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ORIGINAL ARTICLE

Eco-dyeing using *Tamarindus indica* L. seed coat tannin as a natural mordant for textiles with antibacterial activity

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Abstract Tamarind seed coat tannin was extracted and its tannin class was determined. The extracted tannin was employed as a natural mordant alone and in combination with metal mordant namely copper sulphate for cotton, wool and silk fabrics and dyed using natural dyes namely turmeric and pomegranate rind. The colour strength, colour coordinates, wash and light fastness were evaluated and compared for all the three fabrics with and without mordanting. The pre-mordanted fabrics on dyeing gave better colour strength, wash and light fastness than those dyeing obtained without mordanting. The total phenolic content of the extract was calculated and minimum inhibition concentration was 1% against both the *Staphylococcus aureus* and *Escherichia coli* bacteria. The mordanted and dyed fabrics resulted in good antibacterial activity up to 20 washes, when natural mordant was used along with 0.5% and 1% copper sulphate mordant and dyed with natural dyes.

1. Introduction

Tamarindus indica L., commonly known as tamarind tree, is one of the most important multipurpose tree species in the Indian sub-continent. It belongs to the family Caesalpinioideae and almost all the parts of this tree are found to be of some use. The use of fruit pulp has been known for a very long time. Other uses are in food, chemicals and pharmaceuticals (Dagar et al., 1995).

The seed consists of 30% hard but brown seed coat and 70% kernel. The kernels are separated from the seed coat either by roasting or by soaking the seeds in water. The boiled or roasted kernels are eaten and sometimes dried and grounded into flour for making pan-cakes or mixed with rice. The kernels contain polysaccharides (jellose) having a very good sizing property or employed as a sizing agent for jute and textile yarns (The Wealth of India, 2003). Tamarind seeds inhibit the growth of urinary crystals and are used in the treatment of patient's recurrent kidney stones (Natarajan et al.,

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1997). The seed extract is also reported to contain phenolic antioxidants (Tsuda et al., 1993, 1994) and antimicrobial activity (De et al., 1999). The seed coat containing 40% water soluble matter is nothing but a mixture of tannins. It is used for wound healing and as an anti-dysenteric drug. It is also used as a raw material for the preparation of plywood adhesives. With a particular astringent taste, the profile of polyphenolics present in the tamarind seed coat was found to be dominated by proanthocyanidins, i.e., a group of compounds formed by the condensation or polymerization of flavan-3-ol or flavan-3, 4-diol, commonly known as condensed tannins or phlobatannin (Sydjaroen et al., 2005).

Tannin is an astringent vegetable product found in a wide variety of plant parts such as bark, wood, fruit, fruit pods, leaves, roots and plant galls. Tannins are defined as naturally occurring water soluble polyphenolic compounds of high molecular weight (about 500–3000) containing phenolic hydroxyl groups to enable them to form effective crosslinks between proteins and other macromolecules (Ramakrishnan et al., 2006). They are primarily used in the preservation of leather (Swarna et al., 2009), glues, stains and mordants. Meanwhile, natural fibres such as cotton have very low affinity for most of the natural dyes. The tannins play an important role in cotton dyeing to retain colouring matter permanently. The ultimate aim, the purpose of preparing the vegetable fibres with tannin is not so much to fix the colouring matter, as to fix certain metallic salts such as copper, iron, etc., in the form of insoluble tannates. The metal tannates present on the material forms insoluble lakes with the natural dyes during the dyeing process and results in improved fastness properties (Gulrajani, 1999).

Recently, due to increasing awareness of environmental issues and also pollution produced by synthetic dyes, wide spread interest has emerged in the dyeing of textile fibres using natural colourants on account of their better biodegradability and higher compatibility (Ali and El-Mohamedy, 2011; Bechtold et al., 2003) with the environment, relatively low toxicity, allergic reactions. However, most research on natural dyes had been focused on the fundamental aspects of the natural dyes, e.g. the property of dyeing, light fastness and washing fastness (Feng et al., 2007). Little attention has been given to the other functions of the natural dyed materials such as antibacterial properties. Almost all the natural textile material made of cotton, wool, silk, etc., are susceptible to microbial attack as these fabrics offer larger surface area and absorb moisture, thus providing a suitable atmosphere for microbial growth and reproduction. It is also observed that micro-organisms cause degradation of the polymer chains of textile material bringing down the strength and the durability of such products. Hence textiles finished with antibacterial finishes are preferred by the modern consumer.

