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Novel Green Coloration of Cotton Fabric. Part I: Bio-mordanting and Dyeing Characteristics of Cotton Fabrics with Madder, Alkanet, Rhubarb and Curcumin Natural Dyes

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IT was envisioned that rendering the surface charge of cotton fabric cationic at acidic pH would allow it dyeable with natural dyes. For this purpose and with a vision for introducing a completely green process, the fabric was pretreated with tannic acid followed by padding in gelatin aqueous solution and curing. The factors that may affect this pretreatment process were investigated, as reflected by the color strength obtained of the dyed fabrics using madder dye. Furthermore, the pretreated fabric (bio-mordanted) was characterized by FTIR to confirm the success of chemical modification, and a tentative mechanism for the dye fixation is believed to be physical bonds (hydrogen bond and van der Waals bond) and chemical bond (ionic bond). The selected conditions obtained were comparatively applied using four natural dyes, namely, madder (CI Natural Red 9), curcumin (CI Natural Yellow 3), rhubarb (CI Natural Yellow 23), and alkanet (CI Natural Red 20). The successful results presented in this work and the excellent fastness properties obtained suggest its potential as a viable and ecological method for successful bio-mordanting and dyeing of cotton fabrics with natural dyes.

Keywords : Ecological; Green dyeing; natural dye; cotton; Bio-mordanting; Gelatin-tannic acid.

Introduction

Natural dyes have been recognized since ancient Egyptians. They are obtained from natural resources and renewable, and therefore, their uses will continue to inspire scientists due to their ecological, antimicrobial, and UV-protection characteristics [1-6]. The revival use of natural dyes, as well as devising new methods of techniques in textile coloration, is of prime ecological interest [7-10]. However, the use of natural dyes in textile coloration is yet limited due to many reasons such as shades, cost, and fastness properties. The shade and fastness properties of dyed fabrics depend on the mordants used.

Although the use of mordants is necessary for better dyeability and shades, however, their use causes a serious problem for the environment as most of the mordanting process uses toxic metal ions in a large amount that ultimately most of them get discharged into the effluent. Mordants used so far are either metallic or bio-mordant or their combination. Metallic mordant includes copper, chromium, tin, iron, and aluminum [11]. Bio-mordants include Turkey red oil, vegetable oils, and tannin [11]. Tannin is also a coloring matter that can be used by itself for the coloration of textiles after being mixed with metal for mordanting [12]. The use of mordants metallic or bio-mordant or their combination, although enhanced the color characteristics, yet their uses have been with limited success. Recently, tannin

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as bio-mordant for the dyeing of wool with madder dye has been reported to improve the washing fastness [13]. Also, the combination of tannin and metallic mordant was persuaded for high-temperature dyeing of polyester fabrics, but the results obtained were not satisfied regarding the light fastness [11]. Natural dyeing of polydopamine coated-silk has been reported with better light fastness properties [14].

On the other hand, cotton fabric is a cellulosic material that acquires negative charge upon wetting with water due to the presence of hydroxyl groups [15]. This problem has been addressedmostly by chemical modification of cotton to render it cationic before being colored with anionic dyes [16-19]. This approach has also been persuaded for the coloration of cationized cotton with natural dyes [5, 17, 18]. Chitosan, bovine serum albumin, and poly(ethyleneimine) were investigated as coating layers for the cotton fabric to render it dyeable with lac natural dye [20-22]. It has been reported that the treatment of cotton with tannic enhanced its adsorption of aluminum ions that has a good affinity towards Lac dye and thus improved the dyeability of the fabric [23].

In the interest of dyeing cotton fabrics with natural dyes, it was hypothesized that pretreating cotton so that its negative surface change could be changed to positive charge at acidic pH would lead to the successful dyeing of cotton with natural dyes. Madder (CI Natural Red 9), rhubarb (CI Natural Yellow 23), alkanet (CI Natural Red 20) and curcumin (CI Natural Yellow 3) [24, 25] were selected in this study to investigate the effect of dye structure [Fig. 1] on the dyeability of the fabrics.

