Article

Commercially adoptable process for manufacturing Natural dyes for cotton

R Bhuyan[a], C N Saikia[a*] and K K Das[b]
[a]Regional Research Laboratory, Jorhat 785 006, India
[b]Department of Chemistry, Dibrugarh University, Dibrugarh 786 004, India
*Correspondent author, Phone: 2370121; Fax: 0091-(0376)-2370011
E-mail: rnjb1in@yahoo.co.in

Abstract

Dyes were extracted from the stems, roots and leaves of ***Rubia cordifolia* Linn.**, ***Morinda angustifolia* Roxb.** and ***Tectona grandis* Linn. f.**, respectively and the dyeing behaviours of the colour components on cotton were evaluated. The dyeing was carried out with and without the use of mordants and the fastness properties of the fabrics were determined. The absorbance (%) and colour strengths (*K/S*) of the dyes were evaluated and the colour changes were recorded in Hunter co-ordinates and converted to CIELab co-ordinates.

Keywords : Colour strength, Cotton fabric, Dyeing, Hunter co-ordinates, Mordants, ***Morinda angustifolia***, ***Rubia cordifolia***, ***Tectona grandis***.

Introduction

During recent years, the dyes derived from natural sources have emerged as an important alternative to synthetic dyes. Being biodegradable and highly compatible with environment, the natural dyes are free from the defects associated with synthetic dyes such as harmfulness to human body, pollution and wastewater problem (Eom *et al*, 2001; Padhy & Rathi, 1990; Garg *et al*, 1991). The plant kingdom offers a vast source of natural dyes, which can be obtained from leaves, fruits, seeds, flowers, barks and roots depending on the plant species. Various workers have carried out work on extraction of colour components from different dye-bearing plant species (Onal *et al*, 1999; Nishida & Kobayashi, 1992 a, b; Indrayan & Sharma, 1999; Mathur & Bhandari, 2001a,b, c; El-Zawary & Kamel, 1999).

Roots of Morinda angustifolia

In India, the extraction of dyes from natural sources and their use for dyeing of fabrics/fibres and for other purposes, have been known from time immemorial, but in most cases, the processes of extraction and methods of applications have been localized in the areas of abundance of the dye bearing plants. As such, the methodology adopted for extraction of dyes and their utilization are mainly oriented to the needs of local importance. So is the case with the practice of extraction and utilization of dyes from plant sources in the North-eastern region. There is hardly any


Stems of Rubia cordifolia

commercially adoptable process/ technology available for manufacturing natural dyes, though the practice is in vogue from time immemorial. Therefore, authors attempted to isolate the colouring components from roots, stems and leaves of three abundantly available plant species, viz. ***Morinda angustifolia* Roxb.**, ***Rubia cordifolia* Linn.** and ***Tectona grandis* Linn. f.**, respectively, in order

Leaves of Tectona grandis

to optimize the parameters of extraction and also to study the dyeing characteristics of the colour components on cotton with and without the use of mordants.

Process/Technology

Different parts of the selected plant species (5000 g each) were collected from reserve forests near Jorhat (26°4′ N latitude and 94°12′ E longitude), India. The samples were then washed under free flowing water to remove dust and other foreign particles and then air-dried. The air-dried samples were stored at ambient temperature in sealed plastic bags.

Cotton fibre was soaked in distilled water and then scoured in a non-ionic detergent solution containing $2gl^{-1}$ each of soap and soda ash at 80°C for an hour to remove starch and other stiffening agents. The material to liquor (M:L) ratio was maintained at 1:30, after which the fabric was washed with distilled water, dried and ironed. Prior to dyeing or mordanting, the fibre samples were soaked in water for half an hour (Mukherjee, 1999).

Mordants such as copper sulphate ($CuSO_4.5H_2O$) (LR, CDH), stannous chloride ($SnCl_2.2H_2O$) (LR, CDH) and alum [$Al(NH_4)(SO_4)_2\ 12H_2O$] (LR, CDH) were used. Distilled water was used in extraction and for preparation of all chemical solutions. De-ionized water was used for dyeing purposes.

Preparation of samples for extraction of colour components

The air-dried parts of the plants containing about 12-15% moisture were made into powder in a Wiley mill and kept in sealed plastic covers for subsequent use.

Optimization of extraction conditions of dye components

The finely ground plant parts were weighed and both cold and hot water extracts were prepared. In case of cold water extract, the powder was soaked in water 20% (w/v) and kept for 48 hrs. In case of hot water extraction, two sets of experiments were performed. In the first set, the material was cooked in water in a M:L ratio of 1:20 at boiling temperature with the variation of time from 120-240 min. In the second set, the cooking temperature was varied from 50-100°C at M:L ratio 1:20 maintaining the time of cooking at 2 hrs. The resultant liquid was then filtered. The extracted dye solutions were then concentrated and then dried in hot air circulatory oven at 60-70°C till dry material is obtained. The dry material is then removed from the trays and weighed to find out the yield.

