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Novel Green Coloration of Cotton Fabric. Part II: Effect of Different Print Paste Formulations on The Printability of Bio-mordanted Fabric with Madder Natural Dye

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> GELATIN-tannic acid (bio-mordant) treated cotton fabric was printed with natural and synthetic thickeners. Madder natural dye was used with different thickeners include meypro gum, carboxymethyl cellulose, sodium alginate, or acrylate-based synthetic thickener. Different factors such as pH, steaming temperatures, time, auxiliaries (urea, salt, and binder) presence or absence in the print paste formulations were investigated. The data indicated good quality prints using natural thickeners and without auxiliaries compared with the synthetic one. The printed samples reveal very good to excellent fastness properties. Hence, the present process of printing cotton fabric with green print paste formulation and without auxiliaries may find wide application in textile coloration.

Keywords: Ecological, Green printing, Natural dye, Cotton, Bio-mordanting, Gelatin-tannic acid.

Introduction

Textile coloration by printing is a wet process in which the printing paste is composed of water, dye or pigment, thickening agent, and other textile auxiliaries. Therefore, unfixed color or, together with the washed-off components used in the print paste, is heavily polluting the textile effluents [1]. Accordingly, environmental awareness has been growing in textile printing by devising new methods, the substitution of chemicals and/or using new techniques [1-13].

The use of natural dyes in the printing of textiles has been growing. Bioscoured and bleached jute fabric was printed with the natural thickener (guar gum) [14]. Printing with alkanet and rhubarb natural dyes using the technique of pigment printing was also reported [15]. Studies on the printing of chitosan-treated cotton fabric with curcumin have been reported [16].

In continuation of our interest in the green coloration of cotton fabric with natural dyes, the newly green pretreated cotton fabric reported in part one [17] of this work was printed with madder natural dve (CI Natural Red 9, Fig. 1). Madder natural dye was selected in this study as it contains carboxylic and acid hydroxyl groups [18, 19]. Thus, a novel and simple printing paste formulations containing madder natural dye and different thickeners without binder or other conventional printing paste chemicals such as urea and ammonium sulfate are presented. Comparative performance of different thickeners on the printability of cotton fabrics under different printing conditions was investigated. Also, the overall fastness properties of the printed fabrics were evaluated.

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Fig. 1. Main coloring components of madere natural dze,

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 dye used in this study were madder (CI Natural Red 9) and was used as received.

Thickening agents

Meypro gum NP-16 (Meyhall) thickening agent was used at a concentration of 8%. Carboxymethyl cellulose (CMC) sodium salt (medium viscosity) was supplied by BDH laboratory supplies (England) and used at a concentration of 4%. Medium-viscosity sodium alginate was supplied by Ceca Kolloid Chemie, Paris, France, and used at a concentration of 4%. EBCA print 160 Lc (acrylic-based synthetic thickener) was used at a concentration of 3%.

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Auxiliaries

Binder BD (liquid) was kindly supplied by Daico Company, Cairo, Egypt. Urea, diammonium sulfate, and others were all laboratory grade chemicals.

Methods

Print paste formulations

The printing paste formulations were made for printing the cotton fabrics with natural dyes in the presence and absence of printing auxiliaries as follow:

Print paste with auxiliaries

Natural dye solution (50 ml, 10% w/v) + thickener (50 ml, x% w/v) + 4 g urea + 1.25 g diammonium sulphate + binder BD (liquid) 12 ml. X=8% for meypro gum; 4% for sodium alginate, 3% for synthetic thickener; and 4% for CMC.

Print paste without auxiliaries

Natural dye solution (50 ml, 10% w/v) + thickener (50 ml, x% w/v). X=16% for meypro gum; 8% for sodium alginate, 6% for synthetic thickener; and 8% for CMC.

The print paste with auxiliaries and without were made as follows. The dye solution was made by soaking 10 g powder of natural dye in 100 ml distilled water with stirring at room temperature for 30 min. The mixture was filtered through a mesh fabric with squeezing. Then, urea and diammonium sulfate by the amount mentioned above was dissolved in 50 ml dye solution(A). Thickener solution (B) was made by dissolving the amount mentioned in 50 ml distilled water. The print paste was then made by mixing A and B together with a 12 ml binder with stirring at room temperature for 10 min. Finally, the pH of the paste was adjusted to the desired pH by adding drops of glacial acetic acid. This procedure was followed except adding auxiliaries for making the print paste without auxiliaries.

Printing technique

The aforementioned printing pastes of different pHs were applied to cotton fabrics according to the conventional flat screen-printing method [20]. Fixation of the prints was made by drying at room temperature followed by steam fixation at different temperatures and different time intervals.

Washing

The prints made were rinsed with hot water, cold water, and soaped with 2 g/l nonionic detergent (Hostapal (CV, Clariant) at 60 °C for 30 min, thoroughly rinsed and air-dried.

Color measurements

The color strength expressed as (K/S) values of the printed samples were measured by using a Hunter Lab Ultra Scan® PRO (USA) at the maximum wavelength of the dye used. The relative color strength (K/S values) was assessed using the Kubelka–Munk equation (1) [21]

$$K/S = \sqrt{((1-R)^2/2R)}$$
 (1)

where, R is the reflectance of colored samples and K and S are the absorption and scattering coefficients, respectively.

