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Dyeing of silk using *Madhuca longifolia* as natural dye source

V Narayana Swamy[1,a], K N Ninge Gowda[2] & R Sudhakar[2]

[1]Department of Fashion and Apparel Design, The Oxford College of Science, Bangalore 560 102, India

[2]Department of Apparel Technology and Management, Bangalore University, Bangalore 560 001, India

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The dried leaves of *Madhuca longifolia* has been evaluated for their potential as a source for natural dyeing of silk. Dye has been extracted under optimum conditions such as extraction *p*H (10), time (60 min) and temperature (95°C). The extracted dye has been applied on the silk fabrics and a range of shades are obtained using different methods with or without using mordants. It is found that mordants have a significant effect on the color of dyed silk fabrics. The dyed samples have been evaluated for color measurements and standard wash, light and rub fastness tests. The extracted dye is also tested for some of the eco-parameters using atomic absorption spectrophotometry and GC/MS. The test results are compared with set standards to determine the eco-friendliness of natural dye. Their concentrations are found to be lower than the stipulated limits. The dyed samples are also tested for antimicrobial activity against Gram-positive and Gram-negative bacteria. The dyed silk fabrics show acceptable fastness properties and are found to possess antibacterial activity. The results show that *Madhuca longifolia* leaves are promising as a natural colorant, which would, in turn, pave the way for the discovery of a new range of environment-friendly dyes for textile materials.

Keywords: Antimicrobial activity, Mordant, Natural dye, *Madhuca longifolia*, Silk

1 Introduction

Crucial environmental issues are being debated all over the world. Although synthetic dyes are in abundance and a wide range of colors of remarkable fastness properties make their way into the market due to their eco-friendliness, natural dyes are gaining importance and need new natural sources to be explored[1-4]. There are many varieties of wild plant species available in the forests of this region which have the potential to be used as raw material for different forest -based industries[5-7]. Among different plant species, deciduous trees, shed their leaves at one time in a year and create vibrant hues associated with autumn. During each autumn season huge amounts of fallen leaves are available which would otherwise be waste material. *Madhuca longifolia* (Mahua) belongs to the family sapotaceae, commonly known as the butter nut tree; both wild and cultivated types are found predominantly in South India, but available throughout Indian forests. Flavonoids present in these leaves are myricetin, quercetin, β-carotene[8-10]. Leaves have medicinal properties[11-13]. The main objective of this study is the extraction of natural dye from *M. longifolia* waste and its application on silk fabric.

[a]Corresponding author.
E-mail: gvnnarayan@gmail.com

2 Materials and Methods

2.1 Raw Material

Fallen leaves of *Madhuca longifolia* were collected from 15-20 year old trees from Lalbagh, Bangalore. The identity of the plant specimen was confirmed by Lalbagh botanists.

2.2 Fabric

Plain woven, degummed, mulberry silk fabric weighing 40 g/m^2 with a yarn density of 519 ends/dm and 456 picks/dm was selected for dyeing. The fabric was washed in a solution containing 2 g/L non-ionic detergent solution and 1 g/L sodium carbonate at 60°C for 30 min, keeping the material-to-liquor ratio at 1:50. The scoured material was thoroughly washed with running water and dried at room temperature.

2.3 Extraction of Colorant

Dried leaves (500 g) were crushed and mixed with 10 L of water as solvent for dye extraction. The bath temperature was maintained at 90°C for 1h. After the extraction, lost volume was brought up and the extract was cooled down up to the room temperature, filtered to remove the insoluble residues and used for dyeing. The structures of flavonoids present in the leaves are shown in Fig. 1.

