Agr. Nat. Resour. 53 (2019) 188–196

AGRICULTURE AND NATURAL RESOURCES

Journal homepage: http://anres.kasetsart.org

Research article

Antibacterial activity assessment of woolen fabric treated with natural dyes and chitosan

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Article Info

Article history:
Received 3 August 2017
Revised 26 October 2018
Accepted 5 November 2018
Available online 30 April 2019

Keywords:
Antimicrobial activity,
Chitosan,
Mordant,
Natural dye,
Wool fabric.

Abstract

In the textile industry, the finishing process has an important role in the antibacterial characterization of fabrics. Antibacterial growth was investigated in woolen fabric with natural biopolymeric material (chitosan) and natural dyeing (turmeric and madder). The effect of mordant concentration was studied in terms of dyeing concentration, dyeing temperature and dyeing time on the color strength. The results indicated that colorant absorption coefficient/colored substrate scattering coefficient (K/S) values increased with an increase in the chitosan and dyeing concentrations. The highest K/S value for turmeric was 2% chitosan, 55ºC dyeing temperature and 105 min and for madder was 2% chitosan, 73°C temperature and 75 min at 9% shade. Good fastness properties were also achieved using both dyes. The results confirmed that the combination of chitosan and natural dye enhanced the antibacterial activity of woolen fiber against Escherichia coli and Staphylococcus aureus. The impregnation of chitosan in the fabric structure was also examined using scanning electron micrographs.

Introduction

Natural dyes are easily available and provide a sustainable alternative option as a coloring material and they have been used, since the nineteenth century to dye cotton, wool and silk (Shahid and Mohammad, 2013).The demand for natural dyes has reduced due to synthetic dyes becoming more popular in small and large-scale industry, though in developing countries still, some synthetic dyes like azo are used for textiles, leather, toys and plastics product (Ahlström et al., 2005) and may have harmful effects (Sen et al., 2016). Synthetic dyes have their own drawbacks in terms of health, safety and being environment-friendly and are banned in many countries (Singh et al., 2002, Crini et al., 2008). As an alternative, natural dyes have numerous advantages such as resistance to moth invasion, anti-allergic, safer for body contact, non hazardous, economical and recyclable and therefore should be considered as a sustainable colorant for the textile industry (Siva, 2007; Erdawati et al., 2013; Swamy et al., 2014). Natural dyes can be classified according to the nature of raw material such as plants, insects and minerals, and among these, dyes derived from plants have substantial antibacterial quality (Mirjalili et al., 2013). T*urmeric* (*Curcuma longa*) is one such dye; it has been extracted from medicinal plants and used for coloring purposes (Mirjalili et al., 2014). The pigment extract from *C. longa* is known to be a curcuminoid, with small amounts of demethoxy curcumin and bisdemethoxy curcumin,

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https://doi.org/10.34044/j.anres.2019.53.2.14

as shown in Fig. 1 (Bechtold et al., 2009). Turmeric has unique biochemical properties, so that it has been used in wound-healing and for its anti-inflammatory, anticancer, anti-proliferative, antifungal, and antibacterial activity (da Silva et al., 2018). Mirjalili et al. (2013) reported that turmeric shows good antibacterial, wash fastness and dye uptake properties with cotton and silk fabric. In addition, wool fabrics also showed excellent fastness, durability and antibacterial activity properties, when treated with other natural dyes (green tea, turmeric, saffron petals) and aluminum sulfate as a mordant using five washing cycles and 5 hr light exposure (Ghaheh et al., 2014). Madder is another natural dye derived from the roots of *Rubia tinctoria*. The main coloring constituents in madder area mixture of purpurin and munjistin, as shown in Fig. 2 (Saxena et al., 2014). In the middle ages, madder red was expensive in Europe and was typically used for special occasions (Chenciner, 2003). Madder can produce a range of reddish colors from pink to deep scarlet and is applied in the form of chips or powder (Schmidt-Przewoźna, and Brandys, 2016). In an acidic medium, it produces vivid oranges and rich browns on wool fabrics, medium shades of salmon and rose on silk fabrics and paler shades of oranges and salmons on cotton fabrics (Bechtold et al., 2009).