The findings of this work provide a new area of utilization of large quantities of seed coat which are available as residual material during the production of tamarind kernel powder, which is used as a sizing agent. The objectives of this study were to extract tannin from tamarind seed coat and its application as a natural mordant alone and in combination with metal mordant, namely copper sulphate for dyeing cotton, wool and silk fabrics using natural dyes namely turmeric and pomegranate rind. The tannin class, total phenolic content of the extract were determined and characterized. The colour strength, colour coordinates, wash and light fastness were evaluated for all three substrates with and without mordanting. The minimum inhibition concentration (MIC) of extracted tannin for antibacterial activity against Gram-positive bacteria *Staphylococcus aureus* and Gram-negative bacteria *Escherichia coli* were determined qualitatively. Further, the antibacterial activities of mordanted and natural dyed fabrics against both bacteria were evaluated quantitatively and the durability of activity for repeated washing was investigated.

2. Experimental

2.1. Materials

The cotton fabric (124GSM) was purchased from Premier Mills, Mumbai, India, and was soaked in distilled water and then treated with non-ionic detergent solution containing 2 g/L each of soap and soda ash at 80 °C for 60 min to remove starch and other stiffening agents. Ready for dyeing wool (164 GSM) and silk (42 GSM) fabrics were purchased from Kiran Threads, Vapi, India. *Curcuma longa L.* (turmeric) and *Punica granatum L.* (pomegranate rind) were obtained in powder form from local ayurveda market in Mumbai, India. *Tamarandius indica L.* (tamarind) seeds were collected from Thirubuvanam, Tamilnadu, India. Copper sulphate, ferric chloride, gelatine, lead acetate, hydrochloric acid, sulphuric acid and sodium chloride of analytical grade were supplied by S.D. Fine Chemicals, Mumbai. Non-ionic detergent (Auxipon NP) was supplied by Auxichem Ltd., Mumbai. For microbiological testing, Gram-positive bacteria *Staphylococcus aureus* (*S. aureus*) (NCTC 3570) Gram-negative bacteria *Escherichia coli* (*E. coli*) (ATCC 10148) were procured from Haffkine's Institute, Parel, Mumbai, India.

2.2. Methods

2.2.1. Separation of tamarind seed coat for extraction of tannin

The tamarind seeds were dried in a hot oven at 100 °C for 30 min and the coat was removed from the seeds by manual crushing and separated and then ground into powder form.

2.2.2. Tannin extraction procedure

The finely powdered tamarind seed coat (1000 g) was extracted with water (2 L) for 2 h at boil and after cooling it was filtered through a fine muslin cloth and the filtrate was collected separately. The remaining residue was extracted three more times, in order to complete the extraction. The total extract (8 L) was heated to boil and was allowed to stand overnight and filtered again. The clear filtrate was concentrated in a water bath and treated with saturated brine solution. A brownish coloured precipitate thus obtained was filtrated and dried in an oven to yield brown coloured tamarind seed coat tannin in powder form.

2.2.3. Determination of tannin class

The qualitative analysis (Russell, 1935) was carried out by treating 0.5% solution of the above tannin product with various reagents such as aqueous ferric chloride, gelatine, lead acetate and copper sulphate solution. The colour change after the addition of reagent was observed.

2.2.4. Determination of total phenolic content

The total phenolic content (TPC) in both the extracts was determined by the Folin–Ciocalteu method (Singleton et al., 1999). The extract was taken in 20 μL each into separate test tubes, 1.58 mL of water was added followed by the addition of 100 μL of the Folin–Ciocalteu reagent, and mixing. It was allowed to stand for 8.5 min and then 300 μL of the sodium carbonate solution was added and shaken to mix. The solutions were left to stand at 20 °C for 2 h and the absorbance of each solution at 765 nm against the blank (the "0 ml" solution) was determined. All estimations were carried out in triplicate. The calibration curve was plotted using concentrations of 0, 50, 100, 150, 250, 500 and 1000 mg/L gallic acid. The total content of phenolic compounds in the extracts in gallic acid equivalents (GAE) was calculated by the following formula:

$$T = C \cdot V/M$$

where T is the total content of phenolic compounds, mg/g plant extract, in GAE; C is the concentration of gallic acid established from the calibration curve, mg/L; V is the volume of extract, mL; M is the weight of the plant extract, g.

2.2.5. Extraction of colourants

Turmeric and pomegranate rind colourants were extracted using water as the medium. Six grams of the colourants in the form of fine powder were mixed with 300 ml of water and allowed to soak for 30 min and the mixture was then boiled for 60 min. The contents were cooled and filtered. The filtrate was used for dyeing.

