RESEARCH ARTICLE

Optimization of cotton fabrics dyeing process using various natural dye extracts

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OBJECTIVES This study aims to optimize the dyeing parameters of cotton fabrics using natural dye extracts derived from the barks of Tegeran (Cudrania javanensis), Merbau (Intsia bijuga), Tingi (Ceriops tagal), and Jambal (Peltophorum pterocarpum), as well as Jolawe (Terminalia bellirica) fruit peel. METHODS Varied dyeing parameters included dye concentration, dyeing time and temperature, and material to liquor ratio (MLR). The fixative solutions used were alum, lime, and iron (II) sulfate. The optimized parameters were based on the values of color depth and evenness, measured colorimetrically. Furthermore, the obtained results of the dyeing process under optimized conditions were analyzed for color quality by measuring color coordinates as well as the values of color strength (K/S), washing fastness, and light fastness. **RESULTS** The results showed that the optimal dyeing conditions for all natural dye extracts used were the code A extract concentrations (0.0113 g/mL of Tegeran; 0.0115 g/mL of Merbau; 0.0204 g/mL of Jambal; and 0.0582 g/mL of Jolawe), dyeing at 28°C, dyeing time of 30 minutes, and the MLR of 1:30. The resulting color variations were brown, gray, and golden yellow for the Tegeran extract with alum fixative. The highest K/S value was 5.56 for the fabric dyed in Tegeran extract with iron (II) sulfate fixative solution. The washing fastness values for Merbau, Tingi, Jambal, and Jolawe were 3-4 (fairly good) to 4-5 (excellent). Meanwhile, the light fastness values for all dyes were between 4 (good) and 5

(excellent). **CONCLUSIONS** Overall, the standard procedure for cotton fabric dyeing that meets the minimum standards for textile products is obtained.

KEYWORDS cotton fabric; color quality; natural dyes; optimization; standard procedure for dyeing

1. INTRODUCTION

In line with the increasing awareness of environmental sustainability and health, natural dyes are increasingly used in textiles as substitutes for synthetic dyes. Natural dyes can be obtained directly from available materials in nature and are more environmentally friendly than synthetic dyes. Generally, natural dyes have a relatively low wash and light fastness than synthetic ones (Samanta and Agarwal 2011). To improve the quality of natural dyes so that they can be more competitive than synthetic dyes, research and innovation are therefore needed to be conducted.

Based on their application, dyes are classified into six types, namely: mordant dyes, direct dyes, vat dyes, acid dyes, alkaline dyes, and dispersion dyes (Chakraborty 2010). Natural dyes, especially mordant dyes, and vat dyes have been widely used in Indonesia for decades. Since natural dye of the mordant type is water-soluble, it requires a fixator in the application to bind the dye to the fiber of the fabric (Teklemedhin 2018). Natural dyes and natural fibers can form weak hydrogen bonds. Metallic fixatives with cationic properties will strongly bind anionic dyes to fabric fibers. Metal cations (fixative solution) act as a bridge between the dye and the fabric fiber, resulting in better quality and color resistance of the fabric. In addition, through the variation of metal cations as the fixative solution, color variations can be produced for one type of natural dye mordant (Parveen et al. 2019). According to Singh and Bharati (2014), there are three recommended and safe-to-use fixators, namely: alum, lime, and iron (II) sulfate.

Farooq et al. (2013) stated that natural dyes are mainly obtained from various parts of plants (roots, stems, flowers, seeds, or leaves). Tegeran (*Cudrania javanensis*) extract is used as a source of yellow dye. The yellow color of Tegeran wood comes from the content of morin compounds (Kongkiatpaiboon et al. 2016). Merbau (*Intsia bijuga*) bark contains tannin compounds that can be extracted and used as a source of natural brown dye (Rahayuningsih et al. 2020). Tannin compounds are also found in the bark of Tingi (*Ceriops tagal*), with percentages varying from 13 to 40% (Handayani and Maulana 2013; Purwani and Ndawu 2019). Meanwhile, the coloring process of Jambal (*Peltophorum pterocarpum*) bark extract, which also contains tannins, produces a lighter reddishbrown color than that of Tingi (Muthiah and Evvyani 2019). Jolawe (*Terminalia bellirica*) fruit peel also contains tannins and the extract produces a greenish-brown color (Purwani and Ndawu 2019). Morin and tannins are polyphenol compounds. Therefore, their application as a dye on fibers requires a mordant.

