Indian J. Applied & Pure Bio. Special Volume 159-169 (2021).
Environment & Sustainable Development : Problems, Prospects & Mitigation Date : 25th-27th November, 2021
ISSN: 0970-2091
A web of Science Journal

Sustainable utilization of betalains rich *Rivina humilis* L. berries as natural mordant for dyeing silk and wool with eco-friendly natural dye

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Abstract

Natural dyes are sustainable those cover the area of green chemistry. Natural dyes never pollute like synthetic dyes as they are obtained from the renewable resources. The use of non-toxic and eco-friendly natural dyes on textiles has become significant importance due to the increased environmental awareness associated with synthetic dyes. The synthetic dyes are toxic, mutagenic and carcinogenic agents persist as environmental pollutants. Natural dyes are ecofriendly and do not damage human health. Sustainable utilization of natural resources is the proper management of natural resources for the benefit of the entire human community. Eucalyptus is considered one of the most important sources of natural dyes which have ample natural tannins and polyphenols, with the major colouring component. *Rivina humilis* L. (Phytolaccaceae) or blood berry has betalains content in its berries which can be used as sustainable resource of natural mordant for dyeing textile filaments. This species prefers damp, shady sites and is a weed of closed forests, forest margins, disturbed sites, waste areas and urban bushland. This study investigated that Rivina berries extract was used as sustainable resource of natural mordant in dyeing eucalyptus leaves on silk and wool fabrics. The dyeing was carried out with and without mordants by Pre, Post and Simultaneous mordanting method. The variations in the colour strength and coordinates with respect to dyed without and with mordant showed a significant difference in colour obtained. The colour fastness to washing, rubbing (dry and wet), light and perspiration of dyed fabrics were tested according to ISO standards. The results showed that silk and wool dyed fabrics without mordant showed a shade of light brown, while Rivina berries mordants exhibited dark reddish brown shades in simultaneous mordanting method. The pre and post mordanted fabrics showed pale brownish shades compared to unmordanted fabrics. The colour fastness to washing, perspiration, rubbing and light were good except unmordanted fabric in simultaneous

mordanting whereas pre and post mordanted with Rivina berries showed moderate to good fastness compared to unmordanted farbrics. Hence, *Rivina humilis* L. berries which occupy wastelands, bushland can be used as sustainable resource of natural mordant instead of synthetic metallic mordants which harm environment.

Sustainable utilization of natural dyes in textile industry can be attained through low-cost production of natural dyes and conserving biodiversity, application of mordant-like material from renewable resources[7]. The use of natural dyes is beneficial with regard to environmental impact and sustainability, especially if the dyes are extracted from natural resources[26]. Natural dyes are mostly used in colouration of textiles, leather, food substrate since pre-historic times[19]. Natural dyes give good shades and also provide ultraviolet protection, antibacterial and antifungal characteristics[20]. Annually, ten thousand tons of natural dyes are currently being used globally by industries for coloration [(5)]. The natural dyes are mainly used in textile, medicine, paint, pharmaceutical, dermatology, cosmetology, food industry *etc*.[5].

The demand for use of natural dyes has increased due to its non-hazardous nature, biodegradability, eco-friendliness[1)]. The use of non-allergic, non-toxic and eco-friendly natural dyes on textiles has become a significant importance associated with increased environmental awareness[2,4]. Generally, natural dyes lack affinity towards textile substrates which needs mordants. The colour of the substrate does not only depend on a dye, but it is also influenced by the type and concentration of mordant used. Most of the metal mordants are not environment friendly which decreases the sustainability of natural dyes. This makes it essential to explore the use of natural mordants in natural dyeing.

In natural dyeing, the usage of metallic mordants destroys the environmental friendliness of natural dyes contributing to soil and water pollution[21]. The application of mordants in the natural dyeing process improves weak adhesion, colour uniformity, and fastness properties of naturally-dyed textile materials[3]. The natural mordants, mostly tannin-rich compounds which are responsible for forming hydrogen bonds and also help to replace metallic equivalents[10].

Rivina humilis L. (Phytolaccaceae) commonly known as blood berry, is a wild herbaceous bushy perennial plant found in various types of shaded soils. It grows up to a height of 120 cm (4 ft)[24]. The berries of the plant are rich with betalain pigments and it posses antioxidant properties. The studies reported dietary safety, bioactivity with related to pigment and its phytoconstituents[16]. The various aspects of betalains pigment were studied with an intention to use them as water soluble colorant for food, pharma and cosmetics industries[23]. The present study was to utilize betalains rich *Rivina humilis* berries which occupy wastelands can be used as sustainable resource of natural mordant instead of synthetic metallic mordants which harm environment.