2.2.6. Mordanting

Pre-mordanting technique was used for this study. Cotton, wool and silk fabrics were treated with tamarind seed coat mordant solution using 15% concentrations on weight of fabric (owf) and this treatment was carried out at 95 °C for 45 min, keeping the liquor to material ratio 20:1. The cotton, wool and silk fabrics mordanted with 15% tamarind seed coat mordant concentration were further treated with 0.5% and 1% (owf) copper sulphate (Cu) solution at 85 °for 45 min, keeping the liquor to material ratio as 20:1. Cotton, wool and silk fabrics, only mordanted with tamarind seed coat mordant and in combination with copper sulphate mordants, were squeezed and subjected to dyeing.

2.2.7. Dyeing method

Shade used for dyeing on cotton, wool and silk fabrics was 20% (owf). Dyeing was carried out in Rota Dyer machine, Rossari® Labtech, Mumbai, keeping the liquor to material ratio of 20:1. Pre-mordanted fabrics were introduced into the dyeing solution at room temperature and slowly the temperature was raised to 85 °C. The dyeing was continued at this temperature for 60 min. After dyeing, the fabrics were rinsed and air-dried.

2.2.8. Evaluation of colour strength

Dyed samples were evaluated for their colour strength by determining K/S values using a Spectra Scan® 5100+, Computer Color Matching system supplied by Perimer Colorscan, Mumbai, India. An average of four readings taken at four different sample areas was used to get the reflectance values, and Kubelka Munk function (K/S) was calculated (Etters and Hurwitz, 1986).

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

where R is the reflectance at complete opacity; K is the absorption coefficient; S is the scattering coefficient. Dyed fabrics were simultaneously evaluated in terms of CIELAB colour space (L^*, a^* and b^*) values using the Perimer Colorscan. In general, the higher the K/S value, the higher the depth of the colour on the fabric. L^* corresponding to the brightness (100 = white, 0 = black), a^* to the red–green coordinate (+ve = red, −ve = green) and b^* to the yellow–blue coordinate (+ve = yellow, −ve = blue). As a whole, a combination of all these enables one to understand the tonal variations.

2.2.9. Determination of minimum inhibition concentration (MIC) of the extract

The effect of concentration of tamarind seed coat tannin extract for the antimicrobial activity was assessed against Gram-positive bacterium *S. aureus* and Gram-negative bacteria *E. coli*. Five concentrations of the extract, i.e. 0.5%, 1% 5%, 10% and 15% were prepared. A loopful of *S. aureus* (1 mL) was mixed separately in 20 mL saline solution. One millilitre of saline containing the culture was added to a sterilized petri dish and was mixed thoroughly with 20 mL appropriately cooled nutrient agar (containing 1% agar-agar). This solution was allowed to solidify and a single well of 10 mm diameter was made in each of the petri dishes by using a cock-borer. The extracts (30 μL) of 0.5%, 1%, 5%, 10% and 15% concentrations were added in each well using a micropipette. The petri dishes were incubated for 24 h at 37 °C and the zone of inhibition measured. The above procedure was repeated for *E. coli* bacteria. An inhibition zone > 2 mm indicates good anitmicrobial property (Deepti et al., 2004).

The incubated plates were examined for a clear zone of inhibition. The width of the zone of inhibition was calculated using the following equation:

$$W = T - D/2$$

where W is the width of the zone of inhibition (mm); T is the total diameter of the test specimen and clear zone (mm) and D is the diameter of the well (mm).

2.2.10. Determination of antimicrobial activities of mordanted and dyed fabrics

Quantitative assessment of antimicrobial activity exhibited on mordanted and dyed cotton and silk fabrics against both micro-organisms were carried out by AATCC Test 100-2004 (AATCC, 2007a). To evaluate the antimicrobial activities of the treated fabrics, the reduction in the number of bacterial colonies formed with respect to the untreated control after incubation (37 ± 1 °C, 24 h) was determined. The percentage reduction was calculated with the following equation:

$$\text{Reduction rate}\,(\%)\; R = (B - A)/B \times 100$$

where R is the % reduction in bacterial count; A is the number of bacterial colonies recovered from the inoculated treated test specimen swatches in the jar incubated for 24 h contact period; B is the number of bacterial colonies recovered from the inoculated untreated control test specimen swatches in the jar immediately after inoculation (at “0” contact time).

2.2.11. Durability of antibacterial activity to washing

The samples were washed in a Launderometer by using Standard Test Method ISO 105-C06 A1M. One wash of the standard method is equivalent to five home launderings. The specimen size (100×40 mm) was first transferred to a pot containing soap solution (Auxipon NP non-ionic detergent) 4 g/L and 10 steel balls. The temperature was raised to 40 °C and washing was carried out for 45 min. Four washes were given one by one. The samples were then dried in air and the fabric was tested for antibacterial activity as mentioned above.

