

The use of cochineal and *Monascus purpureus* as dyes for cotton fabric

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Received: 20 June 2011; Accepted: 8 August 2012

This study describes the dyeing efficiency of cochineal (*Dactylopius coccus* Costa) and *Monascus purpureus* colorants on cotton fabric. The effects of mordant, dyeing time and temperature on the shades and fastness properties of both colorants were studied. The colour coordinate values indicate that the cochineal colorant exhibited higher colour strength than *M. purpureus* for premordanted cotton fabric (4% o/wf). Optimum dyeing with *M. purpureus* was observed at a temperature of 80 °C for 80 min at a 30:1 liquor ratio. However, for cochineal, the optimum dyeing conditions were 60 °C for 60 min at a 40:1 liquor ratio. Furthermore, both colorants demonstrated moderate to good fastness properties. The results of the study indicate that cochineal has better dyeing properties than *M. purpureus* colorants.

Introduction

The textile processing industry is one of the major environmental polluters [1]. Recently, there has been growing interest in using natural colorants for textile dyeing [2,3]. The general belief is that natural colorants are more ecofriendly than synthetic dyes [4], exhibit better biodegradability and are more environmentally friendly [5]. In order to process 1 t of textiles, 230–270 t of water may be required. This water could pollute the environment throughout dyeing and processing. To overcome these limitations, an alternative is to use natural colorants or dyes. Most natural colorants have little intrinsic affinity for cotton fibres and must be used in conjunction with mordants. A mordant, usually a metallic salt, can itself be fixed on the fibre and also combines with the dyestuff. A link is formed by the mordant between the fibre and the dye, allowing the fixation of certain dyes that have little to no affinity for cotton fibre [6].

Cochineal colorants

The cochineal (*Dactylopius coccus* Costa) is an insect that is widely used as a natural colorant in the food industry [7]. The principal component of the colorant in this species of cochineal is carminic acid ($C_{22}H_{20}O_{13}$), which is a red glucosidal hydroxyanthrapurin that occurs naturally in some scale insects, and is approved for use as a food colorant in the European Union and the United States [8]. Several analytical methods have been reported for the determination of carminic acid using spectrometry [9,10],

enzyme immunoassay [11], capillary electrophoresis [12] and liquid chromatography [13–15]. The chemical structure of carminic acid consists of a core anthraquinone structure linked to a sugar unit (Figure 1a). It is believed that ca. 4000 years ago the Chinese applied these cochineal colorants to silk and leather materials [15–17].

Monascus purpureus colorant

The production and evaluation of microbial-based colorants as textile dyes are currently being studied. Many microorganisms that produce colorants are described in the literature [18]. Fungi are an ecologically interesting source as some species are rich in stable colorants such as anthraquinones, anthraquinone carboxylic acids and pre-anthraquinones [19]. The colorants produced by *Monascus* spp. are oligoketides which have been subdivided into three groups [20]. The orange components are rubropunctatin and monascorubrin, the red colorants come from the analogues rubropunctamine and monascorubramine and the yellow colorants come from the reduced forms of monascin and ankaflavin. There are a few studies concerned with how these colorants are obtained or their properties, such as stability, structural characteristics and so on, but generally those studies focus more on the biological than on the technological aspects of the colorants [21]. Within the technological realm, these colorants have been applied in a few studies for dyeing silk as described above, but few studies have focused on the comparison between the two available natural colorants [21].



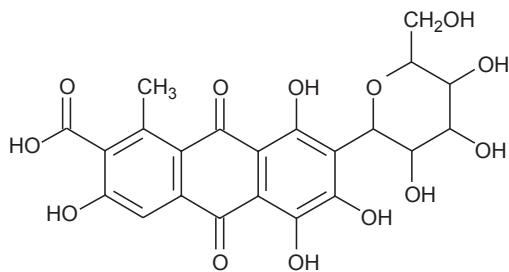


Figure 1 Structure of carminic acid

Therefore, the aim of the present study was to investigate the dyeing efficiency of *Monascus* colorant on cotton fabric and compare it with that of cochineal. Furthermore, factors affecting dyeing and fastness properties were assessed.