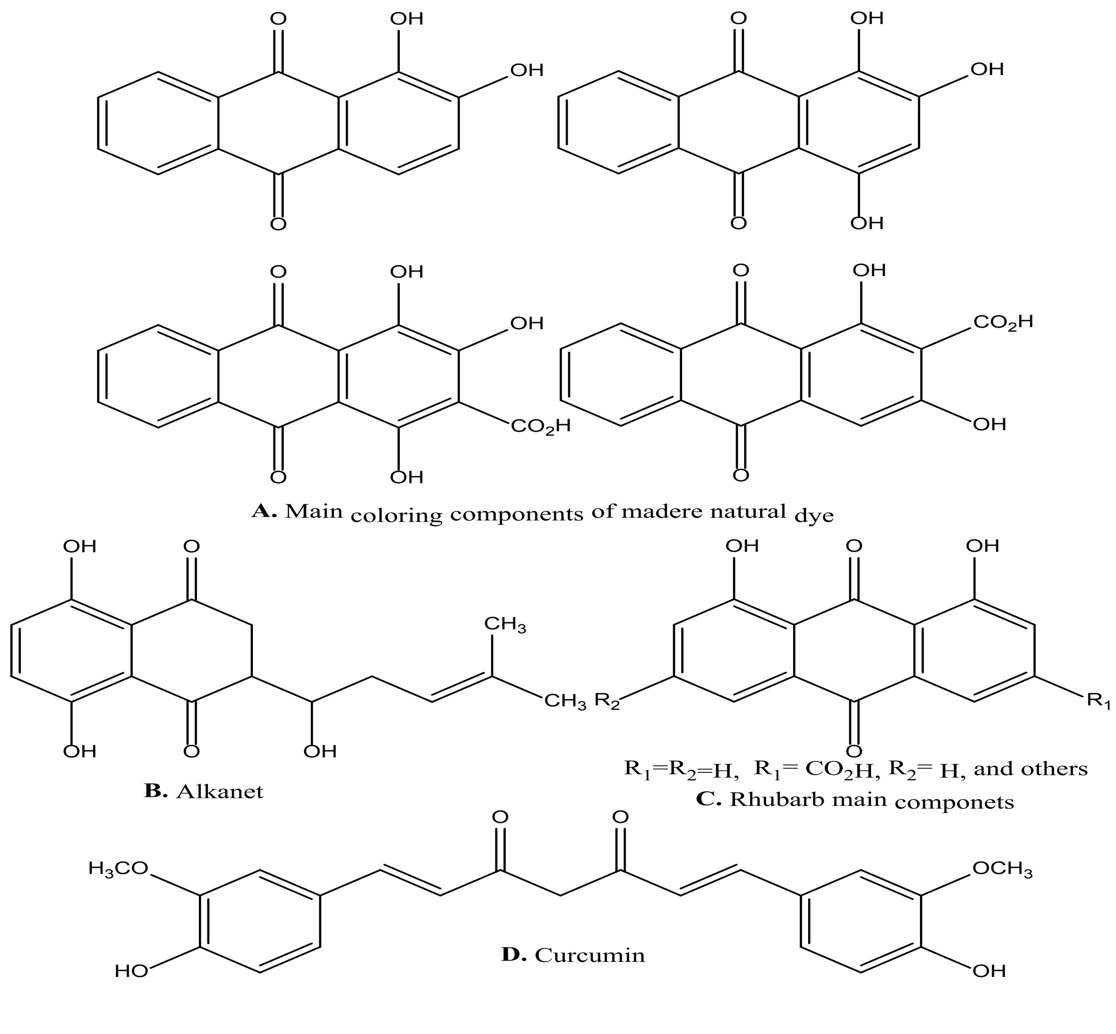


Fig. 1. The chemical structures of the natural dyes used.

For this purpose, a novel pretreatment method of cotton fabric using green bio-mordants composed of tannic acid and gelatin is presented. The pretreated fabrics were then dyed with Madder dye at different conditions, and the optimum dyeing condition was then applied on the pretreated cotton fabrics using different natural and the effect of dye structure on the fabric dyeability was investigated. Furthermore, the overall fastness properties of the dyed samples were evaluated.

Experimental

Materials

Cotton fabric

Mill-scoured and bleached cotton fabric (130 g/m^2) was obtained from Misr El-Mehala Co. (Clariant, Egypt). The fabric was further treated with a solution containing 3 g/l nonionic detergent (Hostapal CV, Clariant) and 5 g/l sodium carbonate at a liquor ratio of 50:1 for 1 h at the boil, after which time it was thoroughly rinsed and dried at room temperature.

Natural dyes

The natural dyes used in this study were madder (CI Natural Red 9), rhubarb (CI Natural Yellow 23), alkanet (CI Natural Red 20) and curcumin (CI Natural Yellow 3). These dyes were purchased from a local market in Egypt and were used as received.

Auxiliaries

Tannic acid, gelatin, sodium chloride and others were all laboratory grade chemicals.

Methods

Bio-mordanting

Cotton fabric was first treated with tannic acid at different concentrations (0-18% w/v) in a LR 1:20 and at 90 ^oC for 1 h. The treated fabric was then thoroughly rinsed with water and air-dried. The tannic acid treated fabric was then treated with gelatin at different concentrations(0-3% w/v) by pad-curing method at 120 ^oC for 5 min.

Dyeing with natural dyes

The dye solution was made by boiling 10 g powder of the natural dye in 100 ml distilled water with stirring for 1 h. The mixture was filtered through a mesh fabric with squeezing. The bio-mordanted fabric was dyed in a LR 1:20 of the dye solution at different pH values (3-8.23) for different durations (10-90 min) and different temperatures (25-100 ^oC) using different exhausting salt concentrations (0-10 g/L).

Washing

The dyed samples were rinsed with hot water, cold water, and soaped with 2 g/l nonionic detergent (Hostapal (CV, Clariant) at 60 ^oC for 30 min, thoroughly rinsed and air-dried.

Measurements

ATR-FT-IR (Perkin Elmer spectrum 100 FT-IR spectrometer) analysis wasperformed to chemical changes of the cotton fabric after being bio-mordanted. The relativecolor strength (K/S values) was assessed using theKubelka–Munk equation (1) [26].

$$\frac{K}{S} = \sqrt{\frac{(1-R)^2}{2R}} \quad \textbf{(1)}$$

where, R is the reflectance of colored samples and K and S are the absorption and scattering coefficients, respectively. The values of K/S were measured at the maximum wavelength of absorption of madder at 355 nm, of rhubarb at 355 nm, of alkanet at 355 nm, and of curcumin at 445 nm.

Fastness testing

The dyedsamples were tested, after washing-off using 2 g/L nonionic detergent (Hostapal CV) at 60^oC for 30 min, according to ISO standard methods [27]. The specific tests were; ISO 105-C02 (1989), color fastness to washing and ISO 105-E04 (1989), color fastness to perspiration, ISO 105-X12(1987), and AATCC test method 16-1993, color fastness to light.

Results and Discussion

It was hypothesized that bio-mordanting of cotton fabric using tannic acid and gelatin would render cotton fabric cationic at acidic pH, and thus, good dyeability with natural dyes would be expected. Therefore, and aiming at presenting a green dyeing approach of cotton fabric with natural dyes and without impacting the environment with metal ions, the present work was investigated using four different natural dyes containing phenolic OH groups and carboxyl groups amenable for bonding with the bio-mordanted fabric. The factors that may affect the pretreatment and dyeing are investigated as a continuous process, i.e., bio-mordanting followed by dyeing with madder natural dye and thus the color strength of the dyed fabric is taken as a function of the success of the pretreatment and dyeing process.

Effect of tannic acid concentration

Pretreatment of cotton fabric would lead to the absorption and fixation of tannic acid inside the fabric by virtue of several hydrogen bonds between the hydroxyl groups present in the cotton fabric and those of polyphenolics present in tannic acid (Scheme 1). Fig. 2 shows the effect of tannic acid on the bio-mordanting of cotton fabric, as reflected by its color strength. It is clear that a specific concentration of tannic acid (15%) would furnish the necessary binding sites onto the fabric. Lower or higher concentrations either not enough or excess in crosslinking the fabric, and thus, a lower dyeability is expected in both cases.

Effect of gelatin concentration

Gelatin is the second component of the bio-mordanting of cotton fabric is the main coating layer that would get crosslinked with the previous tannic acid layer (Scheme 2). As shown in Fig. 3, a similar dyeing behavior with the case of tannic acid effect is also observed. It is shown that 2% of gelatin was enough for having the best dyeability of the bio-mordanted fabric. At zero concentration of gelatin, the dyeability of the tannic-treated cotton was higher than that of cotton fabric, indicating the favoring effect of the tannic pretreatment step. As the concentration of gelatin increases, the color strength increases up to 2% gelatin above which a decline in color strength is observed.

Effect of pH

The effect of the dyeing pH is crucial in this study. As shown in Fig. 4, it is clearly observed that acidic pH (pH 3-4.5) in general is favorable for having a higher color strength of dyed fabric compared with those of slightly acidic to alkaline (pH 5.99-8.23) and the highest color strength value was obtained at pH to 4.5. This result reflects the mechanism of dye fixation that would follow ionic bond formation between the hydroxyl or carboxyl groups present in the dye molecule and the protonated bio-mordanted cotton fabric (Scheme 3).

