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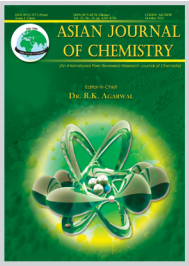

Natural Dyeing of Silk Fabric Using Eco-Friendly Mordants

M. SUNDRARAJAN*, R. RAJIV GANDHI, J. SURESH, S. GOWRI and S. SELVAM

Advanced Green Chemistry Laboratory, Department of Industrial Chemistry, School of Chemistry, Alagappa University, Karaikudi-3, India

*Corresponding author: Fax +91 04565 225202; Tel: +91 94444 96151; E-mail: sundrarajan@yahoo.com

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Parijataka dye (*Nyctanthes arbor-tristis*), which is a natural colourant extracted from orange corolla tube of parijataka flowers were studied for their dyeing potential on silk with a variety of mordants. Colour of the pigment was fixed by the utilization of eco-friendly mordants like $SnCl_2$, $CuSO_4$, $FeSO_4$, $K_2Cr_2O_7$, pomegranate, tannin, alum and lime juice. Some of the mordants used are new for parijataka dyeing. Among the various mordanting method simultaneous mordanting method was the most suitable one for dyeing of parijataka on silk. The range of colour developed on dyed materials are evaluated (in terms L*, a*, b*) and the improvement of colour strength on fabric by mordants were also examined. Dye ability was more in unmordant and lime juice mordanted silk fabric. The colour strength (K/S) and wash fastness values has been found to be good in all dyed samples especially $SnCl_2$, alum and lime juice mordanted fabrics.

Key Words: Natural dye, Parijataka, Silk, Mordant, Dyeing.

INTRODUCTION

Dyes for dyeing textiles, used before the birth of Christ, until about 150 years ago all dyes were natural substances, derived mainly from plants and animals. The natural dyes present in plants and animals are pigmentary molecules[1,2]. Studies to increase the efficiency of the extraction process and identify plants with the highest concentration of dyes were already being conducted in middle of the century[3]. Conventional methods wisdom leads to the belief that natural dyes are friendlier to the environment than their synthetic counter parts. Synthetic dyes are commercially available because of the ease of use and the various colour shades available but environmental problems are concerned. So natural dyes only the options because they present environmental compatibility, low toxicity and low allergic reactions for fabric wearers. Natural dyes can exhibit better biodegradability and generally have a higher compatibility with the environment[4-7]. India is rich in natural wealth and there are ample scope to explore and revive application of natural dyes on textiles, having more and more scientific knowledge base available[8].

Parijataka is one of the most valuable gifts of nature to mankind; the botanical name of parijataka is *Nyctanthes arbor-tristis*. It is a short stalk flower with two white petals and orange corolla tube. The bright orange colour tube size is 0.5 to 0.75 inches. It contains colouring matter nyctanthin, which is identical with cracetin ($C_{20}H_{24}O_4$) from salfron[9]. In the view of polluting aspects of synthetic dyes and long term non-sustainable nature of these petrochemical derivatives, natural dyes are gaining importance. Presently there is an excessive use of synthetic dyes, which is estimated around 10,000,000 tones per annum[10]. However there is a need to improve the dye fastness and the reproducibility of the dyeing process[11]. Vegetable dyes require chemical in the form of metal salts to produce an affinity between the cotton, silk and wool fabrics these chemicals are known as mordants[12]. Natural dyeing of cotton fabric has always posed challenges, although silk is an easy to dye. Several metallic salts and bio-mordants have been used by some researchers[13].

Generally, problems in dyeing with natural dyes are related to low exhaustion of dyes and to the poor fastness of the dyed fabrics. Attempts to overcome these problems have been mainly focused on the use of metallic salts as mordants, which are traditionally used to improve fastness properties, exhaustion and to develop different shades with same dye. Fastness property of natural dye is often enhanced by metal mordants, which form an insoluble complex with dye molecules in which flavonols have greater tendency towards chelate formation due to the presence of hydroxyl-keto functionality [14-18]. The metal ions can act as acceptors to electron donors to form coordinate bonds with dye molecule, which is insoluble in water[19]. Many reports related to mordant research were focused on the effect of synthetic mordant (aluminum sulphate, copper sulphate, lead acetate and potassium dichromate *etc*.,)[20] and no much effort had been made to study the effect of natural mordant in dyeing industry. Considering the importance of the natural dyes

with natural mordant and the harmful effect of synthetic dyes with synthetic mordant, an effort had been made to formulate an eco-friendly herbal dye with a natural mordant.