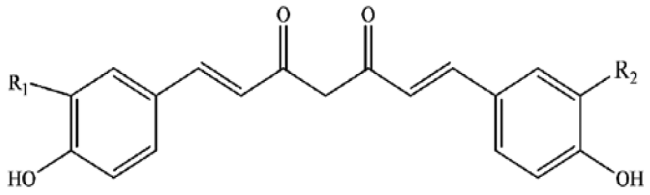

Fig. 1 Structures of curcumin (R1= OCH3 and R2 = OCH3 is curcumin; R1 = R2 = H is bisdemethoxycurcumin; R1 = OCH3 and R2 = H is demethoxycurcumin)

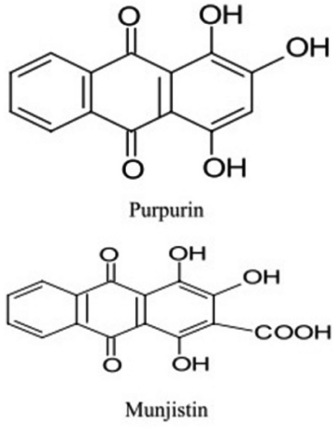

Fig. 2 Structures of purpurin and munjistin

Most of the natural dyes have antibacterial properties but they need some substitute along with mordants (Teli et al., 2013). Application of metallic mordants in the textile dyeing process could cause environmental hazards and be harmful. Hence another choice is a natural mordant like chitosan (Teli et al., 2014). Chitosan is biopolymeric material containing antibacterial properties (Bano et al., 2017). It is a good supplementary agent in textile printing that can be extracted from the shrimp shell waste (Suryawanshi et al., 2019). Treatment with chitosan enhanced the color strength of dyed fabrics as well acting as a good shrink-resistance agent for wool (Davidson et al., 1994; Dev et al., 2009). Nowadays, the textile industry is looking for sustainable production and consumption to address global environmental and economic issues; therefore, natural products are an alternative option in various applications in the textile industry.

The aims of this research work were: 1) to investigate the quality of natural dye madder and turmeric dyes with natural mordant chitosan for woolen fabrics; 2) to examine the effects of chitosan concentration, dye shade percentage, dyeing temperature and dyeing time on the color strength of dyed fabrics; and 3) to assess the antibacterial activity of woolen fabrics treated with mordant and both dyes.

Materials and Methods

Materials

Fabric: The mill-scoured pure (100%) wool fabric was supplied by Madan Textiles Industry Punjab, India. The specifications of the wool fabric are given in Table 1.

Chitosan: The chitosan derivative chitosan chloride was supplied by India Sea Food (regd.) Kochi, Kerala, India. The specifications of the chitosan are given in Table 2.

Chemicals: The acetic acid, hydrogen peroxide, sodium pyrophosphate and sodium hydroxide used in the experiments were all laboratory grade.

Natural Dyes: Raw turmeric root and madder dye powder were purchased from a local market in Bhiwani Haryana, India. Turmeric powder was extracted from the root, with the latter boiled for 1 h followed by filtration and the residual impurities discarded. The madder dye powder was used in the original form as received (Ali et al., 2011).

Table 1 Fabric specifications

Variable	Specification
Fabric	100% wool
EPI/PPI	34/46
Weave	Twill(2/1)weave
GSM	136

EPI/PPI = End Per Inch/Pick Per Inch; GSM = Gram Per Squar.

Table 2 Specifications of chitosan chloride

Variable	Detail
Chemical name	Chitosan chloride
Source	Chemically modified from chitin flakes
Appearance and color	Powder & off white
Solubility	Water soluble
Density	>0.6gm/ml

Pre-treatment

Oxidation of wool with hydrogen peroxide

Oxidation of wool with hydrogen peroxide was carried out using the exhaustion method at pH 9. The fabric sample (1 g) and solvent (30 mL) were used at a liquor ratio 1:30. The treatment of fabric was completed with 18 mL/L hydrogen peroxide and 2 g/L of sodium pyrophosphate at 70°C for 1 hr. After that, the sample was washed carefully and dried under ambient conditions (Julia et al., 1998).

Pre-mordanting using chitosan

After oxidation, the wool samples were immersed in the chitosan solution at a liquor ratio of 1:20. The chitosan solution was prepared by dissolving different concentrations of chitosan (1–2%) in deionized water. The exhaustion procedure was carried out for 1 hr at 60°C. The samples were then rinsed properly and dried properly (Chen et al., 2007).