Effect of dyeing temperature

The effect of dyeing temperature on the color strength obtained is shown in Fig. 5. I t is clear that the color strength of the dyed fabric increases with increasing the dyeing temperature with a remarkable increase observed beyond 70 ^{o}C to have its maximum at 100 ^{o}C. It is believed that increasing the temperature enhances the fiber swelling and the dye diffusion inside the fabric and hence, increased the color strength of the dyed fabric.

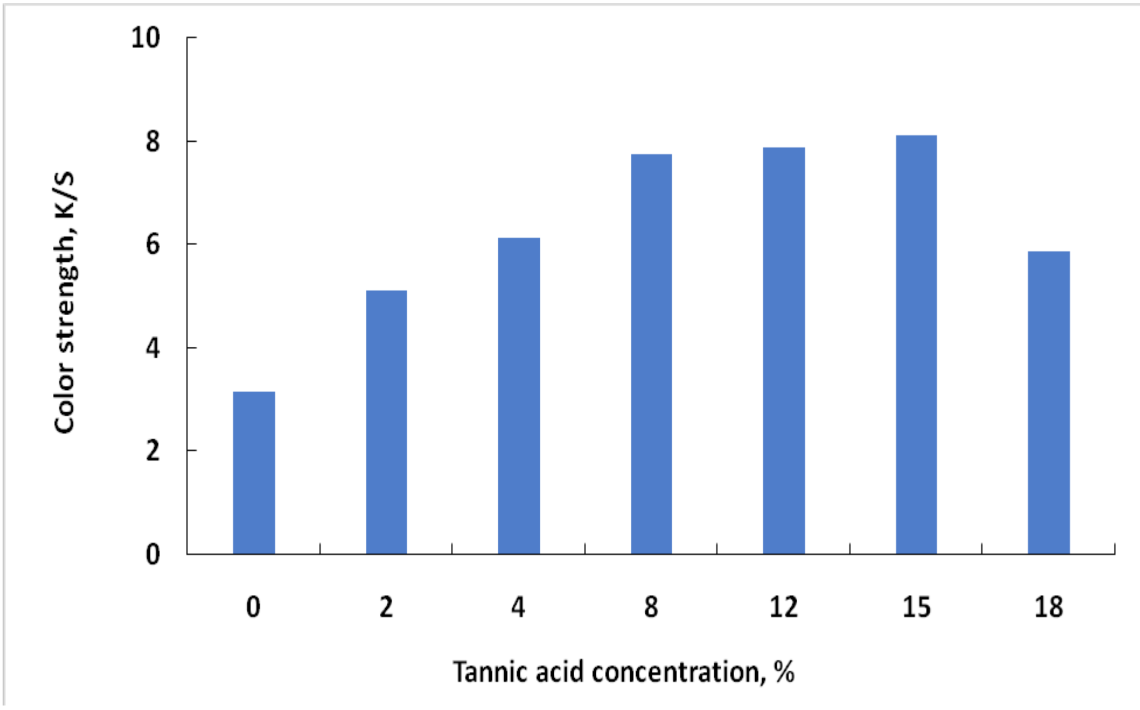
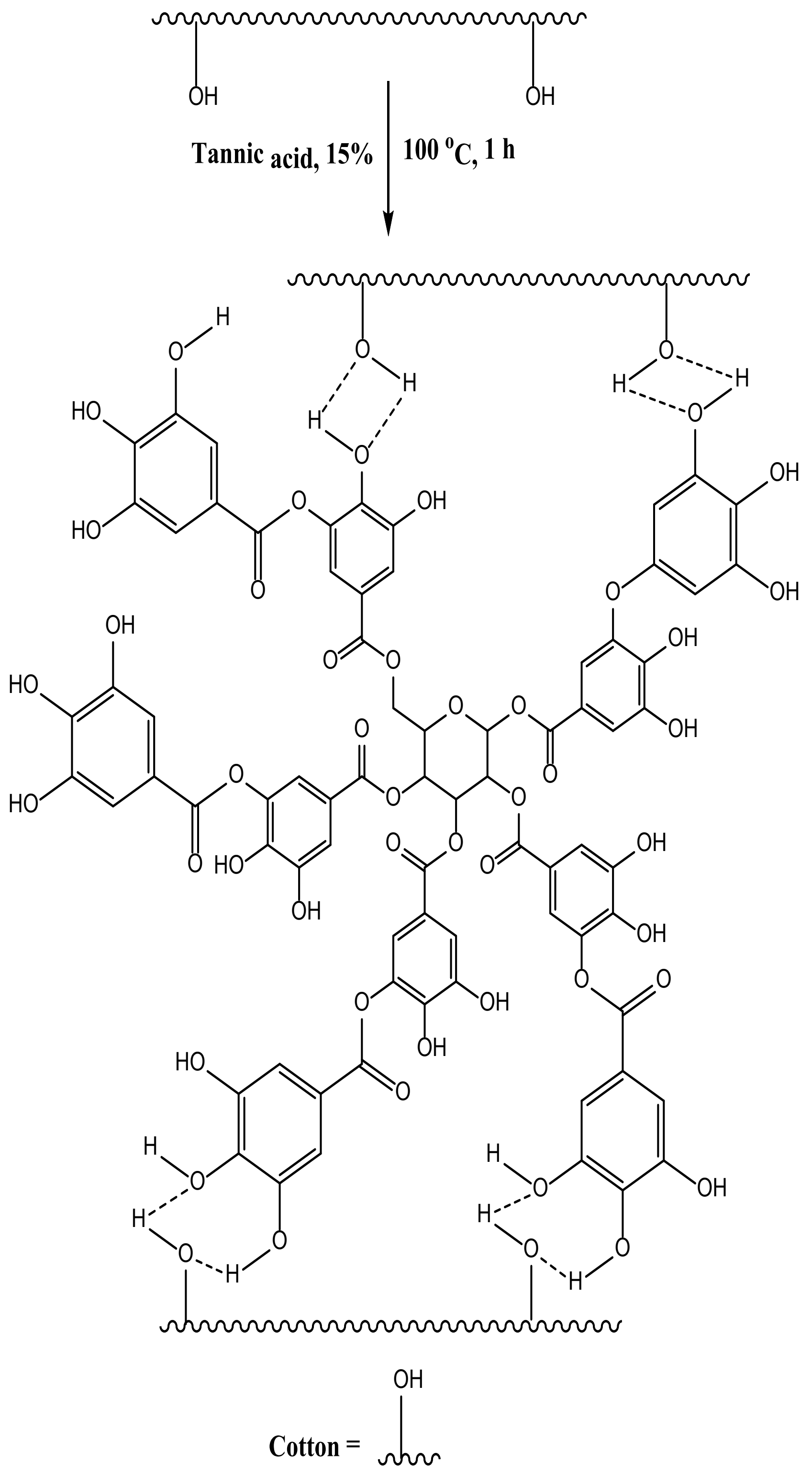


Fig. 2. Effect of tannic acid concentration in the bio-mordanting impact on the color strength of dyed fabric. Conditions: Treatment with tannic acid, LR 1:20, 90 ^{o}C, 1 h, then padding (2-dip-2-nip) in a 2% gelatin, curing at 120 ^{o}C for 5 min, dyeing with madder natural dye, LR 1:20, pH 4,5, 100 ^{o}C for 1h.