Cotton is the most used natural fiber in the textile industry. It is composed of highly polymerized cellulose with excellent tensile strength, water absorbency, and air permeability (Ismail 2018). The quality parameter of the natural dyeing process on textile materials is the stability of the complex formed between the dye and the fiber. The stability of the dye-fiber complex is influenced by the structure of the fiber, the dyeing time and temperature, and the characteristics of the dye molecules (Kovačević et al. 2021). Furthermore, the method applied to extract dyes from the plants also affects the quality of the resulting color. Studies on the optimization of dye extraction have been carried out for Merbau and Tingi barks (Handayani and Maulana 2013; Indah et al. 2010). However, the study on the attempt of optimizing the dyeing process of natural dye extracts on cotton fabrics has not been carried out previously.

The purpose of this study is to determine the optimal conditions for the dyeing process of cotton fabrics using natural dyes derived from the barks of Tegeran, Merbau, Tingi, and Jambal as well as the peel of Jolawe fruit. The optimized bound parameters were based on the color depth and color evenness of the dyed cotton fabric. The independent variables examined in this study were extract concentration, dyeing time and temperature, and the ratio of material to the volume of fixative solution. The dyeing results with optimized conditions were further analyzed by measuring the value of color coordinates, color strength (K/S), washing fastness, and light fastness.

2. MATERIALS AND METHODS

2.1 Materials

The materials used in the dyeing process were concentrated extracts of Tegeran, Merbau, Tingi, Jambal, and Jolawe obtained from Gama Indigo Workshop Gallery, Yogyakarta. Meanwhile, the cotton fabrics with 0.35 thickness and 192 g/m² of weight per area were supplied by PT Primissima Yogyakarta. The technical grades of soda ash, alum, lime, iron (II) sulfate, and distilled water used for the pre-mordanting and post-mordanting processes were purchased from local chemical shops in Yogyakarta.

2.2 Pre-mordanting process

The pre-mordanting process was carried out by immersing the cotton fabric in a mixture of 1% alum and 0.3% soda ash at a material-to-liquor ratio of 1:10 at 100°C for 60 minutes. After the pre-mordanting process, the fabrics were rinsed with water and dried at room temperature overnight.

2.3 Preparation of dye concentrations

Each natural dye extract obtained from Gama Indigo Yogyakarta was tested for the total dye concentration using gravimetric analysis. A total of 10 mL sample was taken with a volume pipette and dried in an oven at a temperature of 65°C until a constant weight was achieved. The initial solution is hereinafter referred to as the extract with the 'code A' dye variation. Furthermore, for variations in the concentration of the dye solution, each code A extract was concentrated separately. A total of 250 mL of natural dye extract solution was heated at a temperature of 70°C on a hot plate stirrer. The extract solution was concentrated until the volume reached 200 mL to obtain the extract of the 'code B' dye variation. Meanwhile, to obtain an extract with the 'code C' dye variation, the solution was concentrated to reach 150 mL of volume. Code B and code C concentrated extract solutions were tested for total dye concentration by applying gravimetric analysis. The dye concentrations for various natural dyes are listed in Table 1.

2.4 Dyeing process

The dyeing of cotton fabrics was carried out by immersion method with three dye variations (A, B, C) at a material-toliquor ratio (MLR) of 1:10 at different temperatures of 28, 40, 60, and 70°C, and for different dyeing times of 10, 20, 30, and 40 minutes. The pre-mordant dry fabric was weighed as much as 1 g and immersed in 10 mL of extract solution. The dyeing process was carried out in a shaker bath with temperature and time settings according to the experimental design. After the dyeing process, the fabrics were rinsed with water and dried at room temperature (28-30°C) for 2 hours.