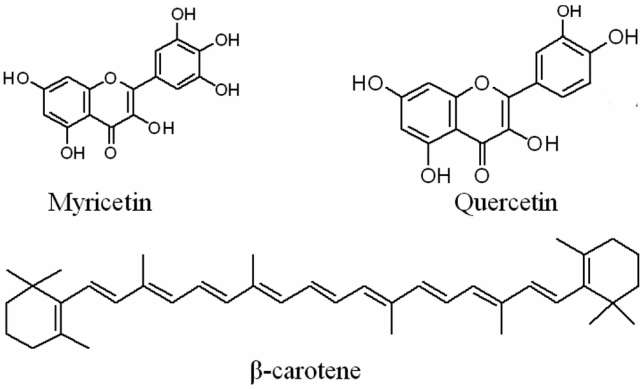

Fig. 1— Structures of Myricetin, Quercetin and β-carotene

2.3.1 Optimization of Extraction Conditions

The dye was extracted maintaining the optimized M : L ratio, temperature and time. Crushed leaves of 500 g and 10 L of distilled water were mixed in a container and heated for 60 min at 95°C from the time it starts boiling. The dye solution was then filtered and evaporated on a water bath to obtain the solid dye. The experiment was repeated three times and average dye yield was recorded[14,15].

The increase in absorbance of the extract was analyzed as a function of extraction time in the wavelength interval of 400-700 nm. For this measurement the extract was filtered through a filter paper and filtrate was diluted using 10 times the volume with distilled water. Absorbance was measured using Shimadzu 1601 PC UV/Vis spectrophotometer.

2.3.2 Effect of Temperature on Dye Stability

To study the effect of temperature on the stability of the dye, a dye bath containing 0.05 g/L dye was prepared from the stock solution (0.2%) in 20 mL water. The dye bath was heated continuously at high temperature in a high pressure beaker dyeing machine at 70 - 130 ºC for a period of 60 min at *p*H 4, 7 and 10. The absorption spectra were recorded before and after the treatment[16]. The percentage degradation of the dye was calculated using the following equation:

$$\text{Degradation } (\%) = \frac{A_i - A_f}{A_i} \times 100$$

where A_i is the area under the curve of the initial dye bath; and A_f, the area under the curve of the dye bath after treatment.

2.4 Dyeing of Silk Fabrics

The dyeing was carried out at 90°C for silk in a dye bath containing 10% dye (owf) at MLR 1: 40 in a beaker dyeing machine for 60 min. The dyed samples were subsequently washed in 5 gpL non-ionic detergent at 60°C for about 5 min and dried at room temperature.

2.4.1 Method of Mordanting

The substrates were pre- and post- mordanted using 2% and 5% (owf) solutions of each of potassium aluminum sulphate, tannic acid and tartaric acid which were employed with MLR ratio 1:20 for 30 min at 60°C. The fabrics were subsequently rinsed and dried.

2.5 Evaluation of Color Strength and Color Co-ordination

Dyed samples were evaluated by using CIELAB color co-ordinates with illuminant D65/10◦ observer on Gretag Macbeth Color Eye 7000 A Spectrophotometer. Four measurements were made for each sample and average results of the four measurements were recorded. The reflectance values over a range of 350–750 nm were recorded. The color strength measurement was based on the ratio of total light absorbed *K* and scattered *S* by the substrate as defined by the Kubelka Munk equation.

2.6 Fastness Testing

Wash fastness was assessed according to ISO 105 C02 using Launderometer. Rub fastness was performed according to ISO-X12 using crockmeter. Light fastness was assessed according to ISO 105 B02 using Xenotest light fastness apparatus.

2.7 Antibacterial Activity

Escherichia coli (E. coli), a Gram-negative bacterium, was selected due to its popularity of being used as a test organism and its resistance to common antimicrobial agents. *Staphyloccus aureus (S. aureus)*, a pathogenic Gram-positive bacterium, was used as it is a major cause of cross-infection in hospitals and the most frequently evaluated species. Antibacterial activity of the dyed samples was assessed using parallel streak method as per AATCC TM 147-2004.

2.8 Elemental Analysis of Dye

The bark samples were weighed to determine the fresh weight and dried in an oven at 95 ºC for 48 h to determine their dry weight. The dry samples were crushed in a mortar and the resulting powder was digested by weighing 0.5 g into an acid washed porcelain crucible and placed in a muffle furnace for 3 h at 500 ºC. The crucibles were removed from the furnace and cooled. Fifteen milliliter of 6 M HCl was added to the powder, then covered and heated on a

steam bath for 25 min. Another 1 mL of HNO_3 was added and evaporated to dryness by continuous heating for one hour. Finally, 5 mL of 6M HCl and 10 mL of water were added and the mixture was heated on a steam bath to complete dissolution. The mixture was cooled and filtered through a Whatman no. 541 filter paper into a 50 mL volumetric flask and the volume was made up to the mark with distilled water[17,18].