Scheme 1. Hydrogen bond fixation of tannic acid with cotton fabric.

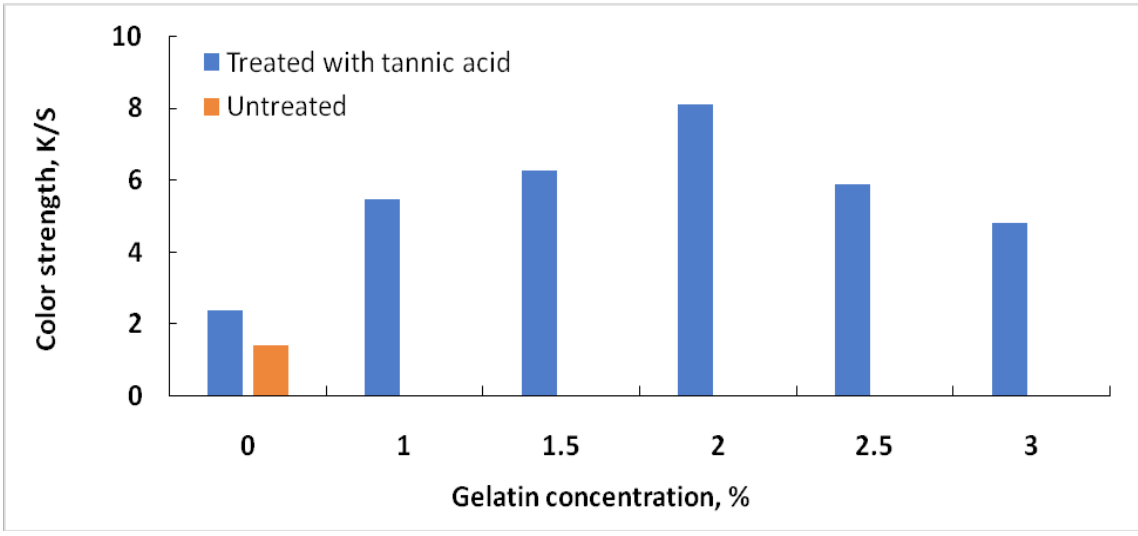
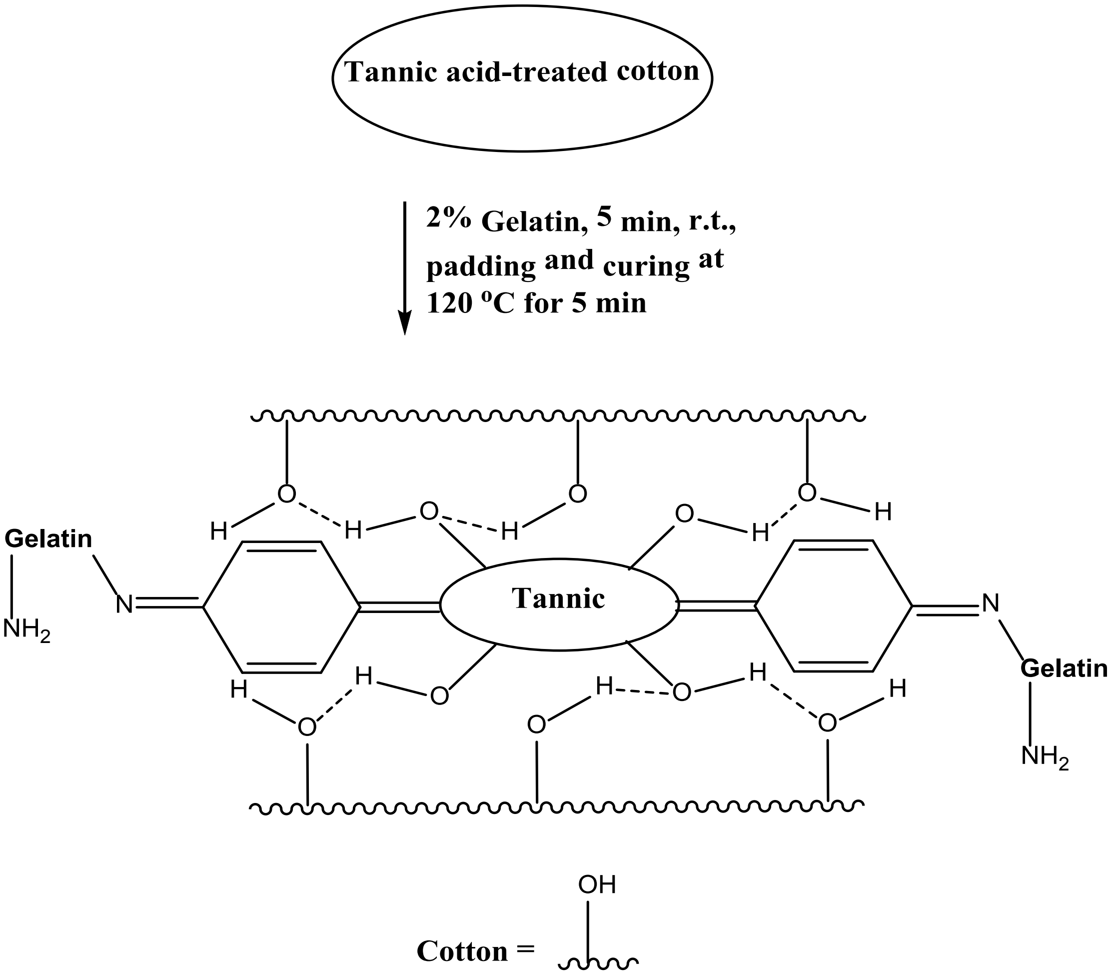


Fig. 3. Effect of gelatin concentration in the bio-mordanting impact on the color strength of dyed fabric. Conditions: Treatment with tannic acid 15%, LR 1 : 20, 90 °C, 1 h, then padding (2-dip-2-nip) in gelatin, curing at 120°C for 5 min, dyeing with madder natural dye, LR 1:20, pH 4,5, 100 °C for 1h.