Absorbance and colour strength measurements

The solid mass so obtained was then subjected to extraction with 70:30 (v/v) alcohol:water mixture and then filtered. The filtrate was evaporated under reduced pressure to get a concentrated mass of the colour component.

Dye solutions of 1-5% were then prepared and definite amounts were taken in the dye-bath by maintaining M:L ratio at 1:10. The absorbance of the dye solutions was recorded before and after dyeing on a Shimadzu 1601 PC UV/ Vis spectrophotometer. An average of 3 measurements at each concentration was recorded. The amount of dye absorbed was calculated (Mathur & Bhandari, 2001a). The colour strength (K/S) values of the dye solutions as well as the dyed samples were evaluated by light reflectance technique and the values were assessed using the Kubelka-Munk equation (Saligram *et al*, 1993).

Dyeing of cotton fibres

For dyeing experiments, the cotton fibres were pre-treated with 2% tannic acid at M:L ratio 1:10 in 95-97°C for 30 min (Shenai, 1984).The fibres were then dyed in a dye-bath containing 3% of each of the dyes extracted from ***Rubia cordifolia, Morinda angustifolia*** and ***Tectona grandis*** dyes in the same M:L ratio. The dyeing was carried out at temperature 97-98°C for 45 min, after which 2% sodium chloride solution on the basis of material was added to the dye-bath and the system was further kept at that temperature for 15 min. The fibres were then washed thoroughly with de-ionized water and dried at room temperature.

Mordanting

Pre- and Post-mordanting methods using 2% solutions each of $CuSO_4.5H_2O$, $SnCl_2.2H_2O$ and $Al(NH_4)(SO_4)_2.12H_2O$ were employed at M:L ratio 1:10 and mordanting was carried out for 30 min at 97-98°C. The fibres were then washed and dried.

Measurement of fastness properties

Colour fastness tests to light, washing and crocking were carried out in a Fad-O-meter, Launder-O-meter and Crock-O-meter, respectively as per the standard methods. The fastness ratings were given in Grey-scales (AATCC, 1968).

Conclusion

While extracting the dye in water, it was observed that with the increase in duration of extraction at boiling temperature, the yield of the dye increased but the rate of increase decreased after 180 min of extraction. So also, the temperature of extraction was varied from room temperature (27°C) to 100°C and the yields were found maximum at 100°C. Therefore, the optimum time of extraction was fixed at 180 min in boiling condition (100°C) for all the three samples (Table 1).

As evident from Table 2, the absorption of dye (%) increased with an increase in concentration and reached maximum at 3% concentration for ***R. cordifolia, M. angustifolia*** and ***T. grandis***. Similarly, *K/S* values also increased with an increase in the dye concentration. Though the *K/S* values went on increasing, maximum absorption (35.35% for ***R. cordifolia;*** 31.58% for ***M. angustifolia;*** and 25.88% for ***T. grandis***) were observed at 3%.

The *K/S* values increased with the increase in concentration of mordants. However, beyond 2% concentration of each of the mordants, the increase in *K/S* values and fastness properties on fabric were not significant enough. It was also evident that a wide variety of shades ranging from yellowish brown to golden brown, grey to dark brown and pink to red could be obtained from the dyes when mordanted with different mordants. The mordant activity of metal ions follow the sequences Cu(II)→Al(III)→Sn(II) for ***R. cordifolia***; Cu(II)→Sn(II)→Al(III) for ***M. angustifolia***; and Cu(II) →Al(III) → Sn(II) for ***T. grandis***. The colour intensity was found to be maximum when mordanted with Cu(II) as compared to Al(III) and Sn(II). The fastness tests were applied to both mordanted and un-mordanted cotton fibres. Fair to good wash and crocking fastness properties (Table 3) were obtained with Cu, while light shades were obtained with $SnCl_2.2H_2O$ and alum.

The results on colour co-ordinates of dyed samples are given in Table 4. It was observed that all the colour co-ordinates were positive with respect to brightness *L*, red-green *a*, yellow-blue *b* and therefore, all of them lie in the yellow-red quadrant of the colour space diagram. Further, the *L* values decreased corresponding to deeper shades on mordanting.

The fibres dyed and post mordanted with $CuSO_4.5H_2O$ showed the low *L* values indicating deeper shades obtained on mordanting with these metal salts compared to that obtained with $SnCl_2.2H_2O$ and alum with high *L* values. Thus for all the dyes, Cu(II) may effectively be used as mordant salts.