Heavy metal concentrations in the digested samples were determined using A6300 Shimadzu flame atomic absorption spectrophotometer with Shimadzu auto sampler (Asc-600). The calibration curves were prepared separately for all the metals by running different concentrations of standard solutions[19].

3 Results and Discussion

Based on the mean highest optical density values, the leaves material to water optimum ratio is determined as 1: 20 and extraction time is found to be 60 min at 95℃. The average dye yield is recorded to be 26.5% which appears as dark brown powder. The absorbance of an extract at the end of the 60 min extract process is shown in Fig. 2.

3.1 Effect of Temperature on Dye Stability

The thermal stability of the dye was studied by recording the absorbance spectra of the dye solution before and after heating. When the dye solutions at different *p*H (4, 7 and 10) are subjected to treatment with temperatures ranging from 70 ºC to 130 ºC, it is found that the dye is most stable under *p*H 4 at 70 ºC and 80 ºC. On the other hand, an appreciable decrease in absorbance is observed for the dye solutions having *p*H 7 and 10 at 80 ºC (Fig. 3).

At 130 ºC, there is almost 90% loss in colour of the dye solution at *p*H 4, and 95% and 100% loss in color at *p*H 7 and 10 respectively. This may be due to the decomposition of the dye molecule that results in colorless products at higher temperatures.

To ascertain that the dye has decomposed at a higher temperature and does not modify into a different chromophore, the maximum wavelength of absorption is determined after treating the dye solution at higher temperatures. Though the absorbance decreases with the increase in temperature, no new peaks are observed, confirming that the dye decomposes and does not convert into a different chromophore when exposed to higher temperatures.

3.2 Color Characteristics

Results with respect to color depth (*K*/*S* values) of dyeing of silk fabrics with *Madhuca longifolia* leaves (shade 10%) obtained with and without the use of mordants are given in Table 1. The results indicate that the *K/S* value of the silk fabrics increases with increasing mordant percentage. The process of dyeing with different mordants gives a shade change from pinkish to dark brown. Varied hues of color are obtained from both pre- and post-mordanting with alum, tannic and tartaric acid.

It can be seen from Table 1 that unmordanted dyed silk fabric shows lower dye uptake (*K*/*S* 1.25) compared to the other samples. This could be attributed to the affinity of mordant with colour and fabric[20,21]. Post -mordanted samples show maximum *K*/*S* values compared to pre-mordanted samples. The presence of OH and C=O in the tannin structure leads to the formation of metal salt-tannin complexes in the post -mordanted silk fabrics, which in turn, results in the higher color strength of the dye in the fabric[22]. The maximum *K*/*S* value of 7.01 is observed in post -mordanted tartaric acid at 5% concentration. This could be explained as due to more population