Materials and Methods

Dyeing of cotton fabric

Industrially bleached woven fabric (100% cotton) was purchased from the South Indian Textile Research Association (SITRA) in Coimbatore, India. The characteristics of the raw cotton fabric were as follows: thickness under 1 kPa pressure, 0.27 mm; yarn number, wrap 14 tex, weft 16.5 tex; fabric counts, wrap 31 threads/cm, weft 35 threads/cm; weight, 115 g/m². To remove the wax and other impurities, cotton yarn (100 g) was placed in boiling water (2 l) to which soap flakes (*ca.* 7 g) and sodium carbonate (3 g) were subsequently added. The mixture was then boiled for 1 h. The cotton fabric was then removed, washed with hot water and cold water in order to avoid breakdown of the emulsion and precipitation of the impurities onto the cotton fabric, squeezed to remove excess liquor and air dried. Finally, the fabric was treated with 1 M hydrochloric acid (HCl) (*ca.* 2 l) at room temperature for 30 min, removed, washed with deionised water until the rinsed water was neutral and dried at room temperature [22]. The dried fabric was subjected to dyeing according to the conditions established by Chairat *et al.* [22].

Extraction of colorant from cochineal

Adult *D. coccus* females were collected in and around Coimbatore, Tamilnadu, India, from several wild cactus pears growing around the wastelands. The cochineals were cleaned over a sieve to eliminate dust and heterogeneous materials such as moult residue or plant material. The insects were then dried at 60 °C in an oven. The dried insects were finely ground in a ceramic mortar, and an amount accurately weighed at *ca.* 0.5 g was mixed with 100 ml of methanol:water (65:35, v/v). The mixture was homogenised in a locally made hand homogeniser for 5 min, and colorant present in the sample was extracted for 30 min in a water bath at 80 °C in a closed container. The sample was cooled and centrifuged at 2191 × g for 15 min. This procedure was repeated twice, and the resulting two supernatants were mixed together. The collected supernatants were concentrated in a rotary evaporator (Büchi, Canada) and lyophilised to obtain a powder [15,23]. The structure of this colorant is shown in Figure 1.

Extraction of colorant from *Monascus purpureus*

M. purpureus culture (MTTC-410) was purchased from the Microbial Type Culture Collection and Gene Bank in

Chandigarh, India. To extract the colorant, *M. purpureus* was cultured in a 5 l Erlenmeyer flask containing 3 l of Czapek–Dox (India) synthetic medium. The flask was incubated for 5 days at room temperature in stagnant position. Following incubation, the mycelium was harvested and the supernatant was filtered through a sterilised muslin cloth. Later, two volumes of 95% (v/v) ethanol and exhausted culture broth were added according to the following procedure: (i) after dilution with *ca.* 60% of the required solvent volume, the resulting mixture was kept on the rotary shaker at 180 rpm at 30 °C for 30 min; (ii) the ethanolic mixture was then centrifuged at 3780 × g for 15 min; (iii) once the supernatant had been recovered, the residue was dispersed in the remaining volume of ethanol and centrifuged again at 3780 × g for 5 min; (iv) the supernatants were then collected and filtered through a preweighed Whatman (USA) GF/C disc (47 mm) and further diluted with 95% (v/v) ethanol to a final volumetric dilution factor of 20, and the scanned spectrum was observed at 350–500 nm.

The colorant was purified according to the method outlined by De Santis *et al.* [24]. Purified colorants were concentrated in a Büchi rotary evaporator and lyophilised to obtain a powder. Using a U-2000 spectrophotometer (Hitachi, Japan), the optical density (OD) was measured at 500 nm [25] and multiplied by the above dilution factor, thus yielding an expression for the produced red colorant in units of absorbance ($UA\lambda$) at a given wavelength (λ_{max}).

