IMPROVEMENT OF DYEING PROPERTIES OF SOME EGYPTIAN COTTON VARIETIES USING CURCUMINE NATURAL DYE BY ENZYMATIC PRETREATMENT

SALEH, S. M[1]., AZZA A. MAHMOUD[1]., AND KH. EL-BADRY[2]

1. Cotton Research Institute, Agricultural Research Center. Giza, Egypt.

2. Faculty of Education, Helwan University, Cairo, Egypt.

(Manuscript received 15 February 2005)

Abstract

This investigation deals with improvement of dyeing properties of Egyptian cotton fabrics of varieties namely Giza 83 and Giza 89, with natural dye from a yellow pigment fraction isolated from the rhizomes curcuma of longa by enzymatic pretreatment. The dyeing technique was performed by simultaneous mordanting technique (sample-dye-mordant) without enzyme, and after the enzymatic treatment. Different mordants namely iron (II) sulphate; alum, potassium dichromate, and copper sulphate were applied in this technique. The mechanism of dyeing is discussed. Dyeing performance in terms of fastness properties and color parameters are studied.The results obtained revealed that the simultaneous mordanting technique(sample-dye-mordant) after the enzymatic treatment resulted in an increase in dye uptake in all cases compared with the untreated or the simultaneous mordanting technique without enzymatic pretreatment samples, and did not affect fastness properties. The color strength (K/S) values, fastness properties and also the response to dyeing were higher for Giza 89 than Giza 83 due to its higher hydroxyl groups on the fabric surface causing increase in the charged sites by using the lone pair of electrons on the functional groups between the molecular structures of the reactants.

INTRODUCTION

There is an interest in the dyeing of textile fibers with natural dyes, on account of their high compatibility with the environment Chavan (1998), and Mahate *et al.* (2003).

There were more interests have been devoted to the use of natural dyes for dyeing and printing of textiles Das (1992). Karyanini et al. (1998) reported that natural dyes are less toxic, non pollutant, very brilliant according to Nishida (1992), rare idea and includes allergic reactions according to Gupta *et al.* (1998).

Problems in dyeing with natural dyes are related to the low exhaustion of the dyes and to the fastness of the dyed fabrics, Grierson *et al.* (1985), Storey (1985), King *et al.* (1970), Tsatsaroni et al. (1994) and Saleh *et al.* (2003).

Attempts to overcome these problems have been mainly focused on the use of metallic salts as mordants, which are traditionally used to improve fastness properties or exhaustion and to develop different shades with the same dye.

Enzymatic treatment has recently been applied to minimize cotton and wool shrinkage and to improve its dyeing properties Riva (1991), and Riva *et al.* (1993). As chemicals are not generally biologically degradable, enzymes have come as a suitable way to take care of environment, Mehra *et al.* (1992).

Recent results indicated that enzymes may be used effectively in the cleaning procedures of cotton. Enzymes and chelating agents in cotton pretreatment were studied by Emilia *et al.* (2001). Effect of enzymatic treatment on the dyeing of cotton and wool fibers with natural dyes was studied by Tsatsaroni *et al.* (1995).

Products of cellulase enzyme for the textile industry are mainly classified in two groups: neutral and acid cellulase based on the pH range in which they are most effective. Acid cellulase is a type of enzyme that works effectively at the pH range of 4.5 -5.5. It is highly active on cotton fibers as compared to the neutral cellulase.

The aim of this work was to study the effect of enzymatic treatment of cotton on their dyeing properties. The results are compared with those obtained with the same dye after treatment of the fabrics carried by simultaneous technique (sample-dye-mordant) according to saleh *et al.* (2003).

MATERIALS AND METHODS

Materials

Bleached cotton fabrics of the Egyptian cotton varieties namely Giza 89 and Giza 83 were purchased from Misr-El-Mehala for weaving and spinning company during the season 2002-2003, and used throughout this study. The specification of both Giza 89 and 83 fibers are given in Table 1.