Dyeing procedure

The treated wool samples were dyed in a dye bath, containing a measured amount of dye and acetic acid (4g/L) with a liquid ratio of 1:30. After that, the sample was heated at different temperatures for different periods, as shown in Fig. 3. The samples were rinsed with cold water and subsequently with a nonionic detergent (3g/L) using a liquor ratio of 1:30 at 45°C for 40 min. Finally, the samples were rinsed with distilled water and dried at ambient temperature.

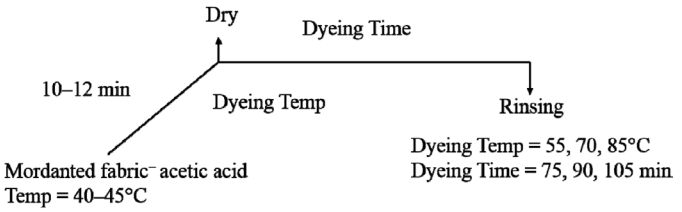

Fig. 3 Dyeing process of mordanted wool fabric at dyeing temperature 55, 70, 85°C and time 75, 90, 105 min

Optimization of dyeing conditions

To optimize the suitable conditions, dyeing was carried out at different concentrations of chitosan, different temperatures and different times, as detailed in Table 3. The experiments were optimizing using the Design Expert software version 7.1 (Stat-Ease, Inc.; Minneapolis, MN, USA) and different runs were formulated as shown in Table 4. Three different percentages (3%, 6% and 9%) of both dyes were used to obtain light, medium and dark shades, respectively (Prabu et al., 2002).

Table 3 Levels of variables chosen for design of experiment

Chitosan concentration (% w/w)	Dyeing temperature (°C)	Dyeing time (min)
1 (-1)	55 (-1)	75 (-1)
1.5 (0)	70 (0)	90 (0)
2 (+1)	90 (+1)	105 (+1)

Table 4 Box-Behnken design for three independent variables

Experimental Run	Chitosan (% weight per weight)	Dyeing time (min)	Dyeing temperature (°C)
1	1.5	75	85
2	1.5	90	70
3	2.0	105	70
4	1.0	105	70
5	1.5	90	70
6	1.0	90	85
7	1.0	90	55
8	2.0	90	85
9	1.5	75	55
10	1.5	90	70
11	1.5	105	85
12	1.5	105	55
13	1.0	75	70
14	1.5	90	70
15	2.0	75	70
16	1.5	90	70
17	2.0	90	55

Testing and analysis methods

Testing method

Color strength

The color yield, expressed as a K/S value using Equation 1, with a wavelength range of 400 nm to 700 nm with 10 nm intervals measured usinga spectrophotometer (Gretag Macbeth color eye 7000A; X-Rite; Grand Rapids, MI, USA) and calculated in accordance with the Kubelka-Munk equation (Teli et al., 2014), as shown in Equation 1:

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

where K is the absorption coefficient, depending on the concentration of colorant; S is the scattering coefficient, caused by the colored substrate; and R is the reflectance of the colored substrate.

Analysis method

Fastness testing

Fastness testing of dyed fabric samples was determined according to IS:764–1984 methods using a Sasmira Launder-O-Meter (Mumbai, India) followed with the IS-3 wash fastness method. The wash fastness rating was examined using a grey scale as per ISO-05-A02 (loss of shade depth) and ISO-105-AO3 (extent of staining). Color

fastness to rubbing (dry and wet) was assessed according to the IS: 766-1984 method using a manual-operated crock meter and grey scale as per ISO-105-AO3 (Kumaresan et al., 2013).

Antimicrobial activity

The treated samples were investigated for antimicrobial properties using the AATTC Test Method (AATCC 100–2004). The *E. coli* method was used to test bacterial activity with a Gram-negative bacterium S. *aureus* and a pathogenic Gram-positive bacterium (Khan et al., 2011). A circular (4 cm diameter) fabric swatch was cut and the untreated wool fabric was used as a control sample. The dilution medium was nutrient broth and the neutralizer was sodium hydroxide. Antimicrobial activity on the wool fabric was examined using a comparative study for the reduction in colony number between treated and untreated fabrics after incubation. The results were expressed as the percentage reduction of bacteria (*R*) using Equation 2:

$$R\ (\%) = (A\text{-}B) / A * 100\% \quad (2)$$

where *A* and *B* are the numbers of bacteria recovered from the untreated and dyed treated wool fabric swatches, respectively.

