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Natural Dyeing of Cotton, Wool and Viscose Fabrics with Sodium Copper Chlorophyllin

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Abstract

This study investigates the optimal dyeing concentrations of cotton, wool, and viscose fabrics with sodium copper chlorophyllin (SCC). Using SSC above the optimal concentration increases the amount of SCC in the wastewater, which is a disadvantage. Pre-finished 100% cotton, wool and viscose 1 x 1 rib knitted fabrics were dyed with varying SCC concentrations. Optimal dyeing concentrations were determined using UV-Vis spectrophotometry, showing dye absorption decreasing as SCC concentration increases. Equilibrium was reached at approximately 7 ml/L for cotton and viscose, and 3 ml/L for wool. Color measurement values and K/S values of the dyed samples were obtained according to the CIELab color system. The K/S values of dyed cotton, viscose, and wool fabrics rose as the concentration of SCC increased, reaching a peak at 9 ml/L for viscose and 12 ml/L for cotton and wool. Rubbing and perspiration fastness were rated good to excellent.

Keywords: Sodium copper chlorophyllin, Natural dyeing, UV-visible spectrophotometer, Color fastness

Pamuk, Yün ve Viskon Kumaşların Sodyum Bakır Klorofil ile Doğal Boyanması

Öz

Bu çalışmanın amacı pamuk, yün ve viskon kumaşların sodyum bakır klorofil (SCC) ile optimum boyama konsatrasyonlarının araştırılmasıdır. Ön terbiyesi yapılmış %100 pamuk, yün ve viskon 1 x 1 ribana örme kumaşlar farklı SCC konsantrasyonlarıyla boyanmıştır. Optimal boyama konsantrasyonlarını belirlemek için UV-Vis spektrofotometre kullanılarak SCC konsantrasyonunun boya çekimine etkisi incelenmiştir. SSC'nin optimal konsantasyondan fazla kullanılması bir dezavantaj olarak atık suyun SCC miktarını artırır. SCC konsantrasyonunun artmasıyla boya çekiminin azaldığı ve pamuk ve viskon için yaklaşık 7 ml/L, yün için ise 3 ml/L civarında dengeye ulaştığı görülmüştür. Numunelerin K/S değerleri ve renk ölçümleri CIELab renk sistemi kullanılarak ölçülmüştür. K/S değerleri yün kumaşlarda 12 ml/L'ye kadar, pamuk ve viskon kumaşlarda ise 9 ml/L'ye kadar artmıştır. Sürtünme ve terlemeye karşı renk haslığı değerlendirilmiş olup boyalı kumaşların haslık değerleri iyi ile mükemmel arasındadır.

Anahtar Kelimeler: Sodyum bakır klorofil, Doğal boyama, UV-vis spektrofotometre, Renk haslık

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1. INTRODUCTION

Compared with synthetic dyes, natural dyes are known to be eco-friendly, biodegradable, less toxic and less allergenic, as they are derived from natural resources classified as plant, animal, mineral and microbial dyes [1-3]. However the use of natural dyes for textile dyeing has been dramatically cut down since the discovery of synthetic dyes in 19th century as synthetic dyes can produce a wide range of bright shades, with fairly good color fastness, dyeing reproducibility and low cost [4,5]. In recent years there is a reviving interest in the dyeing of the textile materials with natural dyes due to the increasing popularity of concept of eco-friendly textiles based on naturally sustainable goods [6-8]. There are some studies on the coloring of different types of fabrics using natural dyestuffs. Bahtiyari et. al. pigmented cotton fabrics using extracts from pomegranate peel, nutshell, orange tree leaves, alkanet roots, and dyer's chamomile via the pigment printing method [9]. In 2015, Davulcu developed polypropylene (PP) fibers dyed with natural dyes by adding hyperbranched polymer (HBP) using melt spinning. They used walnut shell, oak gall, dyer's chamomile, sage tea, and cehri as natural dye sources [10]. Tutak et. al. studied on usability pomegranate peel natural dye solution on wool fabrics [11].

In several studies textile materials are colored in variable tones of mainly yellow, red and blue colors by using the natural dyestuffs that obtained from different types of plants and insects. To obtain a green color, textile materials are usually dyed blue with indigo first and then, the mordanted material is dyed a second time with a yellow plant [12]. Natural green extracts consisting of chlorophyll molecules, which are abundant in all green plants, algae and cyanobacteria, are also used as fabric dyeing colorants [13,14]. Despite attempts to use chlorophyll as a natural green dye, it often yields a dull yellow or brownish color due to the loss of magnesium ions from the chlorophyll molecules during extraction and processing. As a derivative of natural chlorophyll, sodium copper chlorophyllin (SCC) is a natural edible pigment and a water-soluble bright green colorant with good stability [15,16].