2.2.12. Fastness properties

Dyed fabrics were tested for colourfastness to washing according to the ISO Method, ISO 105-C10 (ISO, 2006). A solution containing 5 g/L soap solution was used as the washing liquor. The samples were treated for 45 min at 50 °C using liquor to material ratio of 50:1 in rota machine. After rinsing and drying, the change in colour of the sample was compared with the standard grey scale (rating 1–5, where 1 – poor, 2 – fair, 3 – good, 4 – very good and 5 – excellent).

Dyed fabric was tested for colourfastness to light according to AATCC Test Method 16-2004 (AATCC, 2007b). The light fastness was determined using artificial illumination with Xenon arc light source, Q-Sun Xenon Testing Chamber with black standard temperature = 65 °C with relative humidity of the air in the testing chamber as 40% and daylight filter, wavelength, λ = 420 nm. The samples were compared with the standard scale of blue wool reading (ratings, 1–8, where 1 – poor, 2 – fair, 3 –moderate, 4 – good, 5 – better, 6 – very good, 7 – best and 8 –excellent).

3. Results and discussion

3.1. Qualitative analysis of the extract

In order to find the class of tannin obtained from tamarind seed coat, a set of qualitative experiments were carried out with various reagents and their results are given in Table 1. The result clearly indicates that the tamarind seed coat extract gave identical colour and precipitation reaction when compared with the phlobatannins or condensed tannins as mentioned in the literature (Russell, 1935). It produced a dark green precipitate with aqueous ferric chloride and dark red solids with dilute HCl solution. It clearly confirms that tamarind seed coat extract is of condensed tannins.

Table 1 Qualitative analysis and its characteristic colour change with reagents.

Reagent	Observations
Aqueous ferric chloride	Dark green precipitate
Gelatin	Dirty white precipitate
Lead acetate	Pinkish precipitate
Copper sulphate solution	Faint green
Dilute hydrochloric acid solution	Dark red solids
Dilute sulphuric acid solution	Flesh coloured precipitate

3.2. Total phenolic content

Tannins are made of polyphenolic compounds and they exist widely in plants. In order to determine the total free phenolic group, i.e., total soluble phenols present in the extract, the colorimetric assay (Folin–Ciocalteau method) was carried out. The total phenolic content of the extract was 365.12 mg/g gallic acid equivalent, respectively. The extracts confirmed the presence of phenolic (–OH) groups and thus can be effectively used as mordants for natural dyes on all the three substrates.

3.3. Effect of mordanting, dyeing and its colour characteristics

Results with respect to colour depth, i.e., K/S values of dyeing of cotton, wool and silk fabrics with natural dyes namely turmeric and pomegranate rind (shade, 20%) obtained using with and without mordants are given in Tables 2–4, respectively. The results indicate that the K/S values of the cotton, wool and silk fabrics pre-mordanted with tamarind seed coat mordant and dyed with natural dyes were higher than that of only natural dyed cotton, wool and silk fabrics. The enhancement in the depth of dyeing was due to the presence of tamarind seed coat tannin acting as the mordant containing phenolic hydroxyl group which forms a complex with the dye molecules and leads to higher fixation of dye on the fabric. Also, it can be observed that the K/S values of all three naturally dyed fabrics increased highly with subsequent mordanting with copper sulphate mordant solution of 0.5% and 1% (owf) after tamarind seed coat mordanting for both natural dyes. Here, the enhancement in the colour depth was due to the presence of copper sulphate, a metal mordant which forms insoluble metal tannates with tamarind seed coat tannin phenolic hydroxyl group and further the metal tannates present on the fabrics form an insoluble lake with the reactive group of the dye molecules and lead to further higher fixation of natural dyes on the fabrics. Overall, the order of colour strength (K/S value) of mordanted and dyed fabrics was in the following sequence “wool > silk > cotton”. This may be attributed to wool having more functional groups than silk and cotton.

The colour coordinates L^*, a^* and b^* values, results obtained for cotton, wool and silk fabrics pre-mordanted with tamarind seed coat alone and in combination with copper sulphate mordant and dyed with natural dyes are given in Tables 2–4, respectively. In all pre-mordanted and naturally dyed cotton, wool and silk fabrics, the brightness or L^* values decreased highly resulting in deepening of shades as compared to only naturally dyed samples. From a^* and b^* values, the incorporation of tamarind seed coat mordant alone and in combination with copper sulphate mordant onto cotton, wool and silk fabrics produced good improvement and their values were positive and thus showed shifts in their tones resulting in beautiful colours as compared to only naturally dyed cotton, wool and silk fabrics and the colours obtained are mentioned in Tables 2–4, respectively.