Source of plant materials :

Ripened berries of *R. humilis* L. were

Figure 1: Dried berries of *Rivina humilis* L.

Figure 2: Extraction of dye

collected from shady areas of the environs of Forest campus, IFGTB, Coimbatore. *Eucalyptus camaldulensis* leaves (EC1) clone of IFGTB was obtained from Field Research Station, Panampally, Kerala. The collected Rivina berries and Eucalyptus leaves were shady dried under room temperature for a week. The dried Rivina berries and extraction of dye was shown in the figure 1 and 2.

Textile materials :

The silk fabric, plain weave and 100% ready for dyeing type was procured from Siera Silk Mills Pvt Ltd, Bangalore, India. The wool fabric, plain (100% merino wool) ready for dyeing type was purchased from Sanjay Shah & Associates, Vapi, Gujarat, India.

Extraction of natural dyes :

The natural dyes were extracted by aqueous extraction technique at 80±5ºC for 60-90 minutes using RO water. The plant parts and concentration of sample used are shown in Table-1. The extracted dyes were filtered using nylon fabric and the solutions were used to dye and mordant the fabrics. The recipe for extraction of dyes from *Rivina* berries and Eucalyptus leaves were shown on Table-2.

Table-1. Plant parts and concentration for extraction of dye

Plants Materials Used	Concentration
Rivina berries	100g/l
Eucalyptus Leaves	100g/l

Table-2. Dyeing recipes

Part of plant	MLR	Temperature (°C)	Time (minute)
Rivina berries	1:10	80	30
Eucalyptus Leaves	1:10	85	90

MLR*- Material to liquor ratio

Scouring of Fabrics :

The undyed test fabrics of silk and wool was treated with non-ionic surfactant solution containing 2g/L each of soap and soda ash at 60ºC for 30 minutes to remove dirt and other stiffening particles. The test fabrics were finally washed and dried.

Mordanting :

The mordanting was carried out through

- ***Pre-mordanting :***
  In premordanting process, mordanting was done before dyeing and subsequently the fabric was dyed with natural dye. It is a two bath process in which the first bath is used for mordanting of fabric and in the second bath, dyeing is done with natural dyes.
- ***Simultaneous mordanting :***
  In simultaneous mordanting methods, the mordant are added with natural dye in the same dye bath; dyeing and mordanting take place simultaneously.
- ***Post mordanting :***
  In post mordanting method, the dyeing of fabric was done first; after then mordanting was carried out [9].

Rivina humilis L. berries dye extract was prepared 10% on weight of fabric (o.w.f) for the mordanting. The mordanting was carried out at 80±5ºC and continued for 1 hour.

Dyeing of fabrics :

Before the dyeing process, the test fabrics were immersed in hot water to facilitate uniform penetration of the dye molecules. The fabrics were dyed using Eucalyptus leaves extract at 85±5ºC for 90 minutes. The material was heated at low temperature initially, which was raised to 85±5ºC and gently stirred.

Subsequent treatment of dyed fabrics :

Then the dyed fabrics were washed by boiling for 10 min in the presence of a standard solution of soap at 5 g/l to remove surface adhered dye molecules from the dyed fabric. The washed fabric was then rinsed for 10 min with water at room temperature to remove the soap from the fabric completely. The dyed fabric was then dried.

Colour measurement :

Colour coordinates (L*, a*, b*) and colour strength (K/S) of samples were assessed under light D65 and 10° observer on spectrophotometer (Colour-Eye 7000 A). Colour strength (K/S) of dyed wool fabrics were measured using equation.

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

K= Absorption coefficient, S= Scattering coefficient, R = Reflectance.

K/S indicates the depth of colour, L* represents the lightness, a* defines the red-green coordinates of colour, b* represents the yellow-blue coordinates of colour [22].

Evaluation of colour fastness :

The colour fastness was studied according to ISO 105-C10:2006 for wash fastness[11], ISO 105 X12:2016 for fastness to rubbing[14], ISO 105 B02:2014 for light fastness[13]

and ISO 105 E04:2013 for fastness to perspiration[12] respectively.