Result and Discussion

K/S (Color strength)

The K/S values obtained for turmeric and madder dye with respect to all shades are shown in Tables 5 and 6, respectively. The results were analyzed statistically to determine the optimum parameters and desirability function of the experimental design. The analysis of variance model had coefficient of determination (R^2) values in the range 0.8–0.9 for all shades. Increased chitosan concentration after oxidation increased the K/S value. This might have been due to the simple formation of the amino groups of chitosan and the hydroxyl groups of the wool fabrics after oxidation (Ali et al., 2011). Similarly, the K/S value increased with increased dyeing time and concentration. The color strength was higher at 105 min treatment compared to 75 min; there was different percentage shade increases with both dyes at the different 3%, 6% and 9% dye concentrations.These were attributed to an increase in the driving force for dye transport toward the fiber surface, due to the increased higher concentration gradient of dye occurring in two phases (Zhang et al., 2017). The dyeing temperature did not have a prominent effect on the K/S values. The interaction results of different parameters for turmeric and madder are presented in Figs. 4A–4D and Figs. 5A–5D, respectively.

Table 5 Colorant absorption coefficient/colored substrate scattering coefficient (K/S) values for turmeric-dyed wool samples

Experiment number	K/S_{T3}	K/S_{T6}	K/S_{T9}
1	2.091	3.603	3.566
2	2.809	4.311	4.282
3	4.760	6.370	6.442
4	3.450	5.041	5.002
5	2.311	3.912	3.884
6	2.259	3.761	3.726
7	2.682	4.191	4.146
8	4.000	5.512	5.472
9	2.548	4.101	4.061
10	2.331	3.836	3.827
11	3.087	4.589	4.545
12	3.670	5.220	5.240
13	2.819	4.402	4.367
14	2.387	3.889	3.841
15	4.010	5.570	5.462
16	2.205	3.711	3.767
17	4.110	5.710	5.770

K/S_{T3}, K/S_{T6} and K/S_{T9} representing K/S values for 3%, 6% and 9% shade of turmeric dyed wool, respectively. Values shown as mean of three values.

Table 6 Colorant absorption coefficient/colored substrate scattering coefficient (K/S) values for madder dyed wool samples

Experiment number	K/S_{M3}	K/S_{M6}	K/S_{M9}
1	1.252	4.235	4.797
2	1.245	4.136	4.876
3	4.01	6.824	7.198
4	3.12	6.12	6.465
5	1.897	4.778	5.107
6	1.123	3.947	4.298
7	1.356	4.236	4.798
8	3.021	5.836	6.187
9	1.639	4.502	5.007
10	1.125	3.947	4.298
11	1.352	4.514	4.905
12	1.542	4.601	5.007
13	1.602	4.592	4.954
14	1.528	4.403	4.787
15	4.21	7.11	7.55
16	1.132	4.013	4.573
17	3.221	6.223	6.651

K/S_{T3}, K/S_{T6} and K/S_{T9} representing K/S values for 3%, 6% and 9% shade of turmeric dyed wool, respectively. Values shown as mean of three values.