3.4. Minimum inhibition concentration (MIC) of tamarind seed coat mordant

The antibacterial activity of the tamarind seed coat mordant was tested at five concentrations against the two bacteria

Table 2 *K/S* values, colour coordinates and colour of natural dyes dyed cotton fabric with and without mordanting.

Natural dye (shade, 20%)	Mordants (%)	*K/S* value	Colour coordinates			Colour obtained
			L^*	a^*	b^*	
Turmeric	Nil	1.6690	83.76	−2.13	50.01	Light yellow
	15% T	1.9328	75.93	0.51	41.36	Deep yellow
	15% T + 0.5% Cu	4.1459	69.85	2.19	48.36	Dark greenish yellow
	15% T + 1% Cu	5.0191	68.01	3.16	50.97	Dark greenish yellow
Pomegranate rind	Nil	0.8726	74.02	3.26	19.97	Beige
	15% T	0.9799	73.51	4.19	21.27	Reddish beige
	15% T + 0.5% Cu	1.5505	64.07	6.33	15.59	Deep reddish brown
	15% T + 1% Cu	2.0238	62.74	7.71	17.68	Deep reddish brown

T – tamarind seed coat mordant; Cu – copper sulphate; L^*: lightness (0 = black, 100 = white), L = lightness; $a^*\pm$ = red/green; $b^*\pm$ = yellow/blue.

Table 3 *K/S* values, colour coordinates and colour of natural dyes dyed wool fabric with and without mordanting.

Natural dye (shade, 20%)	Mordants (%)	*K/S* value	Colour coordinates			Colour obtained
			L^*	a^*	b^*	
Turmeric	Nil	7.0037	72.32	3.62	62.11	Bright yellow
	15% T	8.5428	63.04	5.39	49.61	Dark yellow
	15% T + 0.5% Cu	10.529	57.29	5.67	48.39	Deep greenish yellow
	15% T + 1% Cu	11.843	53.72	4.23	45.03	Dark greenish yellow
Pomegranate rind	Nil	5.6507	59.21	8.21	38.69	Light khaki
	15% T	7.7744	54.31	10.8	37.03	Deeper khaki
	15% T + 0.5% Cu	8.9233	46.49	8.46	28.13	Dark greenish khaki
	15% T + 1% Cu	9.6912	43.36	8.92	24.93	Dark greenish khaki

T – tamarind seed coat mordant; Cu – copper sulphate; L^*: lightness (0 = black, 100 = white), L = lightness; $a^*\pm$ = red/green; $b^*\pm$ = yellow/blue.

Table 4 *K/S* values, colour coordinates and colour of natural dyes dyed silk fabric with and without mordanting.

Natural dye (shade, 20%)	Mordants (%)	*K/S* value	Colour coordinates			Colour obtained
			L^*	a^*	b^*	
Turmeric	Nil	2.6676	79.99	−0.72	50	Bright yellow
	15% T	3.8933	67.77	4.47	39.16	Golden yellow
	15% T + 0.5% Cu	7.9712	59.78	4.49	44.68	Dark golden yellow
	15% T + 1% Cu	8.1717	57.88	4.44	40.57	Dark golden yellow
Pomegranate rind	Nil	2.1673	67.72	5.97	30.26	Light khaki
	15% T	4.0377	61.07	8.47	30.46	Dark khaki
	15% T + 0.5% Cu	4.2662	54.41	8.18	24.86	Reddish khaki
	15% T + 1% Cu	5.1861	49.89	9.43	22.62	Dark reddish khaki

T – tamarind seed coat mordant; Cu – copper sulphate; L^*: lightness (0 = black, 100 = white), L = lightness; $a^*\pm$ = red/green; $b^*\pm$ = yellow/blue.

namely *S. aureus* and *E. coli*. The minimum inhibition concentration of the tamarind seed coat mordant at which an inhibition zone width >2 mm observed was taken as the MIC of the mordant against both the bacteria. The results for the clear zone of inhibitions (mm) are shown in Fig. 1 for *S. aureus* and *E. coli*, respectively. The result shows that as the concentration of tamarind seed coat mordant increased from 0.5% to 15%, the width of the clear zone of inhibition (mm) also increased against *S. aureus* and *E. coli* bacteria. At 1% mordant concentration, the width zones of inhibition (mm) were 5.5 and 4 mm for *S. aureus* and *E. coli*, respectively. Even at 0.5%, the tamarind seed coat mordant showed antibacterial activity against *S. aureus* and *E. coli* but the width of the clear zone of inhibition (mm)for both bacteria was only 1.5 and 1 mm, respectively and was lesser than 2 mm which is the minimum requirement for a good antimicrobial agent. Thus, 1% mordant concentration was considered as the MIC against both the *S. aureus* and *E. coli* bacteria. It also was found that the clear zone of inhibition (mm) was higher for *S. aureus* as compared to *E. coli*. This is due to the Gram-positive bacteria being more sensitive to the bactericidal effect of tannins than the Gram-negative bacteria.