Optimisation of the dyeing conditions

Seven samples of cotton fabric with a liquor ratio (LR) of 30:1, each weighing 2 g, were dyed in seven separate baths for 1 h each at 30, 40, 50, 60, 70, 80 or 100 °C. The dyeing was carried out in the presence of 2 g/l of sodium sulphate (Na_2SO_4) to promote exhaustion, without adjusting the dyebath pH to avoid altering the colour of the dye. To study the effect of dyeing time, another set of six samples was dyed at the optimised conditions for 30, 40, 80, 100, 120 and 140 min. The optimum electrolyte concentration for the best degree of exhaustion was determined by six additional dyeing experiments at the optimised temperatures (80 and 90 °C) and times (60 and 120 min) with 0, 1, 2, 4, 6, 8, 10 and 12 g/l of Na_2SO_4 respectively. The optimum LR was determined using nine additional samples dyed at the optimised temperatures (80 and 90 °C), times (60 and 120 min) and electrolyte concentrations (without salt) using LR of 20:1, 25:1, 30:1, 35:1, 40:1, 45:1, 50:1, 55:1 and 60:1.

Mordanting

The cotton fabric samples were subjected to premordanting with alum at 4, 6, 8, 10, 12, 14, 16, 18 and 20% owf for 80 and 60 min at 80 and 90 °C with LR of 40:1 and 30:1 for *M. purpureus* and cochineal colorants respectively. Post-mordanting was also performed for fabric dyed with both colorants using 10% owf of alum in order to study the effect on colour strength and fastness properties under optimised conditions. Furthermore, all the dyeing and mordanting experiments were replicated.

Dyeing

Dyeing was carried out using the *M. purpureus* and cochineal colorants with an optimised colorant concen-

tration in water alone for 60 and 120 min at 80 and 90 °C, with no salt added and a LR of 1:30 and 1:40 respectively. Alum was used as the mordant (4% premordanting and 10% post-mordanting). The dyeing was carried out with a fixed colorant concentration of 5% owf. After dyeing, the samples were washed and dried. Colorant uptake was determined by measuring the absorbance of diluted dyebath samples at the wavelength of maximum absorbance (λ_{max}) of the colorant using a U-2000 spectrometer (Hitachi). The percentage of dyebath exhaustion (%E) was calculated using the following formula:

$$\%E = [(A_0 - A_1/A_0)] \times 100$$

where A_0 and A_1 are the absorbance of the dyebath before and after dyeing at λ_{max} of the colorant used, respectively.

Colour measurement

CIELAB colour coordinates of dyeings were measured using a SF 600 spectrophotometer (Datacolor, Switzerland). Repeated measurements were performed in order to obtain mean values for L^* , a^* , b^* , C^* and h .

Fastness testing

Samples dyed using the optimised conditions were washed under specified conditions to simulate five home washings before evaluation. Wash fastness was tested according to AATCC test method 61-1996 [40]. A crockmeter was used to measure the wet rubbing fastness of the dyed samples according to ISO 105-D02:1993 [26]. Light fastness was measured according to ISO 105-B02:1994 by comparison with the greyscale [27]. The dyed samples were exposed to a xenon arc lamp for 3 days under standard testing conditions.

Statistical analyses

Exhaustion was evaluated by analysis of variance, and each treatment was replicated three times. The standard deviation was calculated for each trial.

Results and Discussion

The mycelial growth was measured and found to be 2.2 ± 1.9 g/l. Although a clear interpretation of the dyeing tests would require fractionation of the *Monascus* red colorants, such an operation was regarded as too expensive for the specific novel application. Here, an economical method of fungal colorant extraction was used. Briefly, the fermentation broth was exhausted and simply diluted with ethanol to dissolve all the suspended colorants. The suspension was refined by centrifugation to remove microbial biomass before using the end product for dyeing.