There are several studies which reported SCC application in dyeing textiles. Park and Park dyed cotton fabrics with natural chlorophyll derivatives (chlorophyllin, Chlin) with and without chitosan treatment to enhance textile coloration and antimicrobial activity [17]. Hou et. al., dyed silk into a bright green shade with SCC and were investigated kinetics and thermodynamics of SCC adsorption on silk in dye bath with and without sodium chloride at different temperatures [15]. Xiu-Liang et. al., were investigated the adsorption kinetics of SCC on silk fabrics and the dyeing mechanism in order to improve the dye up-taking process control and provide a theoretical guidance for dyeing process optimization of SCC on silk fabrics [18]. Zhang and Tao, studied the eco-friendly dyeing of silk fabric with SCC using different mordanting methods as well as a non-mordanting method [19]. Zhao et. al., employed ethylenediaminetetraacetic acid disodium salt and sodium citrate as additives to treat dyed cotton fabrics and studied on photofading of SCC and gardenia yellow [20]. Liu et. al., firstly used SCC as an environmentally friendly dyestuff to dye the wool fibers, then the dyed woolfibers was phosphorylated with phosphoric acid to obtain flame retardant woolfibers [16].

This study aims to explore the potential usability of sodium copper chlorophyll (SCC) as an alternative for the textile dye industry, which is actively seeking sustainable and renewable resources for textile production. The investigation focuses on its effectiveness in dyeing various types of fibers. Fabrics were pre-treated and then dyed with different concentrations of SCC without mordanting. The effect of SCC concentration on dye exhaustion was determined by spectrophotometric method in order to investigate the optimal dyeing concentrations. Color measurements and color fastness of dyed samples against rubbing and perspiration were also evaluated.

2. MATERIAL AND METHOD

2.1. Material

100% carded cotton ring spun yarn with Ne 20/1 yarn count, 100% carded viscose ring spun yarn with Ne 20/1 yarn count and 100% worsted wool yarn with Nm 5/1 yarn count were used. The yarns were knitted with flat knitting machines of 10 gauge thus 100% cotton, 100% wool and 100% viscose 1 x 1 rib fabrics were produced. Tito Co. Ltd. supplied the commercial sodium copper chlorophyll dye liquid.

2.2. Method

2.2.1. Pre-Finishing Process

The cotton fabric underwent bleaching in a laboratory-type machine at 95°C for 4 hours. The bleaching solution consisted of 6% H_2O_2, 4g/L (46°Be) NaOH, 0,5 g/L oil remover soap, 0,5 g/L anticreasing agent, 0,8 g/L ion trapping agent, 0,5 g/L wetting agent, and 0,8 g/L peroxide stabilizer. The liquor ratio was chosen to be 30:1. The fabric was rinsed and neutralized. After bleaching anti-peroxide process was applied. The pre-finishing treatment for wool and viscose fabrics was performed at 40°C for 60 minutes using a solution containing 5 g/L of oil remover soap in a laboratory dyeing machine, with a liquor-to-goods ratio of 30:1.

2.2.2. Dyeing Process

The dyeing of cotton, viscose and wool fabrics were carried out in exhaust dyeing method with a laboratory-type dyeing machine at 60°C for 60 minutes at a liquor-to-goods ratio of 10:1. Dyeing was done with different concentrations of SCC as given in Table 1. The fabric underwent two cold-water washes and was then dried under room temperature conditions.

Table 1. SCC concentrations

Sample code			SCC Conc. (ml/L)
Cotton	**Viscose**	**Wool**	
C1	V1	W1	1
C3	V3	W3	3
C5	V5	W5	5
C7	V7	W7	7
C9	V9	W9	9
C12	V12	W12	12

2.2.3. Dye Exhaustion

The method employed with a Shimadzu UV-1800 UV/VIS spectrophotometer (Eksoy Company, Turkey) relies on an absorbance-concentration curve generated using dye solutions of known concentrations. This curve is then used to determine the concentration of a dye solution with an unknown concentration. Maximum absorption wavelength (λmax) was identified as 626.5 nm. Calibration curve is given in Figure 1. As seen in Figure 1, it is observed that the absorption values of the dye solutions increase linearly with the increase in dye concentrations.