Scheme 2. Crosslinking of gelatin with tannic acid treated-cotton fabaric.

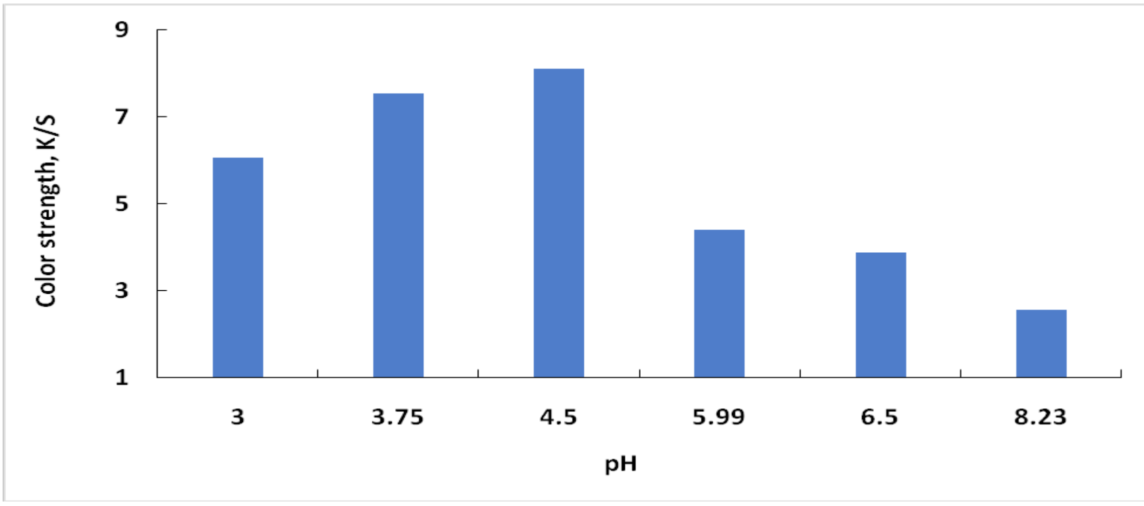
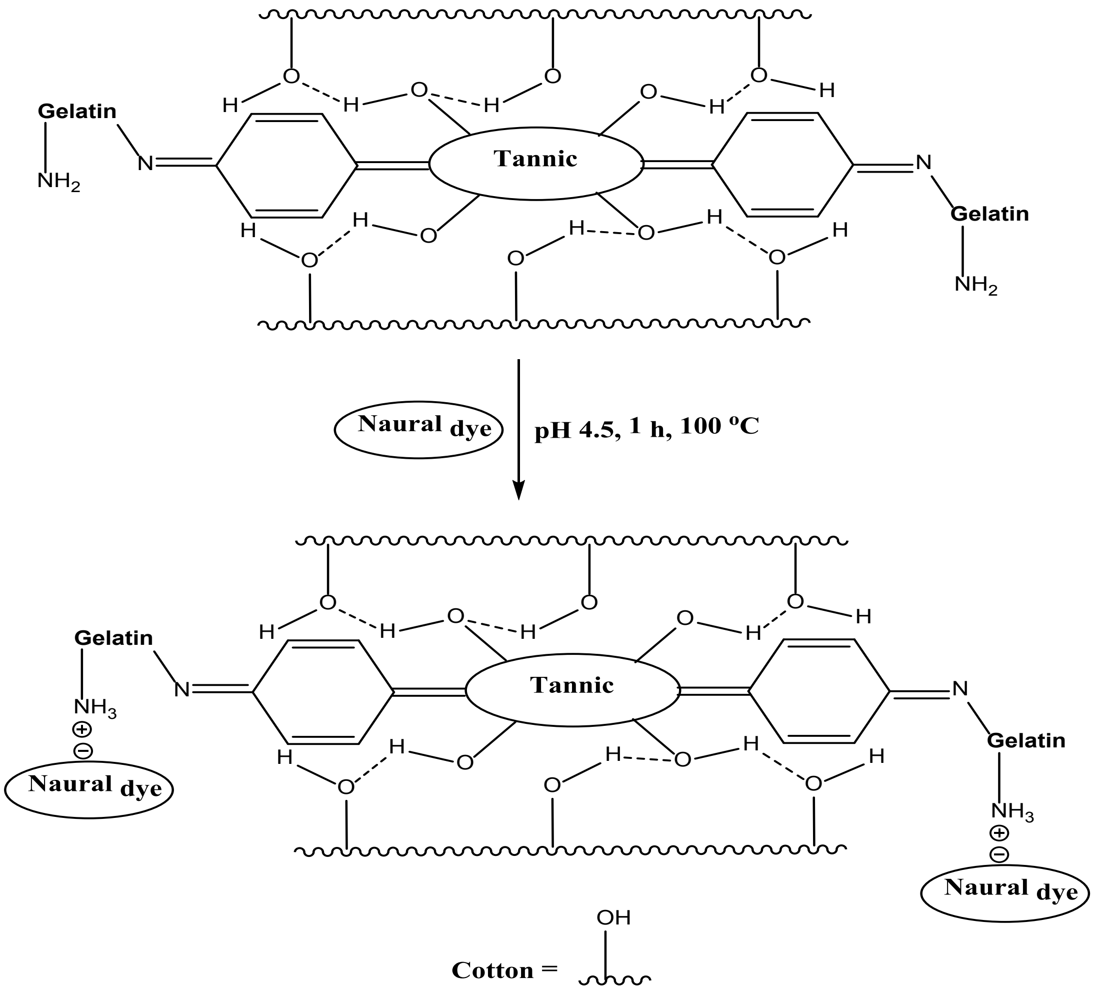


Fig. 4. Effect of dyeing pH on the color strength of the dyed fabric. Conditions: Treatment with tannic acid 15%, LR 1:20, 90 °C, 1 h, then padding (2-dip-2-nip) in a 2% gelatin, curing at 120 °C for 5 min, dyeing with madder natural dye, LR 1:20, 100 °C for 1h.



Scheme 3. Ionic bond fixation of natural dyes with gelatin-tannic acid treated cotton fabaric.