Optimization of extraction condition of dyes from stems of ***Rubia cordifolia***, roots of ***Morinda angustifolia*** and leaves of ***Tectona grandis*** were carried out as these data may help the upcoming entrepreneurs to study the economic viability of such a project on commercial scale. The dyeing experiments also exhibited acceptable performance properties of the dyes so extracted.

Acknowledgement

One of the authors, RB acknowledges the award of Senior Research fellowship of CSIR, India.

References

1. El-Zawahry MM and Kamel NM, Dyeing of wool cloth with *Terminalia belirica* fruit extract, *Indian J Fibre Text Res*, 1999, **24**, 126-130.

2. Eom S, Shin D and Yoon K, Improving the dyeability of natural colourants on cotton by cationization, *Indian J Fibre Text Res*, 2001, **26**, 425-431.

3. Garg A, Shenda S and Gupta KC, Effect of mordants on colour of natural dye extracted from tessue flowers (*Butea monosperma*), *Colourage*, 1991, **38**(2), 50-53.

4. Indrayan AK and Sharma V, Isolation and extraction of medicinally useful dye from the heartwood of *Acacia catechu* using different solvents, *Orient J Chem*, 1999, **15**(1), 191-192.

5. Mathur JP and Bhandari CS, 2001a, Use of beet sugar as wool colourant, *Indian J Fibre Text Res*, 2001, **26**, 313-316.

6. Mathur JP and Bhandari CS, 2001b, Physico- chemical study of dyed wool:

Part I – *Bougainvillaea* as wool colourant with mixed mordant, *Indian J Fibre Text Res,* 2001, **26**, 432-437.

7. Mathur JP and Bhandari CS, 2001c, Physicochemical study of dyed wool: Part II – Red Sandalwood as wool colourant with mixed mordant, *J Inst Chem (India)*,2001, **73**(4), 128-130.

8. Mukherjee A, Dyeing of cotton fabric with *Kigelia pinnata*, *Spathodea campanulata* and *Carissa congesta*. Book of Papers – Convention on Natural Dyes, 114-118, 1999.

9. Nishida K and Kobayashi K, 1992a, Dyeing properties of Natural dyes under after-treatment using metallic mordants, *Am Dyest Rep*, 1992, **81**(5), 61-62.

10. Nishida K and Kobayashi K, 1992b, Dyeing properties of Natural Dye from Natural sources: Part I, *Am Dyest Rep*, 1992, **81**(7), 44-45.

11. Onal A, Yildiz A and Tutar A, Extraction of dyestuff from Valonia oak (*Quercus cerris*): Dyeing of woolen strips, cotton and feathered-leather, *Bull Pure Appl Sci*, 1999, **18C**(2), 77-87.

12. Padhy RN and Rathi D, Effect of mordants on dyeing of cotton with vegetable dyes, *Text Dyer Printer*, 1990, **23**(25), 27-28.

13. Saligram AN, Singh S, Shrivastava RS and Shukla SR, Low-temperature dyeing of silk with direct and basic dyes using redox systems, *Am Dyest Rep*, 1993, **82**(5) 30.

14. Shenai VA, Technology of Textile Processing, Vol VI, Technology of Dyeing, Sevak Publications, Bombay, 27, 1984.

15. *Technical Manual of AATCC*, Vol. 25 (American Association of Textile Chemists and Colorists, U.S.A.), 619-639, 1968.

16. Wyszecki G and Stiles WS, Color Science: Concepts and methods, quantitative data and formulas, John Wiley and Sons Inc., 460, 1967.

Table 1 : Yields of colouring components from on varying time and temperature of extraction

Concentration of powder in water (%)	Temperature (°C)	Time (h)	Yields (%)		
			Rc	Ma	Tg
20	27	48	3.2	2.5	2.1
	50	2	15.1	13.2	11.8
		3	16.4	14.3	12.4
	100	2	18.6	16.6	14.8
		3	20.1	20.5	16.4
		4	21.3	21.5	17.4

Rc, *Rubia cordifolia; Ma, Morinda angustifolia;* Tg, *Tectona grandis*

Table 2 : Absorption (%) and *K/S* values of different concentrations of the natural dyes