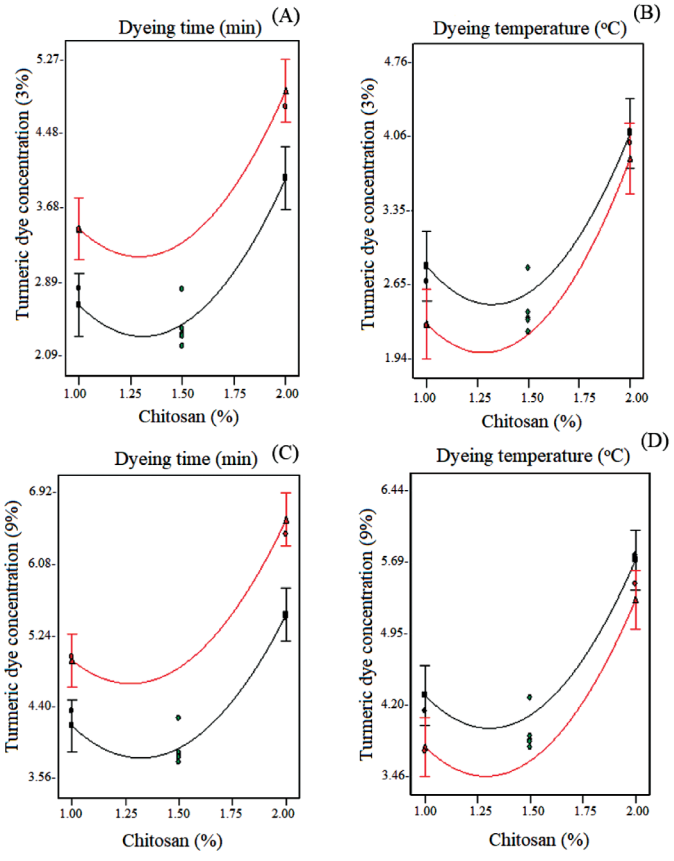

Fig. 4 Interaction graphs of various parameters for turmeric: (A) chitosan percentage and time (T3%); (B) chitosan percentage and temperature (T3%); (C) chitosan percentage and time (T9%); (D) chitosan percentage and temperature (T9%).

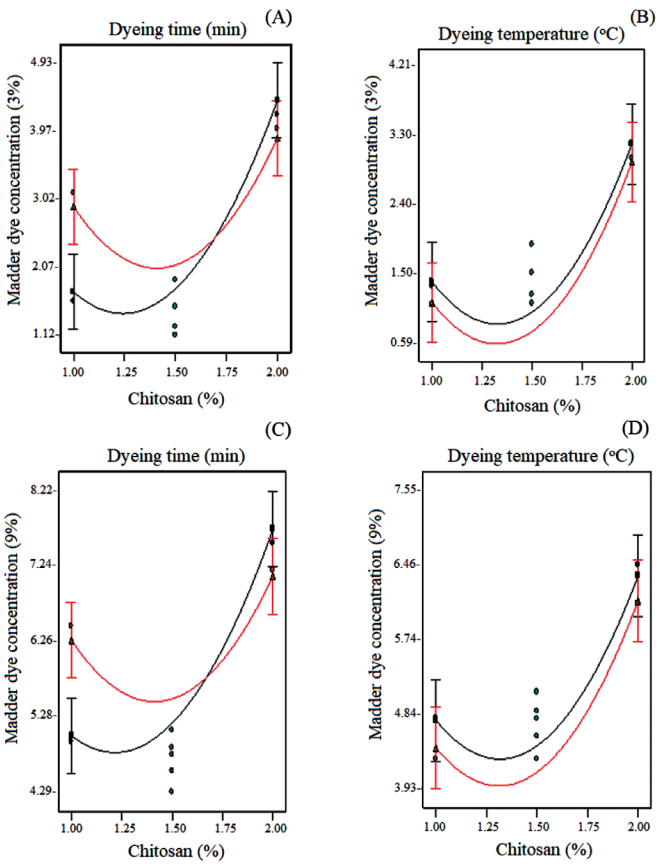

Fig. 5 Interaction graphs of various parameters for madder: (A) chitosan percentage and time (M3%); (B) chitosan percentage and temperature (M3%); (C) chitosan percentage and time (M9%); (D) chitosan percentage and temperature (M9%).

The sorption of chitosan on raw wool fabric was due to ionic interaction between the negative charges of carboxylic groups ($–COO^-$) in the polypeptide macromolecule and the protonated amino groups (NH_3^+) of chitosan. The deacetylated derivative of chitin (protonated form (NH_3^+) in aqueous solution at pH <6) is shown in Fig. 6. In the case of bleached wool, an additional vacancy at cysteic groups (SO_3^-) possibly resulted in higher cross-linking of chitosan, and the proposed mechanism is shown in Fig.7.The turmeric dye has ionizable –OH groups as auxochromes, which attached to the sites available in bleached chitosan-treated wool fiber as shown in Fig. 8.In aqueous medium at optimum pH, these ionizable groups of turmeric were dissolved due to conversion in anionic forms (Rafols et al., 2017). In addition, chitosan formed a strong bond with the sulphonic acid group on the surface of bleached wool which may result in further attraction and react with the turmeric dyes, leading to an increased dyeing rate and a reduction in dyeing time. However, the optimum time obtained was less for madder as it has a greater number of ionizable OH groups compared to turmeric.

