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Application of Chitosan and Catechin to Improve Color Intensity and UV Protection in the Dyeing of Cotton Fabrics with Natural Dyes from *Peristrophe bivalvis*

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Abstract. The use of natural dyes as textile dyes is increasingly in demand since public awareness of the dangers of synthetic dyes have been increasing. Natural dyes produce a weak color intensity and require repeated dyeing to produce the desired color. This paper studied the effect of chitosan and catechin on the intensity and protection of fabrics in natural dyes from *Peristrophe bivalvis* expressed by parameters of UV protection factor (UPF) and color difference values (ΔE). The effects were observed between chitosan treatment and combination of chitosan and catechin treatment of dyed cotton fabric in various concentration. The application on cotton fabric treatment were done before dyeing process, after dyeing process, and combination of both. In result, as the concentration of chitosan and catechin increased, the color intensity increased as evidenced by the increase in the ΔE value. The best UPF value was obtained by combination treatment of chitosan and catechin on cotton fabric with the concentration of chitosan and catechin, respectively 15 g/L and 0.5 g/L in the treatment before and after dyeing resulted in UPF value of 7.22 and a color difference (ΔE) of 36.09. The best lightfastness obtained by a combination treatment of chitosan and catechin with the results increased to 3-4 (good).

1. Introduction

With increasing awareness of the dangers of synthetic dyes to the environment, the use of natural dyes as textile dyes become a trend. However, the performance of natural dyes as fabric dyes is not as great as synthetic dyes. Synthetic dyes produce bright and long-lasting colors while natural dyes produce low color intensity. Dyeing cotton fabrics with natural dyes often requires repeated dyeing to produce the desired color, so it takes a long time to get the final product. Besides, the non-variety of colors produced is also the reason for the lack of commercial use of natural dyes as fabric dyes.

Natural dyes that produce red shades are still very limited, one source of natural dyes that are promising as a source of natural red coloring is *Peristrophe bivalvis* leaves. *Peristrophe bivalvis* contains the pigment phenoxazine which produces red color [1]. Research on the use of natural dyes from *Peristrophe bivalvis* on fabrics has been carried out by Evitasari, et al. [2], the optimum color intensity produced is K/S of 0.597 using $FeSO_4$ as mordant to increase affinity of fabric to the dye, with a concentration of 10.96 o.w.f. % at dyeing temperature of 61°C.

To increase the color intensity and fastness, several ways can be done, one of which is by adding chemicals to the coloring process. In dyeing fabrics with natural dyes from *Peristrophe bivalvis*, this has been done with the addition of a UV absorber by Evitasari and Rahayuningsih [3]. In that study, a benzophenone and benzotriazole UV absorber were used. The addition of UV absorbers to natural dyes

from *Peristrophe bivalvis* can increase the fabric's protection against UV rays but does not increase the intensity of the color. This research will increase the color intensity of the fabric using natural sources. Chitosan has begun to be used as an additive in fabrics to improve various functions of fabrics, including increasing sun fastness, increasing anti-bacterial properties, and anti-wrinkling in fabrics [4], [5]. Chitosan comes from crustaceans, so it is environmentally friendly. Other natural compounds that can be used as additives after the dyeing process are the catechin in green tea. Catechin is a polyphenol and antioxidant compound in green tea. Green tea contains about 30% - 42% by weight of catechin. Catechin can increase the sun-resistant properties of fabrics because they reduce the penetration of ultraviolet rays on fabrics [4]–[9].

This paper studied the effect of chitosan and catechin on the intensity and protection of fabrics in natural dyes from *Peristrophe bivalvis* expressed by parameters of UV protection factor (UPF) and color difference values (ΔE). Observations were made on variations in the concentration of chitosan and catechin as well as variations in the application process of chitosan and catechin before and after dyeing and the combination of both to see the effect of chitosan and catechin on the fabric coloring process.