India has the unique distinction of being bestowed by the nature with all four varieties of silk *i.e*. mulberry, tasar, muga and eri. Eri silk is available in hill areas are better known for its thermal properties and durability. Dyeing eri silk with natural plant dyes will add value to the product that can go a long way in improving economy of hill people. Bombyx mori silks are the strongest natural protein fibers and excellent fibers with many outstanding attractions, including luster, dyeability and moisture absorption. Silk fiber consists of 97 % protein and the others are wax, carbohydrate, pigments and inorganic compounds. The proteins in silk fiber are 75 % fibroin and 25 % sericin by weight, approximately. The sericin makes silk fiber to be strong and lackluster; therefore, it must be degummed before dyeing[21-23].

The purpose of this study is to investigate the dyeing and fastness properties of natural colourant extracted from parijataka flower on silk fabric using novel and eco-friendly mordants. Effect of dye ability was evaluated in terms of dye uptake and K/S value.

EXPERIMENTAL

Degummed and scoured silk fabric was used. The extract of parijataka flower is selected for natural dyeing. Sodium chloride and carbonates were used for exhausting and fixing the dyes. The metal ion mordants used were copper sulphate, potassium dichromate, alum, stannous chloride and ferrous sulphate. In addition, the natural mordants used are pomegranate rind, tannin and lime juice used is yellow colour lemon (citrus limon). Lime juice was taken for mordanting as per the general recipe followed for all mordants.

Dye extraction method: Flowers of coral jasmine (*Nyctanthes arbor-tristis*) were collected from our local residential area. Individual flower of coral jasmine was segregated into two parts (based on the colour) namely orange corolla tube and white corolla lobe. Only the orange corolla tubes were subjected to soxhlet extraction with water at 100 ºC for 2 h. The extract obtained was then filtered. The filtrate was subjected to evaporation for dryness. The dye thus obtained was then purified with alcohol. The extracted dye solution was evaporated to $1/10^{th}$ volume under controlled temperature.

Mordanting: Mordant concentration [3 %], liquor ratio [1:30] and temperature [80 ºC] was used for all mordanting methods.

Pre mordanting method: In premordanting, first the silk fabric should impregnated in each mordant for 0.5 h and then the impregnated samples were dried at room temperature for dyeing process.

Post mordanting method: The fabric is first dyed and then mordanted by impregnation with the mordant solution at room temperature for 0.5 h. It was then removed, washed with water to remove unfixed dye and then dried.

Simultaneous mordanting method: In this method the degummed silk material was entered into dye bath containing both dye and mordant. After 45 min the fabric was washed and dried.

Dyeing recipe: All solid chemicals were taken as w/w percentage and liquids were taken as v/v percentage. The silk fabric was dyed with dye extract, keeping M:L ratio as 1:30. The dye extract was prepared by dissolving 5 g of dye in 100 mL water and the fabric was dipped in the dye bath at 80 ºC in presence of 10 % sodium chloride with constant stirring, after 15 min, 3 % sodium carbonate was added and stirred well. After 0.5 h, the dyed material was washed with cold water and dried at room temperature. The same procedure was followed for all mordanting method.

Measurement of colour strength: The colour strength (K/S) of the dyed samples was determined from the reflectance measurement (R) using Kubelka-Munk equation.

$$K/S = (1-R)^2/2R$$

The reflectance value of the dyed sample was measured by an UV spectrophotometer (Gretamecbeth colour eye UV/2180) at the wavelength of minimum reflectance under CIE D_{65} and 10º observation.

Washing fastness: The dyed silk was tested for fastness properties according to standard ISO Test 3 testing method (ISO 105-C03:1989, Geneva). Dyed samples of 10 × 4 cm were stitched with one of the shorter side of the adjacent bleached fabric and put into the Launderometer at 60 ºC for 0.5 h and then washed with hot water, cold water and dried.

RESULTS AND DISCUSSION

To examine the effect of dye concentration on dye uptake and wash fastness, the dye concentration was varied from 3-9 %. Table-1 shows the results of dye uptake for the all the mordants with the different dye concentration. As the dye concentration increased from 3 to 9 %, dye uptake values increased appreciably. The highest dye uptake was obtained at 9 % dye concentration for all the mordants. Among the different mordants, limejuice showed good dye uptake when compared to others. The dye uptake of different mordants increased in the order with dye concentration: lime juice > unmordant > alum > $SnCl_2$ > $FeSO_4$ > pomegranate > $CuSO_4$ > tannin > $K_2Cr_2O_7$.