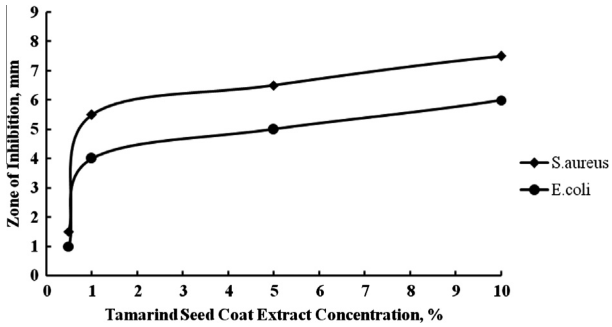

Figure 1 Effect of tamarind seed coat extract concentration on antibacterial activity against *S. aureus* and *E. coli*.

3.5. Antimicrobial activities of mordanted and dyed fabrics

In order to find the effectiveness of the antimicrobial activity of the mordanted and natural dyed fabrics evaluated quantitatively using AATCC Test 100-2004, the results for percent reduction of bacteria for cotton, wool and silk fabrics dyed with natural dyes with and without mordants are given in Table 5.

From Table 5, the results clearly indicate that the cotton pre-mordanted with tamarind seed coat mordant and dyed with turmeric showed higher antibacterial activity as compared to only turmeric dyed cotton fabric against *S. aureus* and *E. coli*. The activity increased from 89.58% to 94.44% for *S. aureus* and for *E. coli* (87.50% to 93.75%). It is clear from the results that the mordanting with tamarind seed coat mordant showed a definite increase in the reduction of the growth of both types of bacteria. A similar trend was observed in the case of pomegranate rind dyed cotton fabrics. Also, in the case of wool and silk fabrics with respect to antibacterial activity against *S. aureus* and *E. coli*, similar results were obtained as in the case of cotton and their results are given in Table 5. For cotton fabrics per-mordanted with tamarind seed coat mordant in combination with 0.5% and 1% copper sulphate mordant and dyed with turmeric, the reduction percentage was 100% against *S. aureus* and *E. coli*. Hence, it can be inferred that 15% tamarind seed coat mordant in combination with 0.5% or 1% copper sulphate mordant concentration can act as a good antibacterial agent against *S. aureus* and *E. coli* bacteria. A similar trend was obtained in the case of pre-mordanted cotton fabrics and dyed with pomegranate rind. In the case of wool and silk fabrics with respect to antibacterial activity against *S. aureus* and *E. coli*, similar results were obtained as in the case of cotton samples and their results are given in Table 5. In general, the antibacterial activities of mordanted and dyed cotton, wool and silk fabrics were higher than that of only naturally dyed fabrics.

3.6. Wash durability of antibacterial activity mordanted and dyed fabrics

An important issue to be considered in any functional treatment is the durability to washing of the functional treatment. For disposable material, it only requires temporary functional property. For all other applications, the functional property should be reasonably resistant to washing. To test the durability to home laundering, the antibacterial activities of pre-mordanted and dyed cotton, wool and silk fabrics were tested after 5, 10, 15 and 20 washes against *S. aureus* and *E. coli*. The findings are given in Tables 6–8.

From Tables 6, it can be observed that the antibacterial activity of treated (0 wash) cotton fabrics pre-mordanted with tamarind seed coat mordant and dyed with turmeric shows a 94.44% and 93.75% reduction in growth against *S. aureus* and *E. coli* bacteria, respectively, but that reduces to 79.16% and 75% after five washes and further decreased drastically to 58.33% and 51.38% after 10 washes, respectively. A similar trend was observed in the case of pomegranate rind dyed cotton fabrics. In general, tamarind seed coat mordant, pre-mordant, and turmeric, pomegranate dyed cotton fabrics are found to have good resistance to bacterial attack. At the same time, these fabrics are found to have good antibacterial activity up to five washes. However, the cotton samples pre-mordanted with tamarind seed coat mordant in combination with 0.5% or 1% copper sulphate and dyed with natural dyes retain almost around 70% antibacterial activity even after 20 washes. This is perhaps due to the presence of copper ions which form insoluble copper tannates with tamarind seed coat tannin phenolic hydroxyl groups and further form insoluble lakes with the dye molecules and retain the antibacterial activity after repeated washings. Similar results were obtained in the case of naturally dyed wool and silk fabrics against *S. aureus* and *E. coli* and their results are shown in Tables 7 and 8, respectively. In general, it was found that the tamarind seed coat mordant alone

Table 5 Antibacterial activity of natural dyes dyed cotton, wool and silk fabrics with and without mordants.