2. Materials and Methods

2.1. Materials

The fabric used was 100% bleached cotton from Yogyakarta. The Mordant used in this research was ferrous sulfate ($FeSO_4$) provided by Gamaindigo. *Peristrophe bivalvis* was grown and cultivated in Yogyakarta. Chitosan (degree of deacetylation 90%) was provided by ChemMix. Catechin was extracted from local green tea.

2.2. Methods

2.2.1. Chitosan Solutions. Chitosan was dissolved in a 0.5% v/v acetic acid solution at various concentrations of 5, 10, and 15 g/L. In the treatment before the dyeing process, the fabric was soaked for 15 minutes in a solution of chitosan at 50°C as much as 20 mL. In the treatment after the dyeing process was carried out without heating.

2.2.2. Catechin Solutions. Fabrics were treated with 15 g/L chitosan and then treated with catechin. The concentration of green tea added was 0.05; 0.1; and 0.5 g/L. In the treatment before the dyeing process, the fabric was soaked for 15 minutes in a 20 mL green tea solution at a temperature of 50°C. In treatment after the dyeing process was done without heating.

2.2.3. Dyeing Process. The pre-mordanting process used ferrous sulfate as mordant with a concentration of 10.96%. Then dyed with natural dye from *Peristrophe bivalvis* leaves at 61°C for one hour in a shaker bath.

2.2.4. UV Protection Factor (UPF) Analysis. Samples were analysed by taking the reflectance value between 290 nm to 400 nm using Shimadzu Spectrophotometer UV-2401PC. The UV Protection Factor (UPF) values were calculated by equation 1, where S_λ is spectrum source (Wm^2nm^{-1}), E_λ is response spectrum, T_λ is transmittance, and $\Delta\lambda$ is the wavelength (nm).

$$UPF = \frac{\sum_{290nm}^{400nm} E_\lambda S_\lambda \Delta\lambda}{\sum_{290nm}^{400nm} E_\lambda S_\lambda T_\lambda \Delta\lambda} \quad (1)$$

2.2.5. Analysis of Color Difference (ΔE). Analysis of color difference values was calculated to determine the change in color intensity based on the parameters of CIELab color coordinates L *, a *, and b *. The L *, a *, and b * values respectively indicate brightness, red-green shades, and yellow-blue shades. The

difference in color compared was the fabric treated with the addition of chitosan and catechin to the fabric without the addition of additives. The color difference value (ΔE) is calculated by equation 2.

$$\Delta E = \left[(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2 \right]^{1/2} \quad (2)$$

2.2.6. *Color Fastness Test*. Fabrics that have been dyed and treated with chitosan and catechin are tested for lightfastness, rubbing, and washing. The test is carried out based on the Indonesian National Standard (SNI). The test results in the visual form expressed in greyscale.

3. Results and Discussions

3.1. Chitosan Treatment on Dyed Cotton with Peristrophe bivalvis

In this study, chitosan treatment on cotton fabric dyed with natural dyes from *Peristrophe bivalvis* leaves can increase the fabric's protection against ultraviolet rays. Physically, there is a significant increase in the color intensity of the fabric with the addition of chitosan. The higher the concentration of chitosan added, the darker the resulting color. Besides, the fabric becomes stiffer. The stiffness of this fabric is due to the antistatic and anti-wrinkle properties of chitosan [10].

The value of UV Protection Factor (UPF) on fabrics dyed with natural dyes from *Peristrophe bivalvis* increases with the addition of chitosan concentrations. The UPF value with the addition of chitosan treatment is presented in Figure 1. The UPF value of untreated dyed cotton was 2.36 based on the previous study [3], the addition of chitosan before staining with a concentration of 5 g/L increased the UPF value to 4.13. The addition of the chitosan concentration value will increase the UPF value, at a concentration of 10 g/L and 15 g/L the UPF value is 4.36 and 4.37 in the application before the dyeing process. The increase in the UPF value along with the increasing chitosan concentration is due to the more evenly the chitosan is bound to the surface of the fabric, thus providing better protection.