TABLE-1
OPTIMIZATION OF PARIJATAKA FLOWER DYED FABRIC IN SIMULTANEOUS MORDANTING METHOD

Mordants	Dye uptake (%) Dye concentration			
	3 %	5 %	7 %	9 %
Unmordant	40	50	70	80
Stannous chloride	50	60	60	75
Ferrous sulphate	40	50	60	70
Copper sulphate	20	40	50	60
Potassium dichromate	20	30	30	40
Alum	40	60	65	80
Tannin	30	30	40	60
Pomegranate	40	55	60	65
Lime juice	60	70	75	90

Table-2 provides the data of dye uptake and wash fastness with the salt concentration varied from 8- 12 %. At 8 % salt concentration, all the mordants and unmordant showing good dye uptake. There is no periodical increase or decrease in the dye ability during the variation of salt concentration. The best

TABLE-2
OPTIMIZATION OF SALT [8-12 %] FOR PARIJATAKA DYED FABRIC IN SIMULTANEOUS MORDANTING METHOD

Mordants	Dye uptake (%)			Wash fastness					
				Change in shade			Change in stain		
	8 %	10 %	12 %	8 %	10 %	12 %	8 %	10 %	12 %
Unmordant	80	70	70	2-3	2-3	2-3	2-3	3	3
Stannous chloride	80	70	70	3-4	2	3-4	3	3-4	3
Ferrous sulphate	70	70	70	4	2	2	2	2-3	2-3
Copper sulphate	60	60	60	1	1-2	1-2	1-2	2	2
Potassium dichromate	50	40	40	2	1	1	2	2	1-2
Alum	80	70	70	3	3	3-4	3-4	3	3-4
Tannin	40	40	40	2	2	2	2	2	2-3
Pomegranate	50	50	40	3	3	2-3	2-3	2	3
Lime juice	90	90	85	3	3-4	3	3	2-3	3-4

dye uptake was obtained for lime juice and the order of dye uptake is as follows lime juice > unmordant > alum > $SnCl_2$ > ferrous sulphate > $CuSO_4$ > pomegranate > $K_2Cr_2O_7$ > tannin. Washing fastness values of dyed samples with various mordants showed good results for limejuice, stannous chloride and unmordanted fabric.

The colourimetric data such as L*, a*, b*, c*, hº and K/S values of the simultaneous mordanted fabric is shown in Table-3. Without mordant, parijataka dye is showing better dye uptake. The K/S values of the different kinds of mordants in simultaneous mordanting was as follows: lime juice > unmordant > $SnCl_2$ > pomegranate > alum > $FeSO_4$ > tannin > $CuSO_4$ > $K_2Cr_2O_7$. It was observed the lime juice (citrus limon) was the best mordant to improve the colour strength of fabric.

TABLE-3
COLOUR STRENGTH (K/S) DATA FOR THE DYED SAMPLES WITH AND WITHOUT MORDANTS

Mordants	L*	a*	b*	c*	hº	K/S
Unmordant	77.50	1.37	75.23	73.84	88.96	7.53
Stannous chloride	78.59	1.70	71.80	71.82	88.65	7.46
Ferrous sulphate	59.93	8.63	76.76	77.25	83.59	5.39
Copper sulphate	60.25	4.91	84.95	75.24	86.69	3.58
Potassium dichromate	67.93	2.14	40.13	40.19	86.95	1.95
Alum	63.31	6.37	60.84	61.18	84.02	6.41
Tannin	57.97	9.56	51.90	52.77	79.57	4.14
Pomegranate	56.09	8.90	73.31	85.09	83.08	6.18
Lime juice	67.80	15.13	107.81	108.87	82.01	8.16

L* = light power; a* = redder; b* = greener; c*= chroma value; hº = hue value K/S = colour strength

Silk is a fibrous protein with several amino acids, which are capable to exist as zwitter ions. Concentration of lime increases the dye uptake also increases. In general, lime juice mordant is showing good dye uptake, better colour strength and acceptable wash fastness by lowering the pH of the dye bath which makes it more acidic. But lime juice mordant does not form any interaction between dye and fibre.

The dye solution was subjected to UV-visible spectral analysis. Fig. 1 shows the absorption at blue region with a wavelength over the range of 430-450 nm, this conform the appearance of yellow colour of parijataka dye. So this area was chosen for investigation.