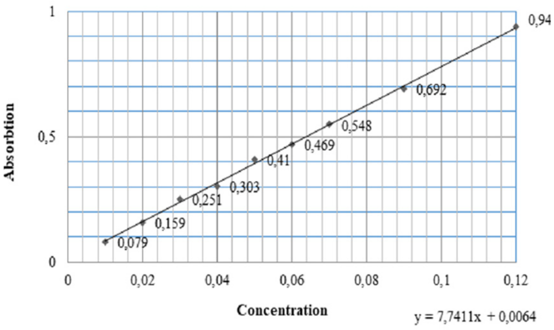

Figure 1. Calibration curve

The exhausted and unexhausted amounts of SCC were determined by calculating the difference in dye concentration in the dyebath before and after dyeing. The percentile of exhausted dye was calculated with Equation (1) by the initial and ultimate concentrations of the dye bath;

Dye Exhaustion (%)= [((Cb- Ca)/Cb) x 100] (1)

Cb represents the initial concentration of the dye bath, while Ca denotes the final concentration of the dye bath [21].

2.2.4. Color Measurements & Color Strength

K/S values and the CIELab color values of the dyed samples were measured by Datacolor 850 spectrophotometer (Eksoy Company, Turkey) using an illuminant D65 and 10° standard observer. Total colour difference ΔE [equation (1)] is calculated to determine the color difference between reference sample and other samples [14].

$$\Delta E = ((\Delta a^2) + (\Delta b^2) + (\Delta L^2))^{1/2} \quad (2)$$

$$K/S = (1-R)^2/2R \quad (3)$$

where, R=relectance, K=absorption coefficient and S=scattering coefficient of dye [22].

2.2.5. Fastness Tests

Color fastness tests to rubbing and perspiration of dyed fabric samples were evaluated according to ISO 105-X12 and ISO 105-E04, respectively.

3. RESULTS AND DISCUSSION

3.1. Dye Exhaustion

To investigate the effects of SCC concentration on the dye exhaustion and K/S values of dyed samples, cotton, viscose and woll fabrics were dyed with different concentrations of SCC as indicated in the experimental section. The results obtained are represented in Table 2.

As seen in Table 2, with increase in the SCC concentration, the amount of exhausted SCC increases. It is expected that the exhausted amount of SCC should increase with increase in the amount of SCC particles available for sorption in the dyeing bath. But percentile value of the dye exhaustion decreases with increase in the SCC concentration, till it reaches the optimum concentration. This situation was considered as the saturation of the fibers in terms of dye uptake in a certain time with the increasing amount of dye molecules in the dyeing bath and less space should left for the remaining dwangye molecules to diffuse into the inner area of fibers. Table 2 indicates that the percentile value of dye exhaustion decreased with an increase in the SCC concentration and reached equilibrium when the SCC concentration is about 7 ml/L for cotton, 7 ml/L for viscose and 3 ml/L for wool fabrics. More increase in the SCC concentration should result the increase in the amount of SCC that should remain in the effluent. That means the load of unexhausted dye remaining in the effluent can be reduced using a maximum amount of 7 ml/L concentration of SCC while dyeing cotton, 7 ml/L concentration of SCC while dyeing viscose and 3 ml/L concentration of SCC while dyeing wool.

Table 2. Effect of concentration of SCC on dye exhaustion and K/S of the dyed cotton, viscose and wool fabrics

Sample code		SCC Conc. (ml/L)	Dye Exhaustion (%)	Dye Exhaustion (mL)	Unexhausted Dye (mL)	K/S
Cotton	C1	1	50,00	0,5	0,5	0,35
	C3	3	30,00	0,9	2,1	0,67
	C5	5	24,00	1,2	3,8	0,84
	C7	7	14,29	1,0	6,0	
	C9	9	16,67	1,5	7,5	0,93
	C12	12	15,83	1,9	10,1	1,11
Viscose	V1	1	40,00	0,4	0,6	0,45
	V3	3	26,66	0,8	0,2	0,79
	V5	5	20,00	1,0	4,0	0,91
	V7	7	15,71	1,1	5,9	
	V9	9	17,77	1,6	7,4	0,96
	V12	12	15,00	1,8	10,2	0,90
Wool	W1	1	50,00	0,5	0,5	0,67
	W3	3	40,00	1,2	1,8	
	W5	5	40,00	2,0	3,0	1,16
	W7	7	37,14	2,6	4,4	1,26
	W9	9	40,00	3,6	5,4	1,31
	W12	12	41,66	5,0	7,0	1,37