The increase in different UPF values occurred in variations in the treatment process of chitosan application on cotton fabrics, before dyeing, after dyeing, and the combination of both. The highest UPF value resulted from the addition of chitosan before and after dyeing, followed by treatment before dyeing, and the lowest UPF value was in the addition of chitosan after the dyeing process. The highest UPF value was obtained when the addition of 15 g/L of chitosan was 6.16 in the addition process before and after dyeing. Followed by treatment before the dyeing process produces a UPF of 4.79 and the lowest is 3.31 in the chitosan application process after dyeing.

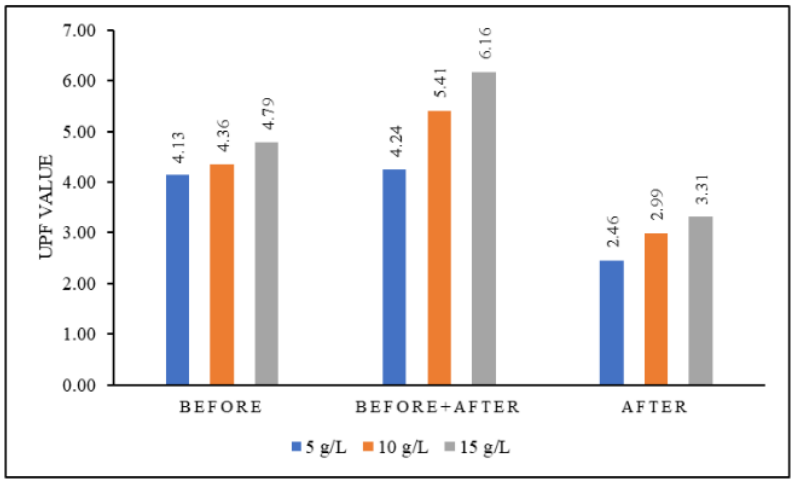

Figure 1. UPF Result with Chitosan Treatment.

Analysis of the color coordinate values and color difference (ΔE) on untreated fabrics and Chitosan treated fabrics are presented in Table 1. The higher the ΔE value, the darker the resulting color will appear, in other words, the higher the color intensity. The highest value of ΔE color difference in the addition of chitosan before and after dyeing was 32.53 at a concentration of 15 g/L. in the addition of chitosan before dyeing the difference was only slightly lower, at 31.56 in the same concentration. However, in the addition of chitosan after dyeing process, it only slightly increased the intensity of the color difference to 9.55 at a chitosan concentration of 15 g/L.

Table 1. Color Coordinates and Color Difference (ΔE) Value on Chitosan Treatment.

Treatment	Chitosan Concentration	L*	a*	b*	ΔE	Shades
Before	5 g/L	47.99	41.05	22.06	25.88	
	10 g/L	49.01	42.26	27.68	29.60	
	15 g/L	40.70	38.49	24.50	31.56	
Before + After	5 g/L	52.44	44.25	24.29	26.50	
	10 g/L	46.34	42.38	26.98	30.67	
	15 g/L	39.05	39.30	23.51	32.53	
After	5 g/L	65.26	32.22	6.00	3.08	
	10 g/L	65.41	32.72	5.66	3.64	
	15 g/L	55.73	32.42	6.25	9.55	

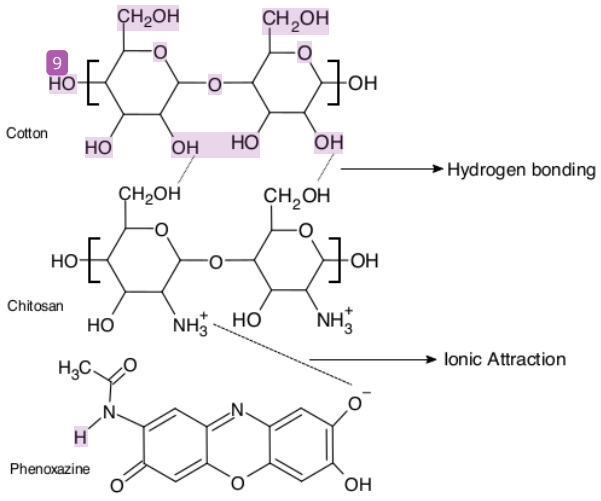

Figure 2. Bonding Mechanism between Cotton, Chitosan, and Phenoxazine.