Effect of dyeing time

The effect of dyeing time on the dyeability of bio-mordanted fabric is shown in Fig. 6. As expected, the process of dyeing proceeds by fiber swelling, dye absorption, and diffusion inside the fiber, followed by its fixation. Therefore, as the time increases, the color strength increases, as shown in the figure, in which a plateau is attained after 60 min, indicating that the dyed fabric has become dyed enough with the dye molecule.

Effect of salt concentration

The effect of salt concentration in the dyebath on the dyeability of the bio-mordanted fabric is shown in Fig. 7. It is clear that the color strength of the dyed fabric without salt addition is slightly better than in the presence of salt. Adding a higher concentration of salt (above 8g/L) decreased the color strength obtained. This result emphasizes again the absorption and fixation of the dye inside the fabric are pH-dependent with an ionic mechanism.

Application on different natural dyes

The optimum dyeing conditions concluded from the above studies were applied using four natural dyes for dyeing the untreated and bio-mordanted cotton fabrics. Fig. 8 shows how the method presented was successful for all dyes and depending on their chemical structures. Images of the dyed samples bio-mordanted and untreated are shown in Fig. 9. The overall results indicate that the fixation of natural dyes on the bio-mordanted fabrics depends on the extent of dye acidity that would lead to a higher dye uptake. Interestingly, the result of K/S values is in a good agreement with the acidity of the dyes, and the values of K/S follow the order madder> rhubarb>alkanet>curcumin.

FTIR of the bio-mordant cotton

ATR-FTIR (Fig. 10) analysis was used to show the changes made in the cotton fabric before tannic acid-gelatin bio-mordanting and after. As shown in the spectra in the region between 3500 to 3100 cm^{-1}, the hydrogen bonding between OH and N-H groups upon bio-mordanting decreased the bandwidth compared with the band of cotton alone before bio-mordanting. This effect in decreasing the bandwidth upon bio-mordanting is also observed at the characteristic pyranose C-O-C stretching vibration of cotton fabric. The bandwidth of FTIR is in the order C (cotton) > CT (cotton-tannic) > CTG (cotton-tannic-gelatin). This suggests that bio-mordanting starting by tannic and ended by gelatin increases the hydrogen bonding process with dehydration possibility upon curing at 120 ^{o}C. The amide bands that typical of gelatin appears at 1660 cm^{-1} (C=O stretching) and 1554 cm^{-1} (N-H deformation) [28]. Interestingly, these peaks are shifted to lower wavenumbers and clearly observed in the CTG sample indicating the involvement of gelatin in a reaction with tannic acid-treated fabric via the formation of imine bond (Schiff base). The imine band vibration band might be overlapped with C=O amide as it also appears nearby 1640 cm^{-1}. The overall result suggests the formation of a crosslinked layer between cotton-tannic sample and gelatin to form the stable un-washable bio-mordanted CTG sample. Thus, the mechanism of bio-mordanting process of cotton fabric with tannic acid-gelatin can be tentatively sketched in Schemes 2 and 3 mentioned above.

Fastness properties

Table 1 shows the fastness characteristics of the dyed bio-mordanted cotton fabrics using different natural dyes. The dyed samples reveal very good to excellent fastness properties using madder and alkanet natural dyes and good to excellent fastness properties using rhubarb and curcumin natural dyes. Interestingly, light fastness properties were very good for all dyes, indicating the suitability of bio-mordanting for better dyeing as a result of chemical bonds between the bio-mordanted cotton fabric and the dye molecule.

Conclusion

Complete green dyeing of bio-mordanted cotton fabric with different natural dyes is presented for the first time. The pH-dependent dyeability with the highest at pH 4.5 indicates the ionic mechanism between the dye and the bio-mordanted fabric. Madder natural dye was the best for having a high dye uptake and very good to excellent fastness properties to emphasize the chemical structure effect and the importance of having high acidic groups in the dye molecule. Hence, the present ecological dyeing approach of cotton fabric with natural dyes without the use of metal ion salts would further inspire scientists for more environmentally friendly successful work ahead in the field of textile coloration.

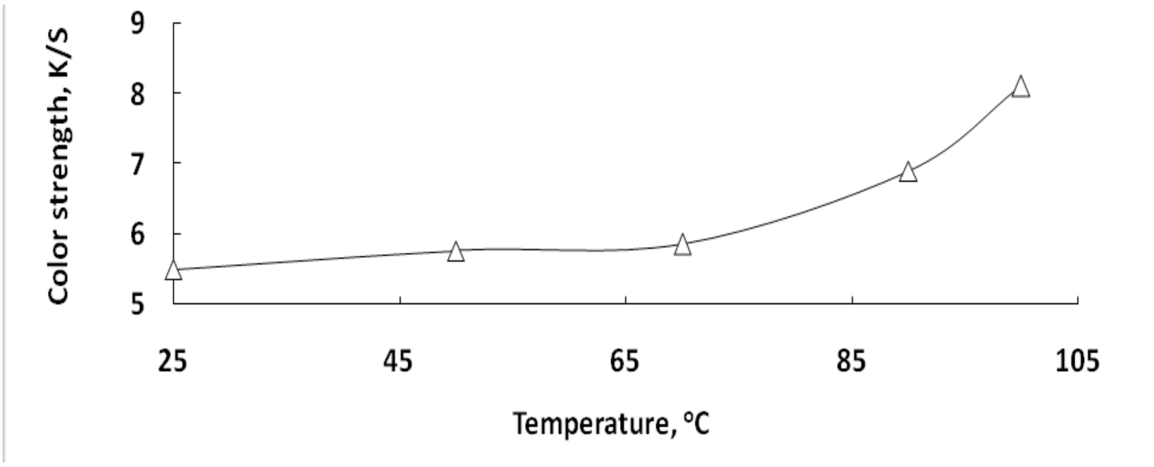


Fig. 5. Effect of dyeing temperature on the color strength of the dyed fabric. Conditions: Treatment with tannic acid 15%, LR 1:20, 90 °C, 1 h, then padding (2-dip-2-nip) in a 2% gelatin, curing at 120 °C for 5 min, dyeing with madder natural dye, LR 1:20, pH 4.5, 1h.

