4-5	5	4-5	4	4-5	4
Bahera 10%	4	4-5	4-5	4	4	4-5	4
Bahera10% + 4% Alum	4	4-5	4-5	4	4	4-5	4
Myrobalan 10%	4	4-5	4-5	4	4-5	4-5	5
Myrobalan 10% + 4% Alum	4	4-5	4-5	4	4-5	4-5	4
Tamarind seed 10%	4-5	4-5	5	4	4	4-5	3
Tamarind seed10% + 4% Alum	4	4-5	5	4	4-5	4-5	3
Tannic acid 10%+ Dye	4-5	4-5	4-5	4-5	4	4-5	4
Tannic acid 10% + 4% Alum	4-5	4-5	4-5	4-5	4	4-5	3

*3.4Colour fastness:*The colour fastness ratings of dyed silk fabrics with and without mordants are presented in Table 2. The washing fastness ratings of the unmordanted dyed sample were found to be 4 for change in colour and 4-5 for staining. The samples mordanted with Amla, tamarind seed coat and tannic acid showed slightly better fastness (4-5 and 4-5) compared to the other samples which recorded a rating of 4 and 4-5 for change in colour and staining respectively. Rubbing and perspiration fastness was also found to be very goodin all the cases except with a slight decrease (4) recorded for wet rubbing with gall nut, bahera, myrobalan and tamarind seed coat mordanted samples. However a considerable improvement was observed in light fastness of the mordanted samples as compared to control. Myrobalan was found to be superior with a rating of 5, whereas other mordants recorded 4 and 3 as compared to a rating 2 in case of unmordanted sample.

IV. CONCULSION

Eco-dyeing of eri silk with the extract of *Tridaxprocumbens* was accomplished with the use of natural mordants. All the bio-mordants used in the study resulted in considerable improvement in colour strength compared to unmordanted sample. Aluminium potassium suplhate when used along with the natural mordants showed further improvement in colour strength in all the cases except tamarind seed coat. All the samples showed acceptable colour fastness ratings.The findings of present work indicate that the leaves of *Tridaxprocumbens*, available abundantly *in the wild* can be a potential source of natural dye for silk. Further the use of biomordants instead of metal mordants on a fibre like eri silk makes the process more sustainable.

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