Fastness testing

The printed samples were tested, after washingoff using 2 g/L nonionic detergent (Hostapal CV) at 60°C for 30 min, according to ISO standard methods [22]. 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

The present work was to study the possibility of green printing of cotton fabrics using different thickeners containing madder natural dye aiming at revealing the mechanism of printing and whether it follows the concept of pigment printing that necessities the use of binder and other printing auxiliaries or follows ionic mechanism with a better printing data without using auxiliaries. Hence, the present work was motivated by the successful formation of bio-mordanted cotton fabric [Fig. 2] to explore its printability using print paste formulations in absence and presence of printing auxiliaries. For this purpose, four thickeners, three natural and one synthetic were investigated. These thickeners are, meypro gum which is a non-ionic galactomannan polysaccharide [23], solidum alginate (anionic), CMC (anionic) and acrylicbased synthetic thickener (anionic).

Effect of the print paste formulation

Fig. 3 shows the effect of print paste formulations on the printability of cotton fabrics (control and bio-mordanted samples) with madder natural dye. It is observed that the color strength value gets enhanced upon bio-mordanting for both print paste formulations, indicating that the bio-mordanting process has created additional binding sites for the fixation of the dye. However, the enhanced value of the color strength obtained using the print paste without auxiliaries is far better for the bio-mordanted sample (535%) compared with those obtained using the print paste with auxiliaries (27%). This result indicates that the printing mechanism using the print paste that contains auxiliaries would follow pigment fixation mechanism for the control sample and pigment and ionic fixation mechanisms for the biomordanted sample. This result is confirmed by the huge enhancement in the color strength value for the bio-mordanted samples using the print paste without auxiliaries compared with the control one, indicating an ionic mechanism between the dye and the fabric.

Effect of pH

Fig. 4 shows the effect of print paste (without auxiliaries) pHs on the printability of biomordanted cotton fabrics with madder natural dye. It is observed that the color strength value increases upon decreasing the pH up to 4,8, and a further decrease in the pH is reverting the trend of fabric printability. The acidic pH impact on enhancing the color strength indicates ionic bond fixation of the dye with the bio-mordanted fabric. However, the lower value of the color strength upon further increases in the acidity might be attributed to the protonation of the natural dyes, and thus ionic repulsion between the dye molecule and the protonated bio-mordanted cotton takes place.



Fig. 2. Bio - mordanted cotton fabric.



Fig. 3. Effect of the print paste formulation on the color strength the printed fabric. Conditions: Meypro gum containing madder natural dye, pH 4.8, steaming at 102 °C for 15 min.



Fig. 4. Effect of the print paste pH on the color strength the printed fabric. Conditions: Meypro gum containing madder natural dye, steaming at 102 °C for 15 min.

Effect of steaming time

Fig. 5 shows the effect of steaming time on the printability of bio-mordanted fabric using print paste without auxiliaries and madder natural dye. It is shown that increasing the steaming time has led to an increase in the color strength obtained up to 15 min above which the color strength started to decline. Prolongation of the steaming time could act as reversibly in destabilizing the dye-fiber bonds and thus lower fixation.

Effect of steaming temperature

Fig. 6 shows the effect of steaming temperature. As shown in the figure, the best data obtained were those obtained at 110 °C above, which a decline in the color strength values is observed, indicating that penetration, diffusion, and fixation of the dye with the fabric necessitates certain temperature above which a reverse action takes place.

Effect of thickener type

Different thickeners were used to elaborate on their structure effect on the printability of biomordanted fabric. Figs. 7, 8 show that the best data obtained was those due to the use of meypro gum was the best among all thickeners, and the data obtained follows the following order. Meypro gum (non-ionic)>CMC (anionic)>synthetic thickener (anionic) >sodium alginate (anionic). This result might be attributed to the charge of the thickener as the more the negative charge in the surface of the thickener, the lower will be the dye uptake by the fabric. Interestingly, control cotton fabric showed low printability data and irresponsible with the thickener used, indicating the absence of a chemical bond. However, biomordanted samples were responsive to the thickener used.

Fastness properties

Table 1 shows the fastness characteristics of prints made by different thickeners containing madder natural dye. The printed samples reveal very good to excellent fastness properties, and the best data obtained was in accordance with the charge of the thickener in which meypro gum thickener was the best. The overall result of the fastness properties indicates the existence of chemical bonds between the bio-mordanted cotton fabric and the dye molecule.

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Fig. 5. Effect of steaming time on the color strength the printed fabric. Condition: Meypro gum containing madder natural dye, pH 4.8, steaming at 102 °C.



Fig. 6. Effect of steaming temperature on the color strength the printed fabric. Condition: Meypro gum containing madder natural dye, pH 4.8, steaming for 15 min.

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Fig. 7. Effect of thickener type on the color strength of the printed fabric. Condition: Meypro gum containing madder natural dye, pH 4.8, steaming at 110 °C for 15 min.



TABLE 1. Color fastness of the printed bio-mordanted cotton fabrics using different thickeners. 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.

Thickeners	Rubbing fastness		Washing fastness			Perspiration fastness						Light
	Dama	Dry Wet	А	SC	SW	Acid			Alkali			fastness
	Dry					Α	SC	SW	Α	SC	SW	
Meypro gum	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	4-5	6
CMC	4-5	4-5	4	4-5	4	4-5	4-5	4	4-5	4-5	4	6
Sodium alginate	4-5	4-5	4	4-5	4	4-5	4	4	4-5	4-5	4-5	6
Synthetic thickener	4-5	4	4	4	4	4	4	4	4	4	4	5-6

Conclusion

An ecological printing process of biomordanted cotton fabric with madder natural dye using green print paste without auxiliaries is presented for the first time. Meypro gum as a natural non-ionic thickening agent was suitable for good prints, and the effect of pH, as well as the effect of print paste formulation, indicated that the fixation of the dye with the bio-mordanted fabric proceeds via ionic mechanism between the positively charged bio-mordanted fabric and the negatively charged dye. The printed samples reveal very good to excellent fastness properties. Hence, the present process of printing cotton fabric with green print paste formulation and without auxiliaries may find wide application in textile coloration.

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