Table 1. Fiber quality measurements of the 2002-2003 Giza 89, and 83

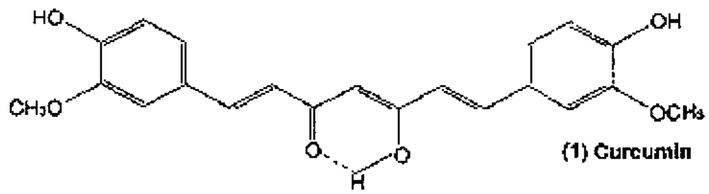
Measurements →	*Value of color attribute*			*Fiber length*		*Fiber strength T and elongation E*		*Fiber fineness F, and maturity Mat %*		
Variety ↓	color	Rd%	+b	ML mm	U.I	T g/tex	E %	M. R	Mat %	F
Giza 89	W	72.2	8.1	27.6	86.2	39.0	7.2	4.3	97	155
Giza 83	C	62.1	11.4	25.1	84.5	33.8	7.6	4.0	90	156

Where Rd% reflectance percent, +b yellowness, U.I uniformity index, and M.R micronaire reading,

Dye

C.I. Natural yellow 3 or Curcumine, $C_{21}H_{20}O_6$, m . p 184-185°C, extracted from the fresh or dried rhizomes of Turmeric-Curcuma Tinctoria, Longa, Rotunda and

Viridiflor. Curcumin is the only natural yellow dye belonging to diferuloyl-methane group. It is a diferuloyl-methane having the following formula:



(1) Curcumin

24

100g of curcumin contains the dye components were boiled in one liter of NaOH (2%) and concentrated to a volume of 500cc. The extracted liquor was used as the foundation of the dye.

Enzyme treatment

Multifunctional acid cellulase enzyme formulation namely Cellusoft^{R} L (Novo Nor disk) was used for the samples pretreatment. The pretreatment was carried out under the following conditions: pH 4.4 (by acetic acid), enzyme dosage 0.7, time 30 min. and temperature 55°C according to Blanch (1992), and Blanchard *et al.* (1997). After treatment, the enzyme inactivated by raising the pH to 9-10 (with soda ash), or by raising the temperature to 75°C for 10 min, followed by thoroughly washing and tumble drying according to Boylston *et al.* (1995).

Mordants

Aqueous solutions containing 2gl/l iron (II) sulphate heptahydrate, alum, potassium dichromate and copper sulphate were used as mordants.

Dyeing treatments

1-Simultaneous (sample-dye-mordant) without enzymatic treatment

10ml of aqueous solutions containing 2gl/l mordant was added to the bath containing the prepared dye. The wetted-out fabric was then added at a liquor ratio of 1:30. The whole is brought slowly to nearly boiling and the fabric was gently moved with a stirring rod from time to time. Simmering continues until either all the dye is taken up or the required shade is reached. The fabric can either be left to cool in the liquid or removed and rinsed in very hot water, followed by subsequent rinses of progressively cool water. The fabric was then gently washed with soap or mild detergent until no further color is lost.

2- Simultaneous (sample-dye-mordant) after enzymatic treatment

All samples after enzymatic treatment were allowed to stand overnight according to Storey (1985), and Tsatsaroni et al. (1994). Then in a 5% ammonia solution for 30 min rinsed and squeezed to inactivate the enzyme and applied in the dye bath containing the mordant at a liquor ratio 1:30. The temperature was raised to 90ºC over 30 min and maintained at this level for 1h. Sodium chloride (10%) or a few drops of 40% acetic acid solution were added to the dyeing liquors, then rinsed with water and dried.

Fastness properties

Dyeing and wash fastness tests were carried out in a Rota dyes apparatus (John Jeffrey's Ltd.). Spectrophotometeric measurements were recorded in a Perkin Elmer double beam Model Lambda 35 equipped with integrating sphere. An air-cooled Hahau Sunset apparatus (Heraeus) with a xenon lamp was used for the light fastness tests. Color changes during the light fastness tests were evaluated in a Vervide color assessment cabinet (Leslie Hubble) with an artificial daylight (D65) lamp.