Assessment of the result of colour fastness:

The grey scale rating was used for assessing the degree of change in colour. The scale consists of nine pairs of standard grey scale colour chips each representing visual difference and contrast. It has 9 possible values, *i.e.* 5, 4-5, 4, 3-4, 3, 2-3, 2, 1-2, 1.

Colour strength and Colour coordinates:

The color strength on dyed silk and wool fabrics s was evaluated in terms of their CIEL *a*b* coordinates and K/S values. The colour strength (K/S value) of the Rivina berries mordanted fabrics was shown in the table 3, 4 and 5. The different shades of colors were obtained increases the aesthetics value of the textile fiber *i.e.* silk and wool fabrics. Analysis of the colour strength (K/S values) enlighted that the optimum mordanting techniques are between simultaneous mordanting (K/S= 5.67) on silk, (K/S= 4.13) on wool and post mordanting (K/S= 3.12) on silk, (K/S= 3.36) on wool techniques which have higher values of colour strengths as compared to pre-mordanting techniques (K/S= 2.67) on silk, (K/S= 2.59). It can also be based on the difference between the colour strength and lightness (L) of the standard and the mordanted fabric, mordants generally increased the uptake of dyes on the fabric[17].The simultaneous mordanted fabrics shows better colour strength (K/S) compared to other both mordanting methods. With the use of mordant, a significant decrease in L* and appreciable effect on other colourimetric properties as indicated by a* and b* were obtained. The lightness value (L) of the dye, did not show a particular trend although, there was a significant increase in colour strength when a mordant was used in the dyeing process. Among the applications of mordant, in all three mordanting methods, it was observed that the higher colour shade was achieved by simultaneous mordanting method. After mordanting, improvement in colour values might be because of a higher extent of exhaustion of Rivina berries on silk and wool resulting in better shades of colour. The effect of rivina berries on colour strength of the dyes on silk and wool fabrics with different mordanting methods was shown in the figure 3 and 4.

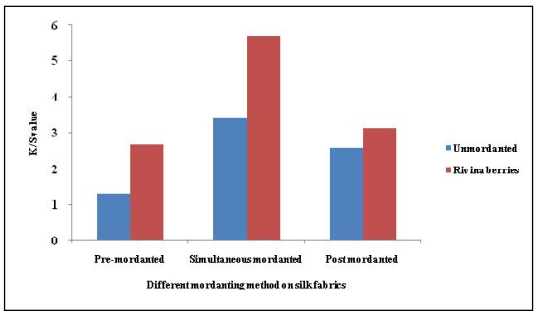

Figure 3. Effect of Rivina berries mordant on colour strength of the dyes using silk fabrics with different mordanting methods

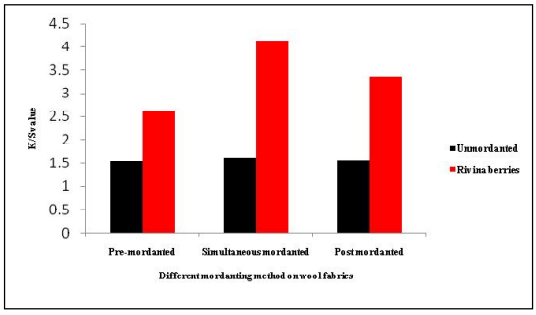

Figure 4. Effect of Rivina berries mordant on colour strength of the dyes using wool fabrics with different mordanting methods

The mordant used for natural dye extraction showed a significant variation in colour strength (K/S) of the dyed fabrics[15,18,28]. The source of plant used for natural dye extraction, mordants type, interaction of dye and mordant and method of dyeing and mordanting method showed a significant effect on colour strength. The result indicates that increased fibre swelling with higher temperature leads to a higher dye uptake and improved dye diffusion[6,25,27].

Table-3. Colour strength and colour coordinates of the dyed silk and wool fabrics using pre mordanting method

Fabrics	Mordants	L*	a*	b*	K/S	Color shades
Silk	Without Mordant	75.6	1.4	-1.1	1.28	
	Rivina Berries	49.8	1.7	-4.1	2.67	
Wool	Without Mordant	74.7	3.3	11.5	1.55	
	Rivina Berries	77.0	-1.8	21.6	2.59	

Table-4. Colour strength and colour coordinates of the dyed silk and wool fabrics using simultaneous mordanting method

Fabrics	Mordants	L*	a*	b*	K/S	Color shades
Silk	Without Mordant	71.8	1.8	0.3	3.41	
	Rivina Berries	58.7	2.0	6.5	5.67	
Wool	Without Mordant	76.0	3.8	14.8	1.61	
	Rivina Berries	83.6	2.2	24.3	4.13	