Effect of temperature

The effect of temperature on colour strength in cotton fabrics dyed with cochineal and *M. purpureus* colorants was examined at different temperatures (20–100 °C), and the results are presented in Figure 2. In general, the colour uptake of cochineal and *M. purpureus* colorants increases as the temperature increases. The maximum exhaustion (83%) for the *M. purpureus* colorant was observed at 80 °C, whereas the cochineal colorant exhibited a maximum activity (86%)

at 90 °C. The mobility of the large dye ions increased at higher temperatures (80 and 90 °C), and subsequently the dyeing rates were also enhanced. The results are consistent with those of a previous study by De Santis *et al.* [24], who reported the maximum dyeing of *Monascus* colorant at 80 °C. There was increased cotton fibre swelling at a higher temperature and less aggregation of colorant, which might augment dye diffusion. This permits additional dye molecules to penetrate into the fabric at high temperature [28]. Moreover, increasing temperature increases the rate of dyeing. In addition, the anomalous behaviour in the temperature range 20–70 °C may not be a result of the propensity of both colorants for surface colouration and colorant aggregation, which yields lower colour strength in this temperature range [5,29]. Achieving temperatures above 90 °C is not always feasible owing to undesired side effects such as fabric damage. However, the amount of colorant uptake by the cotton fabric decreases with increasing temperature, which is indicative of an exothermic process [30,31].

Effect of dyeing time

As shown in Figure 3, the colour strength increases as time progresses for both cochineal and *M. purpureus* colorants. The maximum exhaustion (90%) for the cochineal colorant was obtained at 60 min and decreased for the remaining time up to 140 min. However, in the *M. purpureus* colorant, exhaustion of 85% was obtained at 80 min and gradually decreased up to 120 min. In the case of the cochineal colorant, a plateau in exhaustion was attained from 60 min up to 80 min, and then exhaustion declined slightly as time progressed. When using the *M. purpureus* colorant, a rapid decrease in colour strength was observed after 80 min. The decline in exhaustion might be attributed to desorption of the dye molecules as a consequence of the prolonged dyeing time. The shift in equilibrium may be responsible for the reduced uptake above 60 min (cochineal) and 80 min (*M. purpureus*) owing to colorant aggregation on the surface of the fibre, resulting from the extended dyeing time and the high concentration of colorant remaining in the dyebath.

Effect of salt concentration

Exhaustion in this study was better in the absence of salt. As shown in Figure 4, the highest exhaustion (89% and 83%) was obtained in the control without salt for both colorants. According to Guesmi *et al.* [23], Kamel *et al.* [33] and Ali and El-Mohamedy [34], as the salt concentration increases, the colour strength decreases. The authors also noted that, when no salt was added, exhaustion was at its maximum. Salt is usually required for dyeing cotton with highly anionic dyestuffs such as direct and reactive dyes. The cochineal and *M. purpureus* dyes in this study are not likely to form highly anionic species in aqueous solution. Also, salt can cause the precipitation as well as the aggregation of dyes in solution, which will lead to reduced dye uptake [5,35].

Effect of liquor ratio

The effect of LR on colour strength is shown in Figure 5. Maximal exhaustion was obtained at LR 30:1 for *M. purpureus* (81%) and 40:1 for the cochineal colorant (86%). These results indicate that exhaustion strongly depends upon LR. As LR increases, there is a propensity for dyes simply to remain in the aqueous phase rather than being adsorbed.