2.5 Post-mordanting process

The post-mordanting process was carried out by immersing the dyed fabrics in different fixative solutions (alum, lime, and iron (II) sulfate). Each fixative solution was dissolved separately with boiled water at a concentration of 1:10 (w/v). The solution was stirred for 1 hour with a magnetic stirrer. The solution was then left overnight to precipitate the remaining solids and then filtered through filter paper to collect the filtrate. The post-mordanting process was done in various

TABLE 1. T	he dye cor	ncentration	of the	extracts
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Dye Variation	Dye Concentration (g/mL)						
_	Tegeran	Merbau	Tingi	Jambal	Jolawe		
A	0.0113	0.0115	0.0204	0.0104	0.0582		
В	0.0138	0.0130	0.0294	0.0130	0.0622		
С	0.0186	0.0235	0.0330	0.0193	0.0957		

TABLE 2.	Effect of	dye	concentration	on	the	L* va	lue
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Samples	L*				
	Alum	Lime	Iron (II) sulfate		
Tg_A_1:10_60_20	64.35 ± 0.256	68.42 ± 0.234	34.51 ± 0.333		
Tg_B_1:10_60_20	64.26 ± 0.671	68.36 ± 0.436	34.13 ± 0.557		
Tg_C_1:10_60_20	63.84 ± 0.738	67.42 ± 0.645	33.89 ± 1.003		
Me_A_1:10_60_20	62.45 ± 0.309	61.41 ± 0.493	43.12 ± 0.545		
Me_B_1:10_60_20	61.34 ± 0.830	60.77 ± 0.321	42.53 ± 0.768		
Me_C_1:10_60_20	61.45 ± 1.079	60.56 ± 0.244	41.84 ± 0.776		
Ti_A_1:10_60_20	51.00 ± 1.177	40.35 ± 1.759	30.64 ± 0.590		
Ti_B_1:10_60_20	50.92 ± 1.500	39.65 ± 2.482	29.55 ± 1.199		
Ti_C_1:10_60_20	49.81 ± 1.987	38.14 ± 2.263	29.23 ± 1.357		
Ja_A_1:10_60_20	47.78 ± 0.504	43.00 ± 0.622	34.24 ± 0.479		
Ja_B_1:10_60_20	47.55 ± 0.556	42.73 ± 0.740	33.62 ± 0.719		
Ja_C_1:10_60_20	46.34 ± 0.830	42.28 ± 0.889	32.44 ± 0.903		
JL_A_1:10_60_20	73.64 ± 0.529	67.55 ± 0.710	36.16 ± 0.396		
Jl_B_1:10_60_20	73.25 ± 0.824	66.76 ± 0.985	35.91 ± 0.262		
JL_C_1:10_60_20	73.39 ± 0.739	66.48 ± 0.953	34.84 ± 0.581		

mass of cotton fabric to the volume of fixative solution ratios (MLR) of 1:10, 1:20, 1:30, and 1:40. This process was conducted at 60°C for 10 minutes. After the post-mordanting process, the fabrics were dried at room temperature, followed by the hot soaping process.

2.6 Samples labeling

Samples labeling was determined based on the code of DV_DC_ML_DT_Dt, where: DV is the dye variation (Tegeran, Merbau, Tingi, Jambal, and Jolawe); DC is the dye concentration (A, B, C); ML is the material-to-liquor ratio in fixator ratio (1:10, 1:20, 1:30, 1:40); DT is the dyeing temperature (28, 40, 60, and 70°C); and Dt is the dyeing time (10, 20, 30, and 40 minutes).

2.7 Color measurement

Color measurement of dyed cotton fabric was performed using FRU WR-10 Colorimeter. The color value is determined based on coordinates in the CIELab color space system, namely the "L*" value for color brightness, 0 for black and 100 for white, and a* value for red (positive value) and green (negative value), and b* value for yellow (positive value) and blue (negative value).