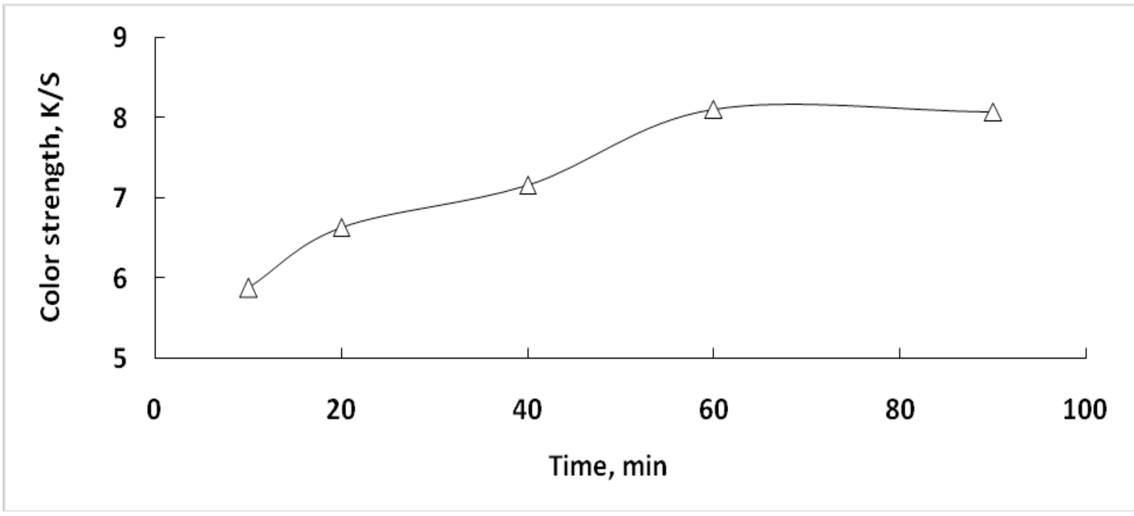


Fig. 6. Effect of dyeing time on the color strength of the dyed fabric. Conditions: Treatment with tannic acid 15%, LR 1:20, 90 °C, 1 h, then padding (2-dip-2-nip) in a 2% gelatin, curing at 120 °C for 5 min, dyeing with madder natural dye, LR 1:20, pH 4.5, at 100 °C.

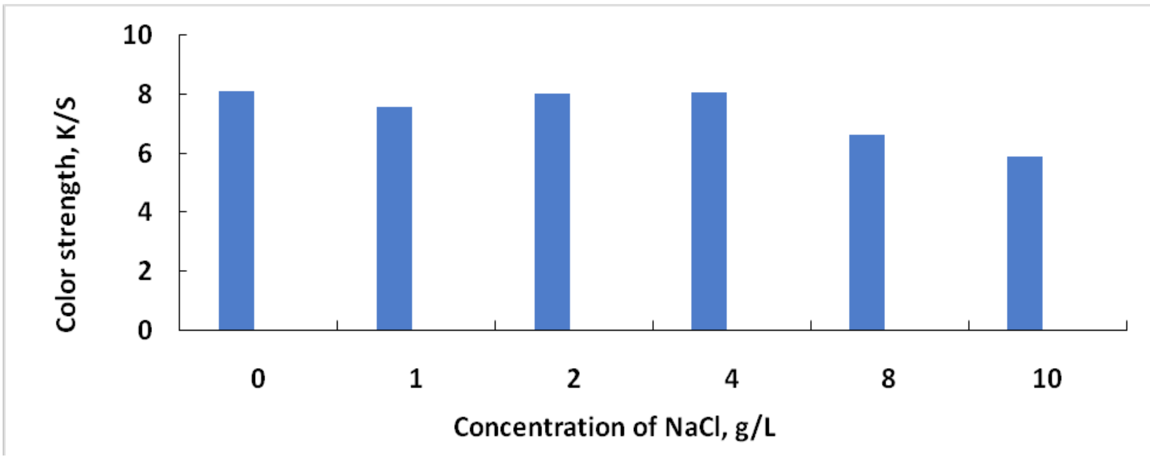


Fig. 7. Effect of salt concentration on the color strength of the dyed fabric. Conditions: Treatment with tannic acid 15%, LR 1:20, 90 °C, 1 h, then padding (2-dip-2-nip) in a 2% gelatin, curing at 120 °C for 5 min, dyeing with madder natural dye, LR 1:20, pH 4.5, at 100 °C, 1 h.

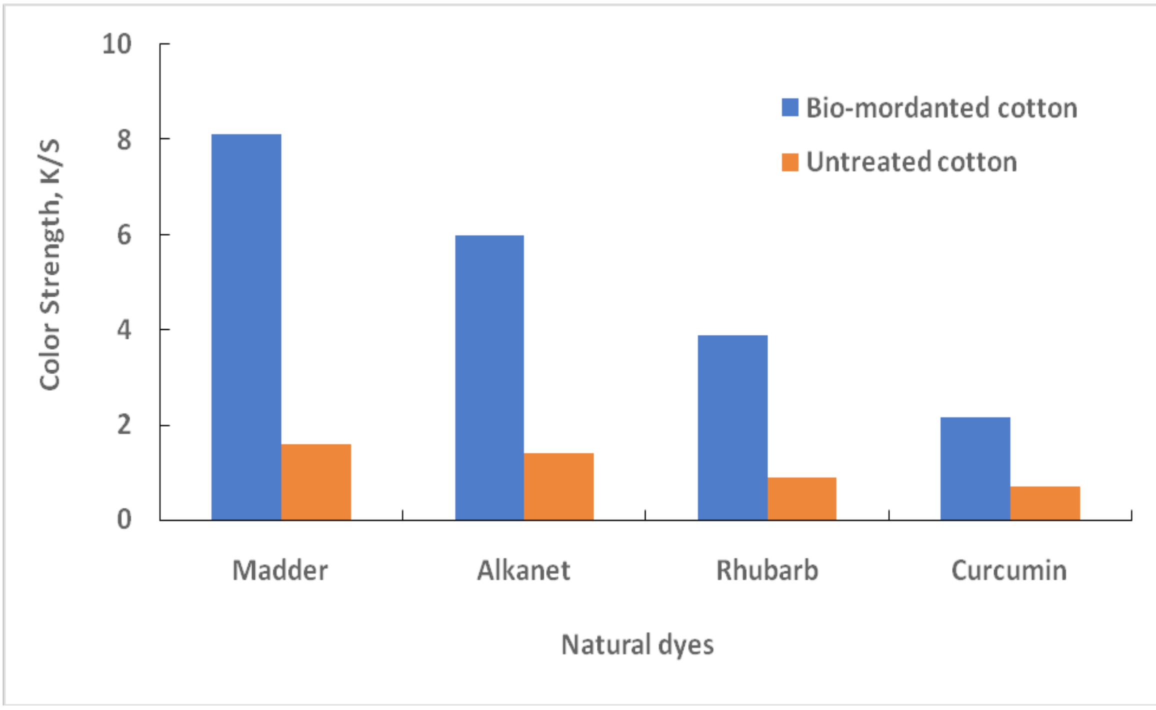


Fig. 8. Dyeing of cotton fabric with different dyes at the optimum conditions.