Determination of the dye adsorbed on the fabric

This is done by extracting the pigment of the dyed fabric with pyridine (25% v/v in water) and measuring the absorbance at 409 nm (λmax of the prepared dye) according to Tsatsaroni *et al.* (1995).

Color values

The dyed fabrics were scanned with D65 illuminant source in the wavelength range 400-700nm using the standard white tile as the reference for all color measurements. The CIE color components L, a, b,c and h were recorded and used throughout this study. The color strength expressed as k/s was also recorded according to Kubilka-Munk equation (1931):

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

Where K is the absorption coefficient, S is the scattering coefficient, and R is the reflectance of dyed samples at the wavelength of the maximum absorption.

Wash fastness

Washing fastness was measured according to ISO 105-C01:1989 (E). Two single fiber adjacent fabrics complying with the relevant sections of F01 to F08 of ISO 105-F- 1989, one of the adjacent fabrics of cotton and the second of wool.

Light fastness

Fastness of light was measured according to the ISO 105:1997 using standard wool serge as reference in all tests.

Perspiration fastness

Fastness of perspiration was measured according to the ISO 105-E04:1994 using 1-Histidine Monohydrochloride (0.5g/l): NaCl (5g/l) and Na_2HPO_4 (2.2-2.5g/l) at pH 5.5 and 8 for acidic and alkaline perspirations respectively.

RESULTS AND DISCUSSION

1- Dyeing Mechanism

The dyeing of cotton fabrics with curcumin dye takes place via a weak complex formation between the dye and the metal ions from the mordant producing a charged complex. On soaking the fabric into the solution, a reaction between the hydroxyl groups in the fabric surface and the charged complex occurred

$$D+M \rightarrow [D\ M]^{+} \quad (1)$$

$$cellOH + [DM]^{+} \rightarrow cellOH{-}{-}{-}DM \quad (2)$$

D=Dye, M= mordant, $[DM]^{+}$ is the intermediate complex. (CellOH) is cellulose molecule and (---) is a coordinate bond between the charged sites or by using the lone pair of electrons on the functional groups in the molecular structure of the reactants.

2- Evaluation of Color components

The results in table 2 reflect that the color strength values (K/S) of Giza 89, and Giza 83 fabric samples dyed without the enzymatic treatment were higher than those dyed with unmordanted samples. These can be attributed to the important role that the mordant forming the intermediate complex which can react chemically or physically with the cellulosic fiber.

Table 2. Color measurements of dyed Giza 83, and Giza 89 cotton fabrics respectively by simultaneous mordanting technique without enzymatic treatment

Mordant	K/S		L*		a*		b*		c*		h*	
	G83	G89	G83	G89	G83	G89	G83	G89	G83	G89	G83	G89
Nil	0.28	0.3	87.7	82.7	2.1	0.8	18.1	20.5	18.2	20.5	96.6	92.2
Cu++	1.10	1.3	84.6	74.7	1.6	0.6	22.9	24.6	23	24.7	85.8	86
Fe++	0.6	0.66	85.7	80.7	1.5	0.54	22.8	26.6	22.8	26.7	88.7	85.5
Cr+++	0.24	0.53	88.8	81.6	0.8	0.21	8.5	11.5	8.6	11.7	84.7	79.8
Alum	0.43	0.46	82.7	80.9	1.4	0.51	18.3	30.9	38.4	40.9	93.1	91.7

Where K/S (color strength), L*(lightness-darkness), a*(red-green component) b*(blue-yellow component), c* (chroma), h*(hue)

The results obtained revealed that the values of L* (lightness-darkness), and a (red-green component) were higher for Giza 83 than that of Giza 89 due to uptake of the dye on the fabric samples of Giza 83 was lower than that of Giza 89.

The results showed that Cu^{2+} is more effective as a mordant more than that of Fe^{2+} ,Cr^{3+} , and Alum for both Giza 83, and Giza 89 to give the maximum dye absorbance (low L* value) for simultaneous mordanting technique without enzymatic pretreatment.