3.2. Color Measurement & Color Strength

To investigate the effects of SCC concentration on the color parameter and color difference, the

CIELab system was used. C7, V7 and W3 were taken as reference samples in the color measurement results of the cotton, viscose and wool fabrics as the percentile dye exhaustion reached equilibrium when the SCC concentration is about 7 ml/L for cotton, 7 ml/L for viscose and 3 ml/L for wool fabrics. As seen in Table 3, L* refers to the lightness-darkness values from 100 to 0, representing white to black. Both the a* value and b* value denote the red-green values that run from negative (green) to positive (red) and the yellow-blue values that run from negative (blue) to positive (yellow) respectively. Characteristic value c* represents the chroma or purity of the color. The dyed cotton and viscose fabrics showed L* values close to each other, while wool fabrics showed lower L* values causing darkness compared to cotton and wool fabrics. Moreover an increase in the SCC concentration of dyeing cotton, viscose and wool fabrics caused similar trends in terms of L*, a*, b*, C* and ho values. In Table 3, the L* value increased when the SCC concentration changed from 1 ml/L to 3 ml/L, but decreased with more increase in the SCC concentration. When calculating the color difference, ΔE, from the reference samples using the L*, a* and b* value, the color difference was higher while dyeing with lower concentrations of SCC than dyeing with higher concentrations of SCC

The K/S values of cotton, viscose and wool fabrics were in a range of 0.35 – 1.11, 0.45 – 0.90 and 0.67 – 1.37 respectively. This indicates that viscose fabric was dyed a deeper color than cotton and wool fabric was dyed a deeper color than viscose. It is clear from Table 3 that the value of K/S depends on the concentration of the SCC used as well as the origin of the fibre. The results revealed that K/S increases with the increased percentage of SCC concentration. This may possibly be due to the increasing amount of dye molecules in the dyeing bath that should be exhausted. The maximum K/S values were obtained at 9 ml/L concentration of SCC while dyeing viscose and 12 ml/L concentration of SCC while dyeing cotton and wool.

Table 3. Effect of concentration of SCC on dye exhaustion of the dyed cotton, viscose and wool fabrics

Sample code		L*	a*	b*	ΔE	K/S
Cotton	**C1**	85,22	-5,41	8,49	4,67	0,35
	C3	80,11	-7,45	9,34	2,11	0,67
	C5	78,00	-8,27	9,32	1,06	0,84
	C7	76,52	-9,33	9,46		
	C9	76,75	-8,23	9,26	0,93	0,93
	C12	75,41	-9,76	9,19	0,64	1,11
Viscose	**V1**	83,69	-4,85	9,19	3,39	0,45
	V3	79,05	-6,59	9,57	1,19	0,79
	V5	77,53	-6,77	9,01	0,42	0,91
	V7	76,49	-6,88	8,92		
	V9	76,87	-7,03	8,51	0,45	0,96
	V12	77,41	-6,81	8,29	0,65	0,90
Wool	**W1**	76,39	-6,34	14,24	2,84	0,67
	W3	72,25	-8,90	13,65		
	W5	70,43	-9,75	13,33	1,07	1,16
	W7	69,55	-10,32	12,93	1,77	1,26
	W9	68,94	-10,22	12,78	1,92	1,31
	W12	68,29	-9,64	12,74	1,84	1,37

3.3. Color Fastness Test Results

Table 4, 5 and 6 outlines the color fastness results to rubbing (dry and wet) and perspiration (acidic and alkaline) of cotton, wool and viscose fabrics dyed with different concentrations of SCC without mordanting. As expected the grades of color fastness to rubbing for the dry test are higher than the grades for the wet test. Color fastness to rubbing for wet test of wool samples ranged 4/5, which were mostly higher than than the cotton and viscose fabrics (ranged grade from 3/4 to grade 4/5). All of the fabrics showed excellent grades of color fastness to rubbing for the dry test (grade 5). In terms of the gray scale assessment, the grades of staining of perspiration fastness on all the dyed cotton, viscose and wool fabrics revealed good fastness with a grade range between 4/5 and 5 both for acidic and alkaline medium, while the grades of color change ranged between 3/4 to 5. From the color fastness test results it can be concluded that SCC dye exhibits good fastness to rubbing and objective fastness to perspiration.