Table-5. Colour strength and colour coordinates of the dyed silk and wool fabrics using post mordanting method

Fabrics	Mordants	L*	a*	b*	K/S	Color shades
Silk	Without Mordant	72.81	1.5	0.5	2.58	
	Rivina Berries	61.74	1.9	5.7	3.12	
Wool	Without Mordant	75.8	3.1	13.0	1.57	
	Rivina Berries	80.1	1.0	23.1	3.36	

Fastness Properties :

It was observed that mordanting with rivina berries shows excellent to good fastness properties. The fastness properties of silk and wool fabrics mordanted with Rivina berries are shown in the table 6-9.

Measurement of color fastness to wash (ISO 105-C10:2006) :

The outcome of wash fastness was shown in the following Table-6. It is clear that, for simultaneous-mordantation color fastness to wash showed 4-5 grey scale rating for Rivina berries mordant for both silk and wool fabric compared to pre and post mordanting method which showed 4 grey scale rating. On the other hand, the post mordanted fabrics also showed appreciable wash fastness than pre mordantation. The simultaneous mordanted fabrics have showed very good wash fastness.

Measurement of color fastness to rubbing (ISO 105 X12:2016) :

Rubbing fastness is measured in both wet and dry conditions according ISO standard. The rubbing fastness of silk and wool fabrics using different mordanting methods dyed with Eucalyptus leaves extracted dye with Rivina berries mordant was highlighted in Table-7. The result was better for simultaneous-mordantation and the rating was 4-5 in dry condition and 4 in wet condition. The pre and post mordanted fabrics also showed for excellent to good rubbing properties.

Measurement of color fastness to light (ISO 105 B02:2014) :

The light fastness of silk and wool fabrics using different mordanting methods dyed with Eucalyptus leaves extracted dye and mordanted with Rivina berries was provided in Table 8. The result was better for all mordantation and the rating was 4-5. Rivina berries mordanted fabrics using simultaneous mordanting is acceptable for good rubbing properties.

Measurement of color fastness to perspiration (ISO 105 E04:2013) :

Table-6. Measurement of colour fastness to wash

Mordanting methods	Mordants	Fabrics	Change in colour	Staining on adjacent fibers					
				Viscose	Acrylic	Polyester	Nylon	Cotton	Tri Acetate
Pre-mordanting	Rivina berries	Silk	4	4	4	4	4	4	4
		Wool	4	4	4	4	4	4	4
	Without Mordant	Silk	4	4	4	4	4	4	4
		Wool	4	4	4	4	4	4	4
Simultaneous mordanting	Rivina berries	Silk	4-5	4-5	4-5	4-5	4-5	4-5	4-5
		Wool	4-5	4-5	4-5	4-5	4-5	4-5	4-5
	Without Mordant	Silk	4	4	4	4	4	4	4
		Wool	4	4	4	4	4	4	4
Post-mordanting	Rivina berries	Silk	4-5	4-5	4-5	4-5	4-5	4-5	4-5
		Wool	4-5	4-5	4-5	4-5	4-5	4-5	4-5
	WM	Silk	4	4	4	4	4	4	4
		Wool	4	4	4	4	4	4	4

Table-7. Measurement of colour fastness to rubbing

Mordanting methods	Mordants	Fabrics	Staining on adjacent fibers	
			Dry	Wet
Pre-mordanting	*Rivina* berries	Silk	4	4
		Wool	4	4
	Without Mordant	Silk	4	4
		Wool	4	4
Simultaneous mordanting	*Rivina* berries	Silk	4-5	4
		Wool	4-5	4
	Without Mordant	Silk	4	4
		Wool	4	4
Post mordanting	*Rivina* berries	Silk	4-5	4
		Wool	4-5	4
	Without Mordant	Silk	4	4
		Wool	4	4

Table-8. Measurement of colour fastness to light

Mordanting methods	Mordants	Fabrics	Colour change
Pre- mordanting	*Rivina* berries	Silk	4-5
		Wool	4-5
	Without Mordant	Silk	3-4
		Wool	3-4
Simultaneousmordanting	*Rivina* berries	Silk	4-5
		Wool	4-5
	Without Mordant	Silk	4-5
		Wool	4-5
Postmordanting	*Rivina* berries	Silk	4-5
		Wool	4-5
	Without Mordant	Silk	4-5
		Wool	4-5