Table 3. Color measurements of dyed Giza 83, and Giza 89 cotton fabrics respectively by simultaneous mordanting technique after enzymatic treatment

Mordant	K/S		L*		a*		b*		c*		h*	
	G83	G89	G83	G89	G83	G89	G83	G89	G83	G89	G83	G89
Nil	0.35	0.39	85.9	85.1	1.0	.26	19.1	19.8	19.2	19.8	93.1	89.3
Cu	1.9	2.4	71	71.2	6.7	8.8	25.6	30.1	26.6	31.3	85.9	83.7
Fe	.66	.65	81.9	82.1	1.9	3 .3	27.8	28.1	27.9	28.3	86.2	83.3
Cr	2.6	3.3	71	72.7	2.1	1.2	38.4	41	38.4	41	93.1	91.8
Alum	0.17	0.17	78.4	79.2	2.1	1.8	23.3	41.3	39.4	41.4	99	97

As shown in Table 2.

Table 3 shows that enzyme treatments resulted in an increase in dye uptake (K/S values) for Giza 89, and Giza 83 compared with the simultaneous mordanting technique without that treatment samples. Giza 89 showed an increase in the color strength values than that of Giza 83 in all cases of treatment due to its higher hydroxyl groups on the fabric surface causing increase in the charged sites by using the lone pair of electrons on the functional groups between the molecular structures of the reactants. Cu^{2+}, and Cr^{3+}could be used as mordant more than that of Fe^{2+} , and Alum for both Giza 83, and Giza 89 to give the maximum dye absorbance (low L* value) for enzymatic treatment technique.

Values of b* (blue-yellow component), c* (Chroma, relating to the color fastness of the sample when compared to the neutral (achromatic) grays which occupies the central spine of the volume), and h (Hue can be classified into warm – yellow, orange, scarlet, crimson and their derivatives and cool- blue, greens, purples, and their derivatives Kamel (1972), were higher for Giza 89 than that of Giza 83 in all cases of treatment due to its higher dye adsorbed on the fabric sample.

The data obtained from tables 2 and 3 show that the enzymatic treatment improves the dyeing up take for both Giza 89, and Giza 83.

3- Dye exhaustion

Table 4. Dye exhaustion of Gaza 89, and Gaza 83 cotton fabrics respectively using simultaneous technique without[a] and after[b] enzymatic treatment respectively

Treatment techniques	Dye(g) per 100g fabric	Giza89	Giza83
		Exhaustion %	Exhaustion%
Treatment a(without enzyme)	0.063	3.15	2.26
	0.087	4.35	3.89
	1.458	72.9	66.8
	1.818	90.9	82.6
Treatment b(after enzyme)	0.063	8.9	6.23
	0.087	26.9	22.5
	1.458	89.2	82.7
	1.818	93.5	91.2

As can be observed from table 4, the amount of dye adsorbed on the fabric samples treated with enzyme then dyed were higher than for the fabric samples untreated with enzyme. The results obtained revealed that the amount of dye adsorbed on the Giza 89 fabric is higher than for Giza 83 for both treatments. Enzymatic treatment resulted in an improvement in dye uptake in all cases reported, which could be attributed to enhanced shrink-resistance properties of the fabric.

4- Evaluation of fastness properties

4-1 Light fastness

Light fastness of dyed substrates depends on various factors: nature of the dye, stability constant of the complex between the dye and the element, reactive group in the dye molecule, and nature of the dye-fiber bond. Tables 5 and 6 show different rating according to the mordant used. The fastness ratings was higher when using Fe^{3+} as a mordant for both pretreated enzymatic and untreated samples for Giza 83, and Giza 89 respectively.

Various fastness data of all the dyed Giza 83 and 89 fabric cotton samples respectively are given in Tables 5, and 6 including those pretreated with enzyme and that untreated.