Natural dyes (shade, 20%)	Mordants (%)	Reduction (%)					
		Cotton		Wool		Silk	
		S. aureus	*E. coli*	*S. aureus*	*E. coli*	*S. aureus*	*E. coli*
Turmeric	Nil	89.58	87.50	90.90	88.36	92.46	88.19
	15% T	94.44	93.75	96.36	95.45	96.23	95.78
	15% T + 0.5% Cu	99.30	100	100	100	100	100
	15% T + 1% Cu	100	100	100	100	100	100
Pomegranate rind	Nil	90.27	88.88	93.22	92.72	91.52	90.96
	15% T	93.75	91.66	96.72	96.36	95.78	95.48
	15% T + 0.5% Cu	100	100	100	100	100	100
	15% T + 1% Cu	100	100	100	100	100	100

T – tamarind seed coat mordant; Cu – copper sulphate.

Table 6 Wash durability of antibacterial activity of natural dyes dyed cotton fabrics with mordants against *S. aureus* and *E. coli*.

Natural dye (shade, 20%)	Mordants (%)	Reduction (%) (0 wash)	Reduction (%) (5 washes)	Reduction (%) (10 washes)	Reduction (%) (15 washes)	Reduction (%) (20 washes)
Turmeric	15% T	94.44 (93.75)[a]	79.16 (75.00)[a]	58.33 (51.38)[a]	–	–
	15%T + 0.5% Cu	99.3 (100)[a]	88.88 (86.80)[a]	83.33 (81.25)[a]	79.16 (76.38)[a]	72.22 (70.83)[a]
	15%T + 1% Cu	100 (100)[a]	90.27 (88.19)[a]	84.72 (82.63)[a]	79.86 (77.77)[a]	74.3 (72.91)[a]
	15% T	93.75 (91.66)[a]	75 (72.91)[a]	55.55 (54.16)[a]	–	–
Pomegranate rind	15% T + 0.5% Cu	100 (100)[a]	89.58 (87.50)[a]	82.63 (81.25)[a]	78.47 (77.08)[a]	70.83 (69.44)[a]
	15% T + 1% Cu	100 (100)[a]	91.66 (90.97)[a]	83.05 (81.94)[a]	79.44 (79.16)[a]	71.52 (72.22)[a]

T – tamarind seed coat mordant; Cu – copper sulphate.
[a] Corresponds to antibacterial activity against *E. coli*.

Table 7 Wash durability of antibacterial activity of natural dyes dyed wool fabrics with mordants against *S. aureus* and *E. coli*.

Natural dye (shade, 20%)	Mordants (%)	Reduction (%) (0 wash)	Reduction (%) (5 washes)	Reduction (%) (10 washes)	Reduction (%) (15 washes)	Reduction (%) (20 washes)
Turmeric	15% T	96.36 (95.45)[a]	78.18 (72.72)[a]	60 (58.18)[a]	–	–
	15% T + 0.5% Cu	100 (100)[a]	95.45 (94.54)[a]	86.18 (85.45)[a]	80 (77.27)[a]	77.27 (74.54)[a]
	15% T + 1% Cu	100 (100)[a]	96.72 (96.36)[a]	87.27 (86.45)[a]	81.63 (78.18)[a]	74.3 (76.36)[a]
Pomegranate rind	15% T	96.72 (96.36)[a]	75.45 (70.00)[a]	58.18 (54.45)[a]	–	–
	15% T + 0.5% Cu	100 (100)[a]	96.36 (95.45)[a]	85.45 (83.63)[a]	78.18 (76.36)[a]	72.72 (70.00)[a]
	15% T + 1% Cu	100 (100)[a]	97.09 (96.36)[a]	86.36 (84.54)[a]	78.72 (77.27)[a]	73.63 (72.72)[a]

T – tamarind seed coat mordant; Cu – copper sulphate.
[a] Corresponds to antibacterial activity against *E. coli*.

Table 8 Wash durability of antibacterial activity of natural dyes dyed silk fabrics with mordants against *S. aureus* and *E. coli*.