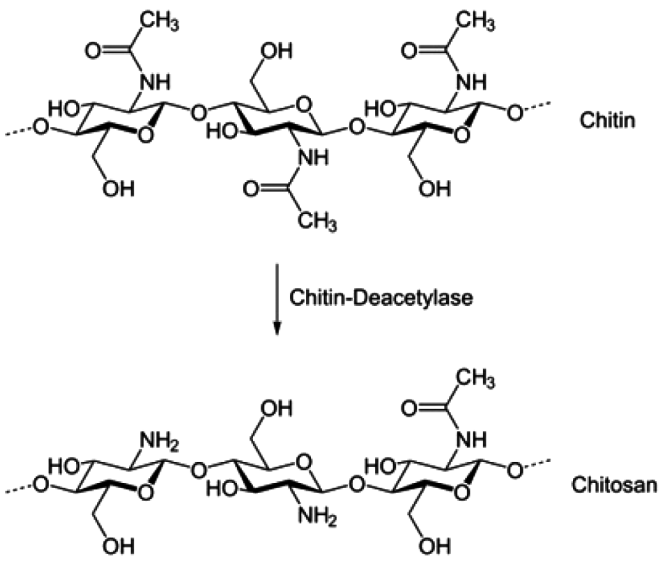

Fig. 6 Deacetylation of chitin to form chitosan

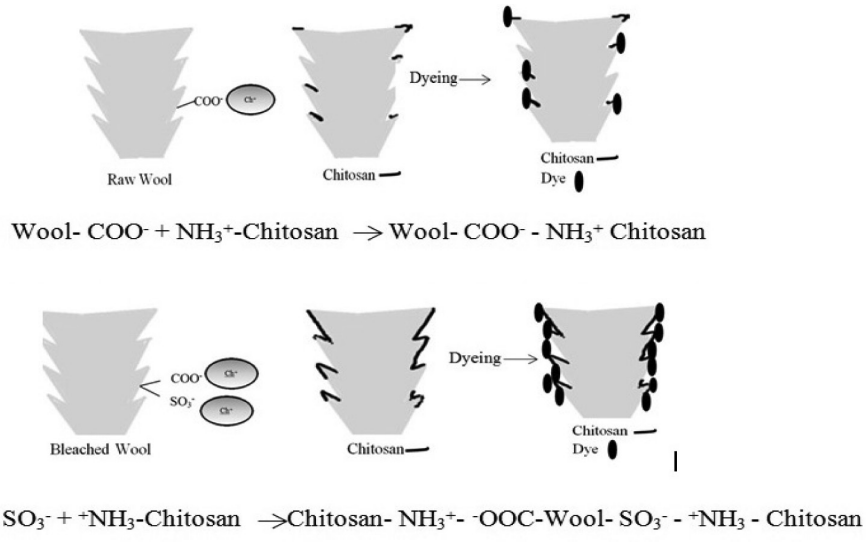

Fig. 7 Proposed mechanism of wool and chitosan attachment

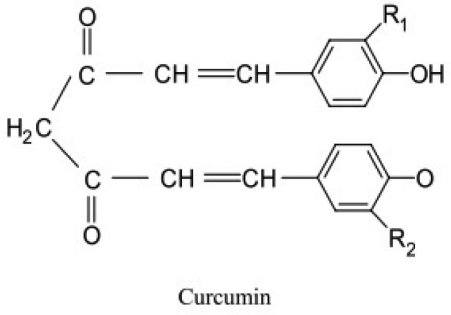

Fig. 8 Ionizable –OH groups of turmeric

The micrograph structures of wool at different chitosan percentages are presented in Fig. 9A and 9B. It was observed that the chitosan percentage on the raw wool increased with the increased concentration of chitosan from 1% to 2%.The values of optimized parameters satisfying the desirability criteria for 'maximizing the K/S value' for turmeric and madder as obtained using the software are given in Tables 7 and 8. Table 7 shows no significant differences (almost similar values of K/S) between the optimized values for 6% and 9% shades for turmeric dyes. Similarly, for madder (Table 8), there were similar values for chitosan (approximately 2%), dyeing time (approximately 105min) and different temperature (approximately 67°C and 55°C) and no significant differences in the K/S values. The effect of an increase in temperature was only for limited time, as for further increase in temperature, there were no changes in the K/S values.