Table 5. Data on various fastness properties of Giza 83 cottonfabric using simultaneous technique without* and after** enzymatic treatment respectively

M	L.F	W. F			P. F					
					Alk.			Acidic		
		Alt	C	W	Alt	C	W	Alt	C	W
Control	1	3	5	5	2/3	5	5	3	5	4/5
Cu*	1	3	5	4/5	¾	4/4	4	3	5	5
Fe*	3/4	4	4/5	4	4/5	5	5	4	4/5	5
Cr*	1	3/4	5	4/5	2	5	4/5	3	4/5	5
Alum*	1	3	4/5	4/5	2/3	5	4/5	3	4/5	4/5
Cu**	1	4	4/5	4/5	4	4	4	4	4/5	4/5
Fe**	3/4	4/5	4/5	4/5	5	4/5	4	4	4/5	4/5
Cr**	1	4/5	4	4/5	4	3/4	4	4	4/5	5
Alum**	1	4	4	4	4/5	4/5	4/5	4	4/5	4

Dye concentration (3% of Fabric weight), mordant concentration 2g /l

L.F Light fastness, W.F Washing fastness, P.F Perspiration fastness, Alternative, C Cotton, W Wool, * simultaneous dyeing without enzymatic treatment ** simultaneous dyeing after enzymatic treatment

4-2 Wash fastness

Generally Circumcise dye exhibits good fastness properties, due to the mineral matter (mordant) processes which might form chelates with the dye inside the substrate. The wash fastness ratings in Tables 5 and 6, show that the pretreatment enzymatic technique did not affect wash fastness properties and there is difference between the pretreated enzymatic and untreated enzymatic samples.

4- 3 Perspiration fastness

The fastness ratings in tables 5 and 6 show that the enzymatic treatment technique did not affect wash fastness properties and there is difference between the pretreated enzymatic and untreated enzymatic samples. With respect to color change for the Giza 83, and Giza 89, dyed fabric samples. However, staining of the adjacent fabrics is to some extent high in both methods. Fastness to alkaline perspiration showed similar results

Table 6. Fastness properties of Giza 89 cotton fabric using simultaneous technique without* and after** enzymatic treatment respectively

Mordant	L.F	W. F			P. F					
					Alk.			Acidic		
		Alt	C	W	Alt	C	W	Alt	C	W
Un mordanted	1	3	5	5	2/3	5	4/5	3	5	4/5
Cu*	2/3	4	4	4	4	5	4/5	4	4/5	4
Fe*	5	4/5	4	5	4	4/5	4/5	4/5	5	5
Cr*	¾	5	5	5	4/5	4/5	5	3	4	4
Alum*	1	3/4	4	3/4	3/4	5	5	4	5	4
Cu**	1	3	4/5	4/5	4/5	4/5	4/5	4	5	4/5
Fe**	4/5	4/5	5	4	4/5	5	4/5	4/5	5	5
Cr**	3	4/5	5	4/5	5	4/5	5	3	4	4
Alum**	1	3	4/5	3/4	3/4	5	5	4	5	4

As shown in table 5

CONCLUSION

The present study revealed that Curcumin can provide a less toxic, non pollutant, very brilliant, and reliable natural dye for dyeing a range of colors depending on the mordant type. Enzymatic pretreatments resulted in an increase in dye uptake in all cases compared with the untreated or the simultaneous mordanting dyeing technique without pretreated enzymatic samples, and did not affect fastness properties. Giza 89 pretreated enzymatic samples were higher than those for Giza 83. The color strength (K/S) values, and fastness properties indicating the higher dyeing response of Giza 89 than Giza 83 in all cases of treatments.