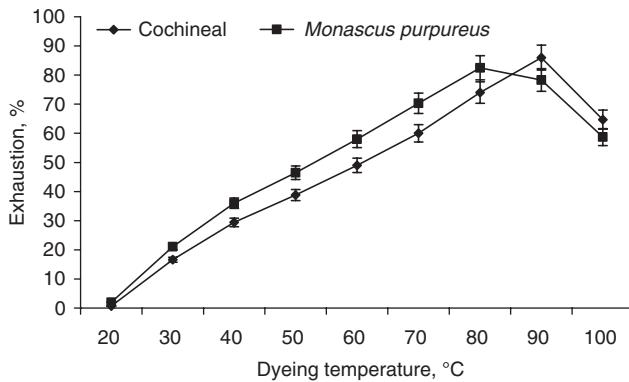


Figure 2 Effect of dyeing temperature on dyebath exhaustion

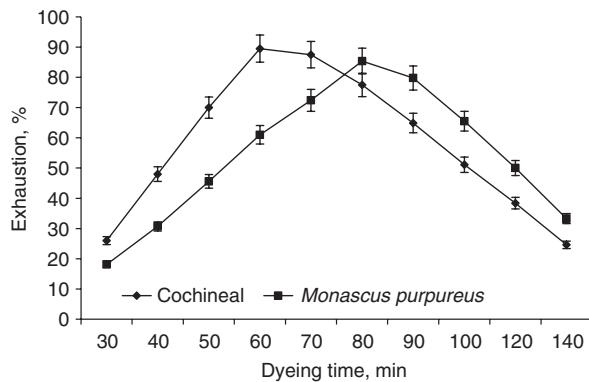


Figure 3 Effect of dyeing time on dyebath exhaustion

The decreased colour strength at the lower LR is because of precipitation of colorant.

Effects of mordanting

The colour of a dye can be altered by treatment with metal salts. Thus, the effects of premordanting and post-mordanting with alum on L^* , a^* , b^* , C^* and h at optimised conditions for cochineal and *M. purpureus* colorants were investigated. The results are presented in Table 1: L^* is the lightness value, positive values of a^* and b^* represent redder and yellower tones and negative values represent greener and bluer tones respectively; C^* is the chroma. Based on the results of the lightness values, the change in hue increased with the addition of alum (premordanting 4% o/wf) at 80 °C cochineal and 90 °C, *M. purpureus*, while it decreased in all other cases. Moreover, there was also a significant decrease in C^* when a higher percentage of alum (10% o/wf) was used for mordanting [36].

Fastness properties

The fastness properties of the cotton samples dyed under optimised conditions either without mordant or with premordanting or post-mordanting are presented in Table 2. There was no significant difference in the washing, rubbing and light fastness properties in the majority of cases. However, with respect to washing and rubbing, the fastness properties are lower in the case of post-mordanting by comparison with premordanting with alum for both colorants. The wash fastness of fabric dyed without mordant and with mordanting is shown in Table 2. These results were assessed in the typical manner in terms of

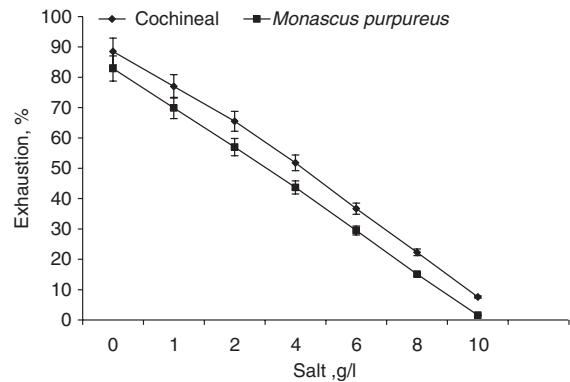


Figure 4 Effect of salt concentration on dyebath exhaustion

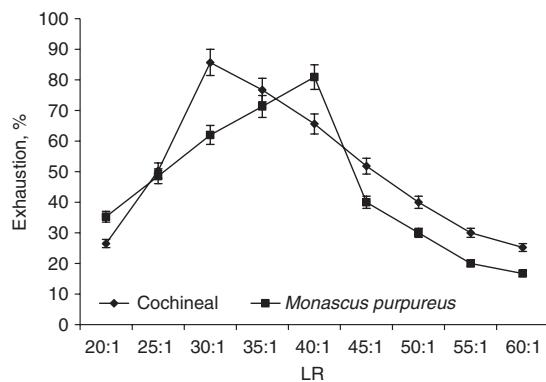


Figure 5 Effect of LR on dyebath exhaustion

greyscale values for the staining of the adjacent cotton material and alterations in shade. Table 2 also shows the rubbing fastness of samples dyed with cochineal and *M. purpureus* colorants in the presence and absence of mordant. Both types of colorant exhibited a fair to good rating in rubbing fastness. This observation is probably due to the fact that both colorants penetrate equally into the cotton fabric [9]. Interestingly, premordanted cotton fabric exhibited a rubbing fastness rating one level higher at good to very good. The increased rating was possibly due to the