The structural properties of treated and untreated wool fabric with chitosan and dyes were examined using Fourier-transform infrared spectroscopy. The biochemical interaction between chitosan and wool fiber polymer was investigated using the changes in the peaks from the characteristic spectra (Fig. 10). In general, no major changes were observed in bands or their intensities for all test fabrics. The broad absorption band which appeared in the range of 3700– 2900 cm−1 indicates the presence of –OH groups in the cellulose polymer (Martínez-Sanz et al., 2011). The presence of $–NH_2$ group in the treated fabric was responsible for introducing a cationic site in the wool polymer resulting in improved dye exhaustion and also interaction with microorganisms for antibacterial properties. Again, the spectrum near 2500 cm^{-1} corresponding to the symmetric stretching of methylene ($–CH_2–$) groups (Babu et al., 2015) and 1500 cm^{-1} related to the C=O stretch of esters were found to be similar in all the tested samples. The absorption peak that appeared at 1300 cm^{-1} for chitosan-treated wool fabric suggested the formation of a Schiff base (C=N double bond) between the aldehydic carbonyl group of cellulose and the amino group of chitosan (Tian et al., 2017). All three samples showed peaks near 1320 and 1100 cm^{-1} being related to –OH bending of C–O–H alcohol groups (Rusu et al., 2016) and 960 cm^{-1} corresponding to C–O mainly to $C_3–O_3H$ secondary alcohol (Bakashi et al., 2018). Moreover, the peak at 700 cm^{-1} corresponding to asymmetric out-of-phase ring stretching of $C_1–O–C_4$ β–glucosidic bonds increased after the dyeing process (Araujo et al., 2018). The increase in the percentage absorption of dye in mordanted samples may have been due to the formation of a chemical bridge between the dye and fabric through natural mordant which was fixed on the fiber and enhanced the fixation of dye (Islam et al., 2016).

Color fastness

Color fastness indicates the resistance of a material to any change in any of its color characteristics (Ramalingam et al., 2016). The results of color fastness are shown in Tables 9 and 10 for turmeric and madder, respectively. It can be clearly seen from both tables that the fastness properties were excellent for both of dyes. This might have been due to electron-donating groups (OH groups) in the dye structure being capable of forming a complex, that doesn't participate during washing or rubbing (Das, 2011).

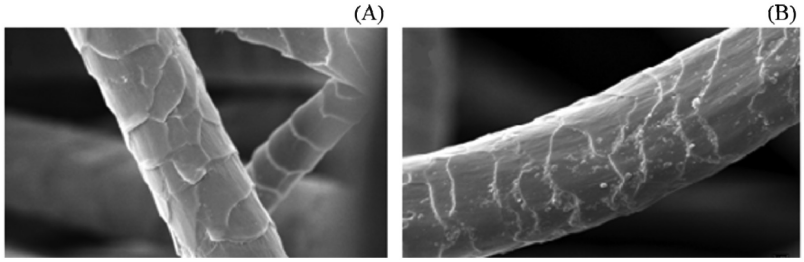

Fig. 9 Macrograph images showing chitosan percentage increase on raw wool with rising concentration: (A) 1% chitosan; (B) 2% chitosan

Table 7 Optimized parameters for turmeric dyed samples to achieve maximum colorant absorption coefficient/colored substrate scattering coefficient (K/S)value

Shade (%)	Chitosan (%w/w)	Dyeing time (min)	Dyeing temperature (°C)	K/S	Desirability
3%	1.97	105	55	4.9	1
6%	1.98	105	67	6.4	1
9%	1.96	104	56	6.5	1

w/w = weight per weight.

Table 8 Optimized parameters for madder dyed samples to achieve maximum colorant absorption coefficient/colored substrate scattering coefficient (K/S) value

Shade (%)	Chitosan (%w/w)	Dyeing time (min)	Dyeing temperature (°C)	K/S	Desirability
3%	1.99	75	75	4.2	1
6%	2	76	59	7.2	1
9%	2	75	73	7.6	1

w/w = weight per weight.