Conditions: Treatment with tannic acid 15%, LR 1:20, 90 °C, 1 h, then padding (2-dip-2-nip) in a 2% gelatin, curing at 120 °C for 5 min, dyeing with natural dye, LR 1:20, pH 4.5, at 100 °C, 1 h.

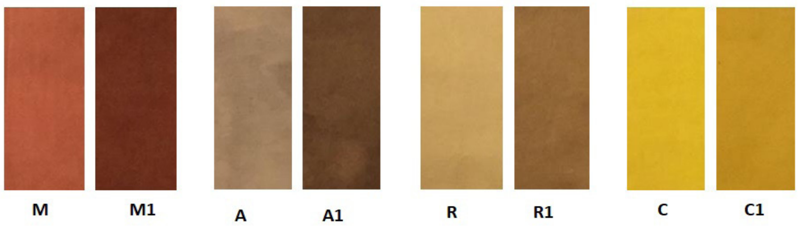


Fig. 9. Color images of dyed untreated and bio-modanted cotton fabrics using mader (M = untreated, M1= bio-mordanted), alkanet (A = untreated, A1= bio-mordanted), rhubarb (R = untreated, R1= bio-mordanted), and curcumin (C = untreated, C1= bio-mordanted).

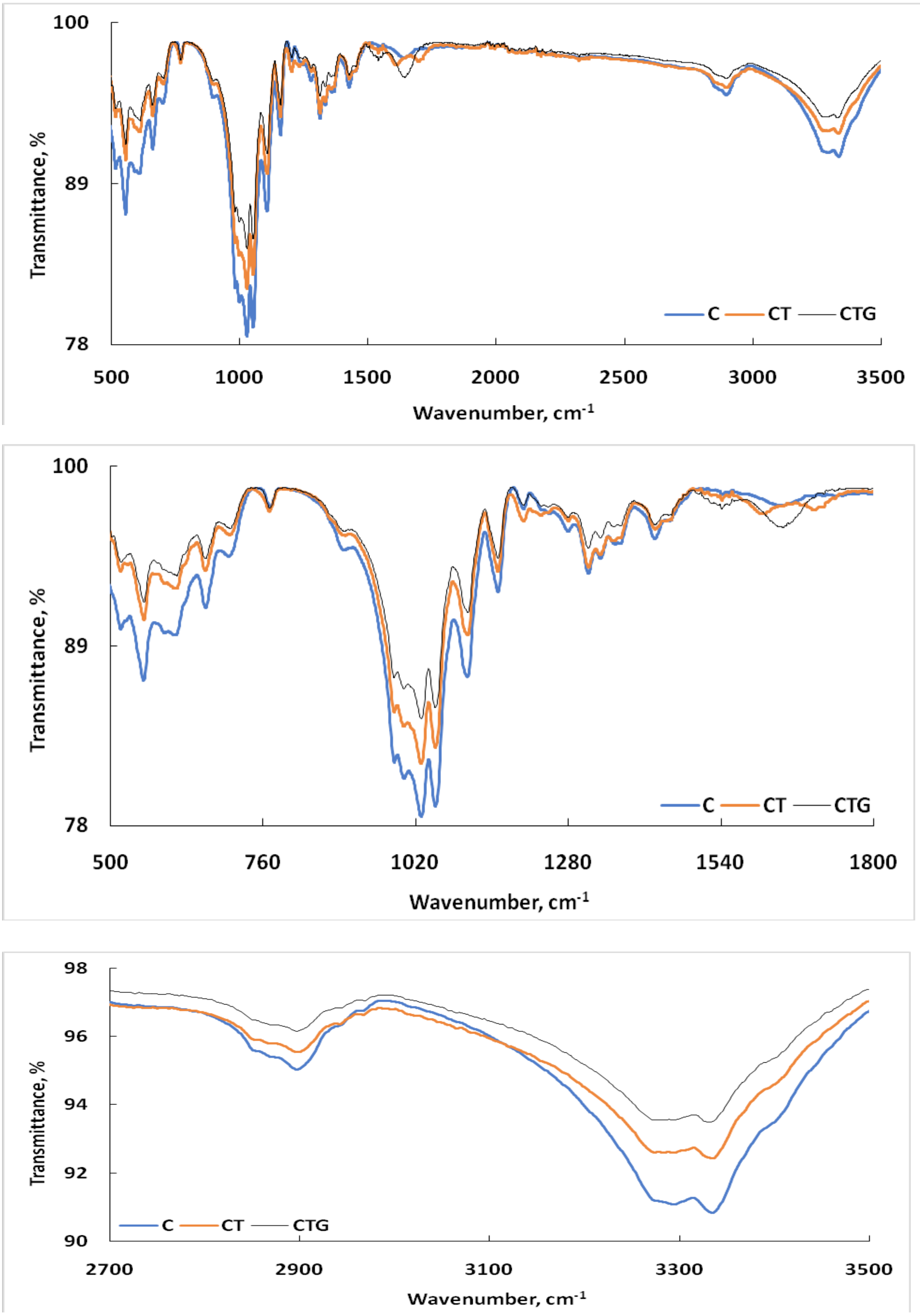


Fig. 10. ATR-FT-IR of cotton (C), cotton-tannic acid treated (CT), and cotton-tannic-gelatin treated samples (Gl). Treatment with tannic acid 15%, LR 1:20, 90 °C, 1 h, then padding (2-dip-2-nip) in a 2% gelatin, curing at 120 °C for 5 min.

TABLE 1. Color fastness of the dyed bio-mordanted cotton fabrics using different natural dyes.

Dyes	Rubbing fastness		Washing fastness			Perspiration fastness						Light fastness
						Acid			Alkali			
	Dry	Wet	A	SC	SW	A	SC	SW	A	SC	SW	
Madder	4-5	4	4-5	4-5	4-5	4-5	4-5	4-5	4	4	4	6
Alkanet	4-5	4	4	4-5	4	4-5	4	4	4	4	4	6
Rhubarb	3-4	3-4	3-4	3-4	3-4	3-4	3-4	3-4	3-4	3-4	3-4	6
Curcumin	4	3-4	3-4	4	3-4	4	4	3-4	4	3-4	4	6

Alternation (A), Staining on wool fabric (SC), Staining on cotton fabric (SC).
Wash & Rubbing and perspiration on gray scale: 1, poor; 2, fair; 3, good; 4, very good; 5, excellent.
Light fastness on blue scale: 1, very poor; 2, poor; 3, fair; 4, moderate; 5, good; 6, very good; 7, excellent; 8, outstanding.

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