The action of alkaline and acidic perspiration to dyed fabric samples was illustrated in Tables-9. The colour fastness to perspiration shows that Rivina berries mordanted fabrics using simultaneous mordanting had good perspiration fastness and the rating was 4. Post mordatation also gave better results of rating about 4. The colour fastness to

Table-9. Measurement of color fastness to perspiration

Mordanting methods	Mordants	Fabrics		Change in colour	Staining on adjacent fibers					
					Wool	Acrylic	Polyester	Nylon	Cotton	Acetate
Pre-mordanting	*Rivina* berries	Silk	Acidic	3-4	3-4	3-4	3-4	3-4	4	3-4
			Alkaline	3-4	3-4	3-4	3-4	3-4	4	3-4
		Wool	Acidic	3-4	3-4	3-4	3-4	3-4	4	3-4
			Alkaline	3-4	3-4	3-4	3-4	3-4	4	3-4
	Without Mordant	Silk	Acidic	3-4	3-4	3-4	3-4	3-4	3-4	3-4
			Alkaline	3-4	3-4	3-4	3-4	3-4	3-4	3-4
		Wool	Acidic	3-4	3-4	3-4	3-4	3-4	3-4	3-4
			Alkaline	3-4	3-4	3-4	3-4	3-4	3-4	3-4
Simultaneous mordanting	*Rivina* berries	Silk	Acidic	4	4-5	4-5	4-5	4-5	4	4-5
			Alkaline	4	4-5	4-5	4-5	4-5	4	4-5
		Wool	Acidic	4	4-5	4-5	4-5	4-5	4	4-5
			Alkaline	4	4-5	4-5	4-5	4-5	4	4-5
	Without Mordant	Silk	Acidic	4	4-5	4-5	4-5	4-5	4	4-5
			Alkaline	4	4-5	4-5	4-5	4-5	4	4-5
		Wool	Acidic	4	4-5	4-5	4-5	4-5	4	4-5
			Alkaline	4	4-5	4-5	4-5	4-5	4	4-5
Post mordanting	*Rivina* berries	Silk	Acidic	4	4-5	4-5	4-5	4-5	4	4-5
			Alkaline	4	4-5	4-5	4-5	4-5	4	4-5
		Wool	Acidic	4	4-5	4-5	4-5	4-5	4	4-5
			Alkaline	4	4-5	4-5	4-5	4-5	4	4-5
	Without Mordant	Silk	Acidic	3-4	3-4	3-4	3-4	3-4	3-4	3-4
			Alkaline	3-4	3-4	3-4	3-4	3-4	3-4	3-4
		Wool	Acidic	3-4	3-4	3-4	3-4	3-4	3-4	3-4
			Alkaline	3-4	3-4	3-4	3-4	3-4	3-4	3-4

perspiration was good in simultaneous mordanted fabrics compared to other mordanting method.

Sustainable use of natural dyes in textile industry can be achieved through low-cost production of natural dyes, conserving biodiversity and utilization of mordant-like material from renewable resources. The suitability of *Rivina humilis* L. berries as a source of betalains rich bio-mordant for *Eucalyptus* leaves natural dye was confirmed. Eucalyptus has ample natural tannins and polyphenols, with the major colouring component. The colour strength (K/S > 5) using simultaneous mordanting on silk fabric, colour values (K/S > 4) on wool fabrics mordanted using rivina berries and excellent colour fastness (wash, light, rubbing and perspiration) were obtained for the simultaneous mordanted dyed fabrics than both pre-mordanting and post mordanting. Application of mordant in dyeing improved dyeing properties in simultaneous mordanting than both pre-mordanting and post mordanting. It was concluded that the color values with respect to K/S and L*, a*, b* values were

found to be influenced by the addition of mordants and also the eucalyptus dye. Rivina berries were found as the influential mordant with respect to color values. Mordants improved K/S values of the dyed fabrics except a few cases when compared to unmordanted dyed fabrics. The technique of simultaneous mordanting gave excellent to very good colour fastness to the dyed silk, wool fabrics compared to unmordanted sample. The substainable utilization of betalains rich rivina berries as a potential source of eco-friendly mordant alternative to synthetic dye in textile industry which has a remarkable environmental benefit.

Authors thank Director, Institute of Forest Genetics and Tree Breeding, Coimbatore for the support.

Conflict of Interest

The authors declare that there is no conflict of interest.

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