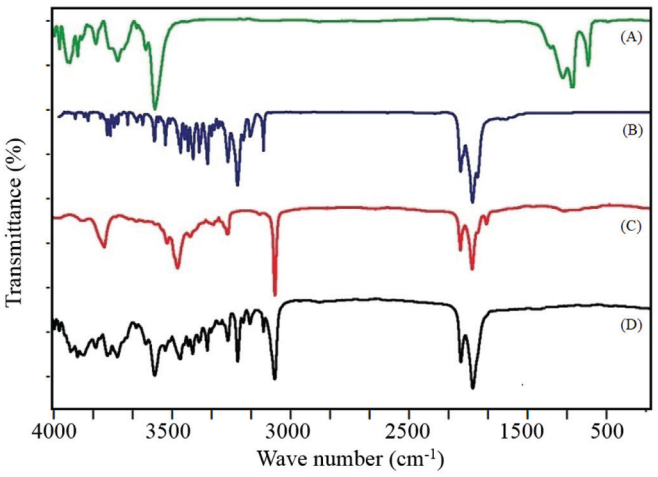

Fig. 10 Fourier-transform infrared spectroscopy images: (A) pure wool fabric; (B) treated with chitosan; (C) treated with chitosan and turmeric; (D) treated with chitosan and madder

Table 9 Color fastness for turmeric dye

Turmeric dye (%)	Wash fastness		Rubbing fastness	
	Color change	Color staining	Dry	Wet
3	4–5	4–5	4–5	3–4
6	4	4–5	4	3–4
9	4–5	4–5	5	4

Table 10 Color fastness for madder dye

Madder dye (%)	Wash fastness		Rubbing fastness	
	Color change	Color staining	Dry	Wet
3	4–5	5	5	4–5
6	5	5	5	4–5
9	4–5	4–5	5	4

Antibacterial assessment

The antibacterial activity of un-mordant-dyed and mordant-dyed fabrics was examined against *S. aureus*, *E. coli*, and *P. aeruginosa* and the results are shown in Table 11. With 2% chitosan as mordant, maximum color strength was achieved, as this was identified as the optimum mordant concentration. All experimental samples of turmeric and madder had high antibacterial activity against the tested microorganisms (bacteria). This could be attributed to the linkage of chromophores to the glycoside in natural dyes. It was also noted that the antibacterial activity of dyed samples showed significant increases after mordanting and dye concentration increases.

This research concluded that natural dyes and mordant are effective substitutes for synthetic dyes to reduce adverse environmental and economic conditions. The dyeing of wool fabric with natural dyes was successfully carried out using chitosan as an eco-friendly mordant. The K/S values varied with the dye and mordant percentage combination; hence the parameters were optimized to produce a maximum K/S

value for both dyes. The results showed a maximum K/S value at turmeric was 2% chitosan, 55ºC dyeing temperature and 105 min and for madder was 2% chitosan, 73°C temperature and 75 min at 9% shade. Moreover, the fixation of chitosan as a mordant on wool fabric was confirmed using scanning electron micrographs. The color fastness property for both wool fabrics was excellent. The turmeric and madder were suitable as an antibacterial agent for *E. coli* and *S. aureus* microorganisms and this increased with increased dye and mordant concentrations. Further investigation should investigate different organisms with different mordants and different types of fabric.

Table 11 Antibacterial activity of dyed woolen fabrics

Fabric sample	Bacterial reduction (%)	
	S. aureus	E.coli
Turmeric		
Unmordanted + 3% turmeric	94.2	93.0
Mordanted + 3% turmeric	99.4	99.2
Mordanted + 6% turmeric	99.6	99.5
Mordanted + 9% turmeric	99.8	99.7
Madder		
Unmordanted + 3% madder	95.3	96.2
Mordanted + 3% madder	99.2	99.1
Mordanted + 6% madder	99.5	99.4
Mordanted + 9% madder	99.6	99.7

Conflict of Interest

The authors have no conflicts of interest to declare.

Acknowledgments

The authors acknowledge Fashion and Apparel Engineering, The Technological Institute of Textile and Sciences, Bhiwani Haryana, India for funding and facilities.

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