Natural dye (shade, 20%)	Mordants (%)	Reduction (%) (0 wash)	Reduction (%) (5 washes)	Reduction (%) (10 washes)	Reduction (%) (15 washes)	Reduction (%) (20 washes)
Turmeric	15% T	96.23 (95.78)*	77.71 (77.56)*	63.85 (60.84)*	–	–
	15% T + 0.5% Cu	100 (100)*	93.37 (93.37)*	83.43 (82.83)*	77.71 (77.40)*	72.59 (72.13)*
	15% T + 1% Cu	100 (100)*	94.42 (94.12)*	84.18 (83.88)*	78.91 (78.31)*	73.19 (72.89)*
	15% T	95.78 (95.48)*	76.5 (75.90)*	62.34 (59.34)*	–	–
Pomegranate rind	15% T + 0.5% Cu	100 (100)*	92.62 (92.16)*	81.47 (81.17)*	77.4 (76.95)*	69.87 (69.27)*
	15% T + 1% Cu	100 (100)*	93.07 (92.46)*	82.22 (81.62)*	78.61 (78.16)*	70.33 (69.87)*

T – tamarind seed coat mordant; Cu – copper sulphate.
* Corresponds to antibacterial activity against *E. coli*.

Table 9 Light and wash fastness ratings of natural dyes dyed cotton, wool and silk fabrics with and without mordants.

Natural dye (shade, 20%)	Mordants	Light fastness			Wash fastness					
					Change in colour			Staining cotton/wool	Staining wool/cotton	Staining silk/cotton
		Cotton	Wool	Silk	Cotton	Wool	Silk			
Turmeric	Nil	2	2	2	2–3	2–3	2–3	4	4	4
	15% T	5	5	5	4	5	4–5	5	5	5
	15% T + 0.5% $CuSO_4$	6	6	6	5	5	5	5	5	5
	15% T + 1% $CuSO_4$	6	6	6	5	5	5	5	5	5
Pomegranate rind	Nil	3	3	3	2	2	2	4	4	4
	15% T	5	5	5	4–5	5	4–5	5	5	5
	15% T + 0.5% $CuSO_4$	6	6	6	5	5	5	5	5	5
	15% T + 1% $CuSO_4$	6	6	6	5	5	5	5	5	5

T – tamarind seed coat mordant; Cu – copper sulphate, Light fastness rating:1 – poor,2 – fair,3 – moderate,4 – good,5 – better,6 – very good,7 – best and 8 – excellent, Wash fastness rating: 1 – poor, 2 – fair, 3 – good, 4 – very good and 5 –excellent.

can be used to impart a temporary finish to cotton, wool and silk fabrics dyed with both natural dyes. This type of finish may be required for materials which do not need laundering as in the case of hygiene markets such as bandage gauze, sanitary napkins and other wound healing products. However, for a durable antibacterial finish to washing for cotton, wool and silk fabrics with natural dyes, the tamarind seed coat mordant has to be used in combination with copper sulphate mordant. Out of the two concentrations of copper sulphate, even 0.5% (owf) copper sulphate mordant concentration can be sufficiently enough to obtain good antibacterial activity against *S. aureus* and *E. coli* bacteria.

3.7. Fastness properties

The wash and light fastness properties of cotton, wool and silk fabrics mordanted and dyed with natural dyes are given in Table 9. The wash fastness properties with respect to change in colour were of the order of 4–5 (midway between very good to excellent). The staining of all three naturally dyed samples revealed good fastness with grade 5. The light fastness was in the range of 5, i.e., better for cotton, wool and silk per-mordanted with tamarind seed coat mordant and naturally dyed fabrics. However, the light fastness improved one rating and was in the range of 6, i.e., very good for all three substrates per-mordanted with tamarind seed coat mordant in combination with 0.5% and 1% (owf) copper sulphate mordant concentration and dyed with both natural dyes, which is quite acceptable.

4. Conclusions

Tamarind seed coat extract can be successfully employed as a natural mordant for dyeing cotton, wool and silk fabrics with natural dyes. The tamarind seed coat mordanted alone, and in combination with copper sulphate mordant and natural dyes dyed cotton, wool and silk fabrics showed higher K/S values as compared to only natural dyes dyed fabrics. It is possible to produce fabrics with good antibacterial activity using the tamarind seed coat mordant alone against Gram-positive *S. aureus* and Gram-negative *E. coli* bacteria. The only limitation is that such treated fabric does not have good wash durability after five washes. With the help of copper sulphate mordanting, the antibacterial activity of cotton, wool and silk fabrics with two natural dyes was found to have been enhanced significantly against *S. aureus* and *E. coli*. These fabrics are found to have a wash durability of up to 20 washes. Even using 0.5% (owf) copper sulphate mordant treatment after 15% (owf) tamarind seed coat mordant and dyed with natural dyes for all three substrates is adequately enough to obtain a good antibacterial activity against *S. aureus* and *E. coli* bacteria. The wash and light fastness properties of the dyeing on fabrics were found to be enhanced when mordanting was done. This process is promising and should be easily industrialized, popularized among the local natural dyers in order to obtain an eco-friendly product with value added properties.

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