The increasing amount of dye absorbed into the cotton fabric is caused by the amine group in chitosan, so there is an empty site to be filled with phenoxazine dyes [11]. Cotton fabrics have a low affinity for dyes because they are both negatively charged (anions) in solution. Chitosan acts as a mordant in fabrics because chitosan is positively charged (cation) in acidic conditions, so chitosan becomes a bridge between cotton fabric and dyes, in this study phenoxazine [6], [9]. Chitosan will form hydrogen bonds with cotton fabric, and form ionic bonds with phenoxazine dyes. The bonding mechanism that occurs between cotton, chitosan, and phenoxazine is presented in Figure 2.

3.2. Combination of Chitosan and Catechin Treatment on Dyed Cotton with Peristrophe bivalvis

In this study, the addition of a combination of chitosan and catechin from green tea to fabrics dyed with natural dyes from *Peristrophe bivalvis* leaves was intended to increase the fabric's protection against ultraviolet rays. The color produced on the cotton fabrics with the addition of chitosan and catechin was darker than the fabric treated with chitosan only. The higher the catechin concentration added, the darker the resulting color.

The value of UV Protection actor (UPF) on fabrics stained with natural dyes from *Peristrophe bivalvis* leaves with a combination treatment of chitosan and catechin is presented in Figure 3. The lowest UPF value with the addition of catechin after dyeing is with a concentration of 0.05 g/L, the UPF value is 2.61. The highest increase in UPF was achieved at the addition of the catechin concentration of 0.5 g/L by the treatment before and after the dyeing process at 7.22, the UPF value increased by 205%.

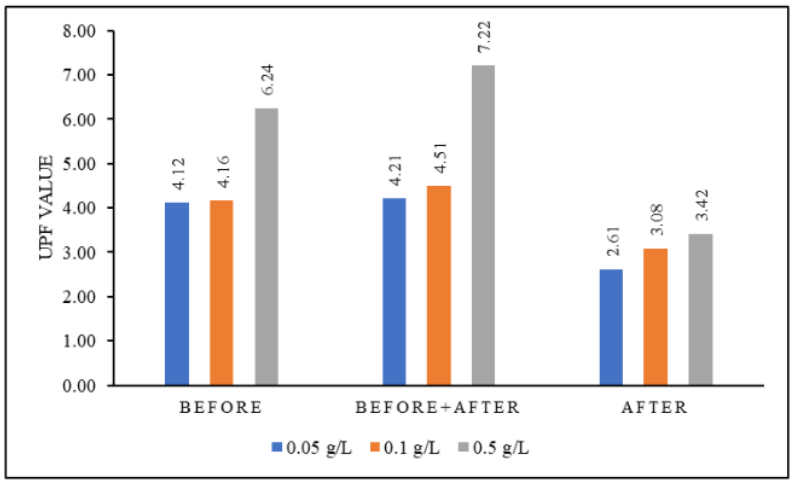

Figure 3. UPF Result with Combination of Chitosan and Catechin Treatment.

In the variation of treatment, the highest UPF value resulted from the addition of catechin before and after dyeing, followed by treatment before dyeing, and the lowest UPF value was in the addition of chitosan and catechin after the dyeing process. The increase in the UPF value occurred due to the addition of chitosan which functions as a mordant, and the addition of catechin as antioxidants so that they can ward off ultraviolet rays. The catechin extracted from green tea had a light brown color. This has an impact on the resulting fabric to be darker than the original color. Although catechin gave a dark color, they did not change the purplish-red color produced by phenoxazine from *Peristrophe bivalvis* leaf extract.