Table 4. Color fastness to perspiration (acidic)

Sample code	Acidic						Change
	Color staining*						
	1	2	3	4	5	6	
C1	5	4/5	5	5	5	5	4/5
C3	5	4/5	5	5	5	5	4/5
C5	5	4/5	5	5	5	5	4/5
C7	5	4/5	5	5	5	5	5
C9	5	4/5	5	5	5	5	5
C12	5	4/5	5	5	5	5	5
V1	5	5	5	5	5	5	5
V3	5	5	5	5	5	5	5
V5	5	5	5	5	5	5	4
V7	5	4/5	4/5	4/5	5	4/5	4
V9	5	4/5	4/5	4/5	4/5	4/5	3/4
V12	5	4/5	4/5	4/5	4/5	4/5	4
W1	5	5	5	5	5	5	4/5
W3	5	5	5	5	5	5	4/5
W5	5	5	5	5	5	5	4/5
W7	5	5	5	5	5	5	4/5
W9	5	5	5	5	5	5	5
W12	5	4/5	4/5	4/5	5	4/5	5

*Multifiber (1-Acetate, 2-Cotton, 3-Polyamide, 4-Polyester, 5-Acrylic, 6-Wool)

Table 5. Color fastness to perspiration (alkaline)

Sample code	Alkaline						Change
	Color staining*						
	1	2	3	4	5	6	
C1	5	5	5	5	5	5	4/5
C3	5	5	5	5	4/5	5	4/5
C5	5	5	5	5	4/5	5	4/5
C7	5	5	5	5	4/5	5	4/5
C9	5	5	4/5	4/5	4/5	4/5	4/5
C12	4/5	4/5	4/5	4/5	4/5	4	4/5
V1	5	5	5	5	5	5	4/5
V3	5	5	5	5	5	5	4/5
V5	5	4/5	4/5	4/5	5	5	3/4
V7	5	4/5	4/5	4/5	5	4/5	3/4
V9	5	4/5	4/5	4/5	5	4/5	3/4
V12	5	4/5	4/5	4/5	5	4/5	4
W1	5	5	5	5	5	5	4/5
W3	5	4/5	4/5	4/5	4/5	5	4/5
W5	5	4/5	4/5	4/5	4/5	5	4/5
W7	5	4/5	4/5	4/5	4/5	5	4/5
W9	5	5	4/5	4/5	4/5	5	5
W12	5	5	5	5	5	4/5	4/5

*Multifiber (1-Acetate, 2-Cotton, 3-Polyamide, 4-Polyester, 5-Acrylic, 6-Wool)

Table 6. Color fastness to rubbing

Sample code	Rubbing	
	Dry	Wet
C1	5	4/5
C3	5	4/5
C5	5	4
C7	5	4
C9	5	4
C12	5	3/4
V1	5	4/5
V3	5	4
V5	5	3/4
V7	5	3/4
V9	5	3/4
V12	5	3/4
W1	5	4/5
W3	5	4/5
W5	5	4/5
W7	5	4/5
W9	5	4/5
W12	5	4/5

4. CONCLUSION

The effects of SCC concentration on the dye exhaustion and color parameters in dyeing cotton, viscose and wool fabrics has been studied. Fabrics were pre-treated and then dyed with different concentrations of SCC without mordanting. The dye exhaustion values which were determined by spectrophotometric method indicates that the percentile dye exhaustion decreased with an increase in the SCC concentration and reached equilibrium when the SCC concentration is about 7 ml/L for cotton, 7 ml/L for viscose and 3ml/L for wool fabrics. More increase in the SCC concentration results with an increase in the amount of SCC that should remain in the effluent. The K/S values of dyed cotton, viscose and wool fabrics increased with increasing the concentration of SCC up to 9 ml/L while dyeing viscose and up to 12 ml/L while dyeing cotton and wool. The fastness ratings to rubbing and perspiration of cotton, viscose and wool fabrics dyed with different concentrations of SCC are good to excellent. Only in few cases color fastness to rubbing for wet test and color change after the color fastness to perspiration were observed fair to good level. Based on the color fastness results, it can be said that the fastness values of the SCC are suitable for the dyeing process.

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