Table 1 Colorimetric properties of the dyeings

Dyeing	Colour coordinates				
	L^*	a^*	b^*	C^*	h
Raw cotton	88.83	-0.25	4.59	38.26	84.94
Dyed with cochineal colorant	79.80	11.72	4.82	42.92	45.72
Dyeing with <i>M. purpureus</i> colorant	81.48	0.20	1.19	38.21	42.54
Cochineal, premordanted with 4% alum	59.26	15.14	6.12	36.22	32.31
<i>M. purpureus</i> , premordanted with 4% alum	68.20	0.40	9.04	26.16	22.81
Cochineal, post-mordanted with 10% alum	62.60	19.16	-0.21	22.18	28.12
<i>M. purpureus</i> , post-mordanted with 10% alum	76.36	0.12	2.02	33.16	88.14

Table 2 Fastness properties of cotton dyed with cochineal and *Monascus purpureus* colorants with and without mordanting

Dyeing	Wash fastness		Rubbing fastness		
	Colour change	Staining of cotton	Dry	Wet	Light fastness
Dyed with cochineal colorant	3–4	4	3–4	3–4	4–5
Dyeing with <i>M. purpureus</i> colorant	2–3	3–4	2–3	2–3	4
Cochineal, premordanted with 4% alum	4	3–4	4	3	4
<i>M. purpureus</i> , premordanted with 4% alum	4	3–4	4	3	3–4
Cochineal, post-mordanted with 10% alum	3–4	3–4	3–4	2–3	4
<i>M. purpureus</i> , post-mordanted with 10% alum	3–4	3–4	3–4	2–3	3–4

enhanced penetration of colorant into the fabric. The light fastness values of fabric dyed with the cochineal and *M. purpureus* colorants with and without mordant are shown in Table 2. The dyeings derived from *M. purpureus* were slightly less fast to light compared to those of cochineal. Mordanting (pre- or post-) also led to a small reduction in photostability. The higher light fastness properties of the cochineal colorant can be attributed to strong intramolecular H-bonding [37]. These bonds enhance the stability of the compound by reducing the electron density at the chromophore. As a result, the sensitivity of the colorant towards photochemical oxidation is reduced. These results indicate that samples dyed without pre-mordant have better light fastness when compare to samples dyed with post-mordant; this might be due to improved colorant aggregation on the fabric [38,39].

Conclusion

In this research, the dyeing properties of cochineal and *M. purpureus* colorants were studied and the dyeing conditions were optimised. When compared with the fungal colorant, the cochineal colorant was found to have much higher colour strength on cotton fabric. In the case of pre- and post-mordanting with alum, there was no significant change in hue, but there was a significant decrease in chroma. Shades of the dyeings ranged from pink to light red. Compared with the fabric dyed with the cochineal colorant, fabric dyed with the *M. purpureus* colorant was duller. In most cases, pre- and post-mordanting with alum resulted in small changes in the fastness properties. The use of mordants in the dyeing of cotton fabric resulted in an increase in depth of shade for both colorants. Premordanting yielded the deepest shades, with superior wash and rubbing fastness. Mordanting led to a half-point reduction in light fastness ratings.

Acknowledgements

LPS would like to thank the South Indian Textile Research Association (SITRA) for their technical assistance, and P Senthilkumar of the Department of Textile Technology, PSG College of Technology, Coimbatore, India.

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