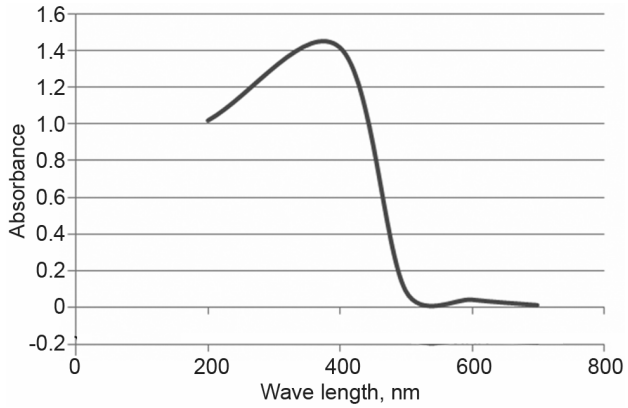

Fig. 2 — UV-Vis spectra of dye extract

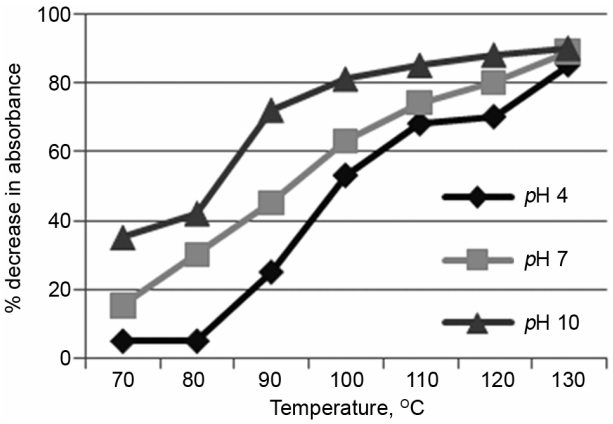

Fig. 3 — Effect of temperature and *p*H on dye yield

Table 1— The Calorimetric data of silk samples dyed with *Madhuca longifolia* leaves extract

Sample	Mordanting method	Mordant conc., %	*K/S*	L*	a*	b*	C	h
Undyed	-	-	-	90.42	2.15	0.11	2.71	365.52
Control	-	-	1.25	54.26	12.02	10.91	45.05	49.59
Mordanted								
Aluminium potassium								
sulphate (A)	Premordanting	2	5.01	48.45	11.96	12.32	45.24	63.98
		5	6.15	46.68	11.42	10.85	45.42	70.28
	Postmordanting	2	6.35	46.53	11.23	12.42	40.17	69.12
		5	6.51	45.44	11.11	12.82	39.95	70.10
Tannic acid (B)								
	Premordanting	2	4.81	48.68	11.12	10.85	45.83	63.58
		5	4.95	48.61	11.91	12.01	46.14	62.89
	Postmordanting	2	5.14	48.60	12.60	8.55	45.92	74.82
		5	5.50	46.78	11.43	10.58	45.28	69.35
Tartaric acid (C)								
	Premordanting	2	5.82	46.15	5.85	12.58	44.19	73.15
		5	6.11	48.64	11.56	12.41	46.18	63.98
	Postmordanting	2	6.14	45.01	9.19	5.92	40.82	69.25
		5	7.01	44.33	7.12	10.14	43.09	63.01

Table 2—Fastness properties for silk fabric dyed *with Madhuca longifolia* leaves extract

Sample	Mordanting method	Mordant conc., %	Light fastness	Wash fastness		Perspiration fastness				Rub fastness	
				CC	CS	Acidic		Alkali		Dry	Wet
						CC	CS	CC	CS		
Control	-	-	2	3	5	3	5	3	5	5	4
Mordanted											
Aluminium potassium											
sulphate (A)	Premordanting	2	3	3	5	4	5	4	5	5	4
		5	3	4	5	4	5	4	5	5	4-5
	Postmordanting	2	3	3	5	4	5	4-5	5	5	4-5
		5	2	3-4	5	4	5	4	5	5	4-5
Tannic acid (B)											
	Premordanting	2	3	2-3	5	3	5	4	5	5	4
		5	2	3	5	3-4	5	4	5	5	4
	Postmordanting	2	3	4	5	4	5	4-5	5	5	4
		5	3	4	5	4	5	4	5	5	4-5
Tartaric acid (C)											
	Premordanting	2	3	3	5	4	5	4	5	5	3
		5	3	4	5	4	5	4	5	5	3-4
	Postmordanting	2	3	4	5	4	5	4-5	5	5	4-5
		5	4	4	5	4	5	4	5	5	4-5

cc-Color change, cs-Color staining .

effect of mordant which possibly is bound to the maximum number of molecules of dye in the fabric[23].

The mordants not only cause a difference in hue color and significant changes in *K/S* values, but also in L* values. The data indicate that the maximum (44.33) dark colour is obtained by using post-mordanted tartaric acid at 5%. The sample without the use of mordants has the lightest color (54.26). Maximum (12.60) redness is found by using post - mordanted tannic acid at 2% and minimum (5.85) was found in pre-mordanted tartaric acid at 2%. The maximum (12.82) yellowness is obtained by using post-mordanted alum at 5%, whereas minimum (5.92) yellowness is obtained using tartaric acid post-mordanted at 2%.