Fig. 2 explains about the effect of different kinds of mordants on fabric for the dye parijataka. Parijataka a yellow colour dye, which has good dye uptake and poor wash fastness on fabric without any use of mordants. Among the various mordants used for dyeing stannous chloride and ferrous sulphate produced better result in post mordanting, but alum, tannin and lime juice produced better dyeability in premordanting method. All the three methods of mordanting produced identical result for copper sulphate and potassium dichromate. While comparing the effect of all mordants and different mordanting methods, lime juice mordant and simultaneous mordanting method produced good dye uptake and wash fastness. Fig. 3 explains about the different hue achieved on silk fabric with different mordants by simultaneous mordanting method. The best yellow shades with parijataka dye were obtained for lime juice, unmordant, $SnCl_2$ and alum. All other mordants giving contaminated yellow shade.

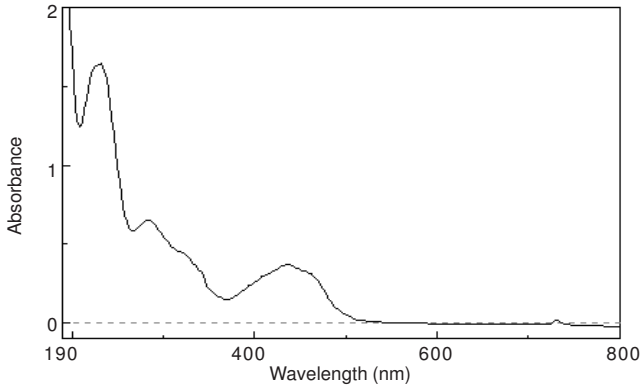

Fig.1. UV-Visible spectrum of *Nyctanthes arbor-tristis*

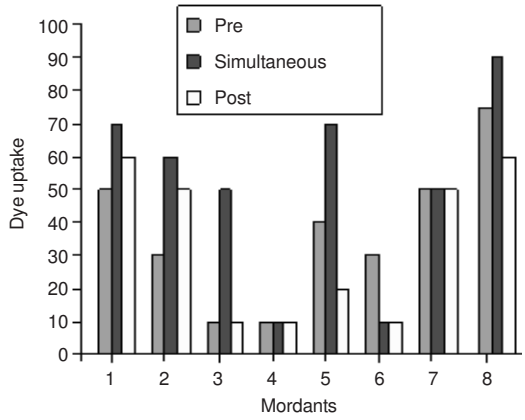

Fig. 2. Effect of mordanting methods (1. Stannous chloride, 2. Ferrous sulphate, 3. Copper sulphate, 4. Potassium dichromate, 5. Alum, 6. Tannin, 7. Pomegranate rind, 8. Lime juice)

Mordants	Dyed Samples
Unmordant	
Stannous Chloride	
Ferrous sulphate	
Copper sulphate	
Potassium dichromate	
Alum	
Tannin	
Pomegranate-rind	
Lime juice	

Fig. 3. Dyed samples of parijataka in simultaneous mordanting method

Conclusion

The above results have strongly exposed that the mordanting method suitable for parijataka dyeing on silk is simultaneous mordanting because of its best dye uptake and better depth of shade. The effect of different mordants and unmordant on K/S values of dyed silk fabrics were investigated. The order of K/S values is lime juice > unmordant > $SnCl_2$ > pomegranate > alum > $FeSO_4$ > tannin > $CuSO_4$ > $K_2Cr_2O_7$. The optimum dye and salt concentration were set up to be 9 % and 8 % respectively. The dye uptake with various dye concentration and salt concentration follows the order: lime juice > unmordant > alum > $SnCl_2$ > $FeSO_4$ > pomegranate > $CuSO_4$ > tannin > $K_2Cr_2O_7$ / limejuice > unmordant > alum > $CuSO_4$ > ferrous sulphate > $SnCl_2$ > pomegranate > $K_2Cr_2O_7$ > tannin respectively. Similarly lime juice, $SnCl_2$ and unmordanting have caused improved fastness properties with simultaneous mordanting. Thus the net enhancement of dye uptake and wash fastness values has been found to be best for lime juice and unmordant compared to other mordants. Commonly mordants improve the dye uptake of natural dyes but for parijataka dye there is no significant difference except in lime juice. As a result addition of mordant is not necessary for parijataka dyeing on silk fabric.

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