REFERENCES

1. Blanch R. M., 1997. ATA jr 8 (5), 68.
2. Bolystone E. K., and J. J. Herbert. 1995. Text. Res. J 65, 429.
3. Das, S. colorage .1992. Vol. XXX1X, No.9, Sept., p. 52.
4. Emilia C, L. Anita, S George, and I. Rusznak. 2001. J. Biotechnology 89 271-279.
5. Grierson S., D. G. Duff. and R. S. Sinclair 1985. J. Soc. Dyers Colourists, 101 220.
6. Gupta V. K., R. A. Sachon, V. P. Singh, and V. P Sharma 1998. The indian Textile Journal, Vol. 108,No. 4, 16 -19.
7. Kamel J., and E. Hoeppner. 1972. "Natural Dyes Plants and Processes" USA SBN chpt. (Trade cloth).
8. Katyaynini V. K., M. Jacob. 1998. The Indian Textile Journal, Vol. 108, No. 4, p. 86.
9. King J., and R. A. Stewart 1970. in Dyes and Dyeing. Griffin and George Ltd.
10. Kubilka p., and F. Munk. 1931. Techn. Z. Physik 12, 593.
11. Mahate G., Sakshi, and R. K. Sunanda 2003. Indian Journal of Fibre& Textile Rresearch, Vol. 28, No 1, Mar, pp. 86-89.
12. Mehra R. H., and A.R. Mehra 1992. Spcial Issue, Colourage, Vol. xxxix, July.
13. Nishida K., and K. Kobayashi. 1992. American Dyestuff Reporter, Vol. 81, No. 7, p. 44.
14. Riva a., J. C., and Sanchez, R. Prieto 1993. J. Soc. Dyers Colourists, 109 210.
15. Saleh S. M., and M. A. El-Gamal 2003. Egypt. J. Appl. Sci.18 (12B), 559-568
16. Tsatsaroni E. G., and I. C. Eleftheriadis. 1994. J. Soc. Dyers Colourists, 110, 313.
17. Tsatsaroni E. G., and M. Liakopoulou- Kyriakides 1995. Dyes and Pigments Vol. 3, 203-209.

تحسين خواص الصبـــاغة لبعض أصناف القطن المصري باستخدام صبغة الكركم الطبيعية مع المعالجة بالأنزيمات

صلاح منصور صالح [١] ، عزة عبد العزيز محمود[١] ، خالد البدرى[٢]

١. مركز البحوث الزراعية- معهد بحوث القطن –-الجيزة – مصر.
٢. جامعة حلوان – كلية التربية – – شعبة النسيج

يهدف هذا البحث إلي دراسة تأثير المعالجة بالأنزيم (سيلولاز) على صفات الأقطان المصبوغة بالصبغات الطبيعية (الكركم) مقارنة مع المنسوجات الغير معالجة وتم استخدام نسيج صنفين هما جيزة ٨٩ وجيزة٨٣ من موسم ٢٠٠٣ من شركة مصر للغزل والنسيج بالمحلة الكبرى، وكانت النتائج كالتالي:

١- قوة امتصاص الصبغة:

وجد أن جيزة ٨٩ أعطت استجابة أعلى من جيزة ٨٣ مع استخدام المواد المثبتة وكذلك بعد المعالجة بالأنزيم وفى مكونات اللون: وجد أن جيزة ٨٣ سجل استجابة أعلى من٨٩.

٢- المواد المثبتة:

وجد أن النحاس أعطى أعلى نتائج عن الحديد والكروميوم والشبة مع جيزة ٨٣ عن جيزة٨٩ في المعالجة بدون إنزيم أما عند المعالجة بالأنزيم وجد أن النحاس والكروميوم أحسن من الحديد والشبة.

٣- درجة نفاذية الصبغة للأنسجة:

وجد أن امتصاص الصبغة سواء المعالجة أو الغير معالجة كان في صنف جيزة٨٩ أكثر استجابة من صنف جيزة٨٣.

٤- اختبارات الثبات للون (الضوء والغسيل والعرق الحامضى والقلوى) :

وجد أنه لا يوجد فروق نوعية فى أختبارات للثبات للعرق سواء العينات المعالجة بالأنزيم أو الغير معالجة . كـــــما وجد أن أستجابة صنف جيزة ٨٣ لأختبارات العرق أعلى منها فى صنف جيزة ٨٩ .

وكنتيجة نهائية لهذا البحث فقد وجد أن المعالجة بأنزيم السليولاز أدى إلى زيادة امتصـــاص الصبغة الطبيعية وثباتها لكل الصفات التي تم قياسها لكل من جيزة ٨٩ و جيزة ٨٣ و قــد وجــد أن استجابة صنف جيزة ٨٩ أعلي من صنف جيزة٨٣.