Dyes from Plant species	Wavelength (nm)	Dye concentration (%)	Absorbance		Absorption (%)	*K/S*
			Before dyeing	After dyeing		
Rc	410	1	0.19	0.16	15.38	12.35
		2	0.22	0.16	25.33	14.26
		3	0.29	0.19	35.35	15.59
		4	0.36	0.25	29.91	17.73
		5	0.40	0.29	26.55	19.01
Ma	440	1	0.16	0.14	12.50	9.52
		2	0.17	0.13	23.53	10.28
		3	0.19	0.13	31.58	11.11
		4	0.20	0.15	25.00	12.63
		5	0.22	0.17	22.73	14.88
Tg	380	1	0.15	0.13	10.45	9.51
		2	0.16	0.13	13.12	10.31
		3	0.19	0.14	25.88	11.30
		4	0.20	0.16	20.29	13.02
		5	0.21	0.17	20.77	14.50

Rc, *Rubia cordifolia*; Ma, *Morinda angustifolia;* Tg, *Tectona grandis.*

Table 3 : Shades and fastness properties of dyed and mordanted cotton fabric

Dyes from Plant species	Mordants	Method Of mordanting	Light fastness	Crocking fastness		Wash fastness	Shade
				Dry	Wet		
Rc	Nil		4	4	5	4	Pink
	$CuSO_4.5H_2O$	I	4	4	6	4	Brown
		II	4	4	6	5	Brown
	$SnCl_2.2H_2O$	I	3	4	5	4	Pinkish yellow
		II	4	4	5	4	Pinkish yellow
	Alum	I	3	3	5	3	Light pink
		II	4	4	5	4	Brown
Ma	Nil		3	3	4	4	Pale pink
	$CuSO_4.5H_2O$	I	5	4	5	5	Dark brown
		II	4	4	6	4	Brown
	$SnCl_2.2H_2O$	I	4	3	5	4	Yellowish brown
		II	4	3	4	5	Yellowish brown
	Alum	I	4	4	5	5	Pale pink
		II	4	4	5	5	Pale pink

Tg	Nil		3	3	4	3	Grey
	$CuSO_4.5H_2O$	I	4	4	5	4	Grey
		II	4	4	5	3	Dark grey
	$SnCl_2.2H_2O$	I	3	3	4	3	Light grey
		II	3	3	4	3	Light brown
	Alum	I	3	3	4	3	Light brown
		II	3	3	4	3	Light brown

Rc, *Rubia cordifolia*; Ma, *Morinda angustifolia*; Tg, *Tectona grandis*.
I – pre- mordanting; II – post-mordanting
1 – very poor, 2 – poor, 3 – fair, 4 – very fair, 5 – good, 6 – very good.
AATCC Method : Light fastness – Method 16B 1964.
Wash fastness – Method 36-1965
Crocking fastness – Method 8-1961

Dyed cotton fabric samples ➤

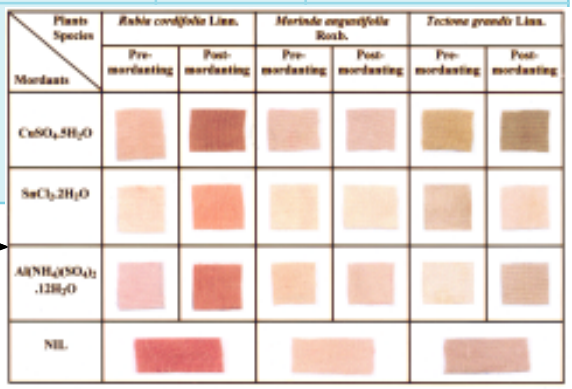

Table 4 : Colour co-ordinates of dyed and mordanted cotton samples

Dyes from Plant species	Mordants	Mordanting technique	Colour co-ordinates		
			L	*a*	*b*
Rc	Nil	-	44.97	58.88	11.98
	$CuSO_4.5H_2O$	I	43.87	48.91	28.17
		II	35.98	35.19	30.17
	$SnCl_2.2H_2O$	I	60.17	40.13	19.85
		II	75.12	26.91	31.18
	Alum	I	42.87	47.18	18.27
		II	39.72	37.12	40.13
Ma	Nil	-	21.02	9.80	6.93
	$CuSO_4.5H_2O$	I	41.01	12.14	20.12
		II	40.31	11.79	19.98
	$SnCl_2.2H_2O$	I	23.37	10.98	7.81
		II	43.44	12.14	65.24
	Alum	I	23.14	10.14	7.12
		II	24.06	10.42	7.15
Tg	Nil	-	49.16	15.54	8.34
	$CuSO_4.5H_2O$	I	43.27	18.67	14.34
		II	35.00	25.78	20.44
	$SnCl_2.2H_2O$	I	59.27	16.54	34.32
		II	53.87	15.32	45.10
	Alum	I	51.86	13.49	30.14
		II	45.11	14.01	24.25

Rc, *Rubia cordifolia*; Ma, *Morinda angustifolia*; Tg, *Tectona grandis*.
I – pre- mordanting; II – post- mordanting