3.3 Fastness Evaluation

Table 2 shows the fastness properties of the silk samples dyed with extracted dye. These results are

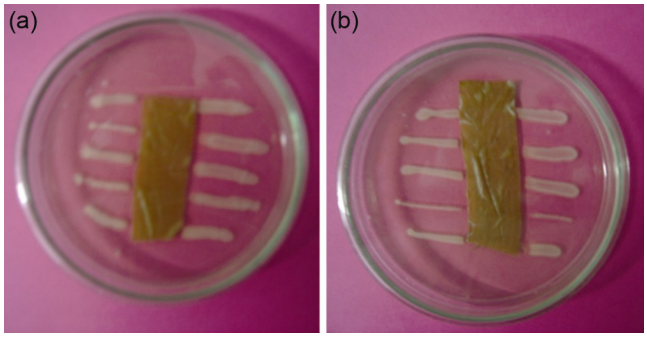

Fig. 4— Antibacterial activity of unmordanted silk fabrics on (a) *E. coli* and (b) *S. aureus*

assessed in the usual way in terms of the gray scale values for the staining of adjacent silk and cotton material. As can be seen, mordanted samples show improved fastness properties compared to unmordanted samples. Post -mordanted with tartaric acid and tannic acid samples show a good to very good rating to wash fastness in comparison to other samples. Similarly, post mordanted samples show moderate to good light fastness properties, the in proved light fastness properties of the dye can be attributed to the strong ionic bonding. This enhances the stability of the compound by a reduction in electron density at the chromophore. However, silk samples exhibit commercially acceptable grade (4-5) perspiration and rub fastness properties as a result of a good fixation of the dye in the fibre.

3.4 Antimicrobial Activity

The extract of *Madhuca longifolia* leaves has been screened for antimicrobial activities against selected bacteria (*E. coli* and *S. aureus*). The tested micro-organisms show a high degree of sensitivity. This is evident from the zone of clearance (Fig. 4). A minimum of 1-mm zone of inhibition is observed in all the cases. As tannin is known for inheriting general antimicrobial properties[24-25], the extract of *Madhuca longifolia* can be said to be antibacterial in nature.

3.5 Elemental Analysis of Dyed Samples

The metal content in dyed silk samples is shown in Table 3. Results show the extremely low quantities of heavy metals extracted from *Madhuca longifolia* leaves extract. As the concentrations are much below the stipulated limits the extracted dye can be considered eco-friendly.

Table 3—Concentration of red listed chemicals in the extracted natural dye

Parameters	Permissible range, ppm	Dye , ppm
Heavy metals		
Arsenic	0.02	0.001
Cadmium	0.1	Nil
Chromium	1.0	Nil
Cobalt	1.0	0.002
Copper	10	1.25
Lead	0.08	Nil
Mercury	0.02	Nil
Nickel	1	0.02
Zinc	10	Nil
Pesticides	--	NT
Banned aryl amines	--	NT

Nt- Not traceable.

4 Conclusion

The study shows that natural dye can be efficiently extracted from the waste parts of *Madhuca longifolia*. Maximum extraction is observed at *p*H 10, temperature 95°C and time 60 min. Mordant treatment not only improves the color strength and fastness properties of this natural dye but also results in numerous shades; therefore, this natural dye colourant source can be a promising alternative to synthetic dyes. Fabrics show antibacterial activity against Gram-positive and Gram-negative bacteria, the antimicrobial tests demonstrate an exciting opportunity for the dyed textile as a potential choice in developing protective clothing against common infections in hospitals and hotels. As the leaves of *Madhuca longifolia* are from a renewable source and are abundantly available, the natural dye colorant obtained may be considered a potential source of eco-friendly natural dye for textiles. The current findings clearly demonstrate that the extraction of natural colorants from waste leaves of plants can also be a sustainable technique towards waste utilization.

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