Analysis of color coordinate values and color difference (ΔE) on the combination treatment of chitosan and catechin are presented in Table 2. The higher the ΔE value, the darker the color will be. The highest value of ΔE color difference in the addition of chitosan before and after coloring was 36.09 at a concentration of 15 g/L. The addition of chitosan before dyeing obtained ΔE 33.61 at the same concentration. However, in the addition of chitosan after coloring, there was only a slight difference in color to 13.78 at a concentration of 15 g/L. The insignificant increase in the value of ΔE in the

application process of chitosan and catechin after dyeing shows that chitosan does not have the ability like mordant metal. Mordant metals such as $FeSO_4$ have the ability to darken natural dyes in fabrics. The application of chitosan after dyeing was able to lock the color on the cotton but did not increase the color intensity.

Table 2. Color Coordinates and ΔE on Combination of Chitosan and Catechin Treatment.

Treatment	Catechin Concentration	L*	a*	b*	ΔE	Shades
Before	0.05 g/L	51.48	45.52	25.34	28.38	
	0.10 g/L	43.06	38.41	25.31	30.28	
	0.15 g/L	34.68	31.29	15.81	31.59	
Before + After	0.05 g/L	38.70	34.87	20.98	30.43	
	0.10 g/L	41.83	41.81	27.40	33.61	
	0.15 g/L	42.98	43.30	31.38	36.09	
After	0.05 g/L	68.08	36.22	7.46	7.87	
	0.10 g/L	63.89	41.53	8.77	12.61	
	0.15 g/L	63.68	41.73	11.90	13.78	

Catechin bonds to fabrics just like phenoxazine dyes bond to fabrics. Catechin can be bound to the metal mordant and chitosan. The mechanism for the formation of complex compounds between cotton, chitosan, and catechin is presented in Figure 4.

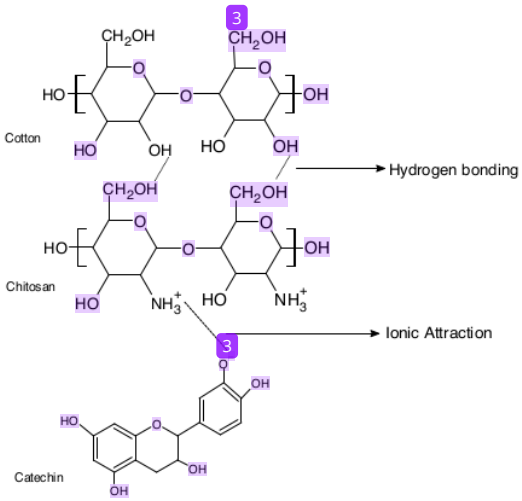

Figure 4. Bonding Mechanism between Cotton, Chitosan, and Catechin.

3.3. Color Fastness Properties

The fabric dyed with natural dyes from the leaves of *Peristrophe bivalvis* without treatment has fair fastness to sunlight and washing with a value of 2-3 for both [2]. The treatment of chitosan and catechin in dyeing fabrics with natural dyes from *Peristrophe bivalvis* leaves increased the fastness value, both from sunlight and washing. The best fastness resistance is obtained in fabric treatment with a combination of chitosan and catechin with a fastness rating to sunlight and washing of 3-4 or good and 3 or fair, respectively.

Table 3. Fastness Rating to Sunlight, Washing and Rubbing.

Treatment	Fastness		
	Sunlight	Washing	Rubbing
Untreated	2 – 3 (Fair)	2 – 3 (Fair)	4 – 5 (Good)
Chitosan	3-4 (Good)	3 (Fair)	4 – 5 (Good)
Chitosan & Catechin	3-4 (Good)	3 (Fair)	4 – 5 (Good)

4. Conclusion

The addition of chitosan and catechin can increase the color intensity of the fabric dyeing with natural dyes from *Peristrophe bivalvis*. As the concentration of chitosan and catechin increased, the color intensity increased as evidenced by the increase in the UPF and ΔE value. The best UPF value was obtained by combination treatment of chitosan and catechin on cotton fabric with the concentration of chitosan and catechin, respectively 15 g/L and 0.5 g/L in the treatment before and after dyeing resulted in UPF value of 7.22 and a color difference (ΔE) of 36.09. Supported by the results of the fastness to sunlight obtained by a combination treatment of chitosan and catechin with the results increased to 3-4 (good).

Acknowledgments

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