The color strength analysis of the fabric was carried out using the Shimadzu UV-2401PC spectrophotometer in a wavelength range of 350-750 nm. The test results are the reflectance values (%R). The color strength (K/S) is obtained from the conversion of the %R value using the Kubelka-Munk equation (1).

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

2.8 Color fastness properties

The color fastness to washing test was performed according to ISO 105-C06:2010. A 4 cm x 10 cm of dyed fabric was

placed between a 100% white cotton fabric and a 100% white polyester fabric in the same size and then sewn together on their shortest side. The fabrics were put into a closed vessel containing 150 mL of AATCC soap (4 g/L) and sodium perborate (1 g/L) solution and 10 marbles. The washing process was conducted in the Launder O Meter machine at 40°C for 30 minutes. The fabrics were then rinsed with 0.2 g/L of glacial acetic solution before being dried at room temperature. The color change was measured using the grayscale, with a scale of 0 (poor) to 5 (excellent).

The color fastness to light test was determined according to the American Association of Textile Chemists and Colorists (AATCC) 16. The test was performed by irradiating the fabrics using a Xenon Arc lamp ($48W/m^2$)for 30 minutes. The color change between the tested fabrics and the un-irradiated fabrics was measured using the grayscale, with a scale of 0 (poor) to 5 (excellent).

3. RESULTS AND DISCUSSION

3.1 Effect of dye concentration

The optimized parameters for the cotton fabric dyeing process using natural dyes (Tegeran, Merbau, Tingi, Jambal, and Jolawe) were selected based on the color depth and color evenness of the dyed cotton fabric. The color depth value was determined on the L* value of the CIELab color coordinate measurement results. A smaller L* value indicates the darker shade produced, meaning that a greater amount of dye is attached to the cotton fiber. The color evenness was represented by the standard deviation value obtained from the L* values of several parts on the fabric surface. The lower the standard deviation value, the narrower the distribution of L* values, which means the color evenness is higher. From the L* values displayed in Table 2, it can be seen that all types of natural dyes for each type of fixator have a similar pattern: increased dye concentration resulted in a slightly deeper color. On the other hand, the decrease in the L* value is proven to

be insignificant. The insignificance of the L* value is due to the equilibrium that occurs between the binding and the releasing of the dye on the fabric. Thus, the optimal dye concentration is only determined by the standard deviation of the L* value. Table 2 shows that the standard deviation of the L * value increases with a higher concentration of dye. The higher the standard deviation value, the wider the distribution of L* values, and the lower the color evenness value of the fabric. The decrease in the color evenness value is due to the large number of aggregates formed in a more concentrated dye solution. The aggregates can induce blocking of the dye at a specific part of the fabric so that the color evenness is reduced. Based on the standard deviation of the L* value, code A dye concentration was proven to be the optimal concentration in the cotton fabric dyeing process using natural dyes examined in this study.

3.2 Effect of dyeing temperature

As presented in Table 3, each natural dye shows the same trend in heat treatment; there was a slight decrease in the L* value along with the increase in temperature, accompanied by a decrease in the L* value when a certain temperature was reached. The deeper color result produced with increasing temperature is due to swelling of the cotton fiber leading to higher dye uptake by the fiber (Farooq et al. 2013). However, since the change in the L* value was insignificant, the chosen optimal temperature was 28°C, taking into account the energy efficiency used in the dyeing process.

3.3 Effect of dyeing times

From Table 4 below, it can be concluded that the longer the dyeing time, the lower the L^* value achieved. It is possible

TABLE 3. Effect of dyeing temperature on the L* value.

as more dyes are attached to the fiber. However, the average L* value of the cotton fabric during the 40-minute dyeing time did not deviate significantly from the L* value of the 30-minute dyeing time. At a specific dyeing time, an equilibrium between the binding and the releasing of the dye on the fiber will be achieved, so that the amount of dye absorbed into the fiber will be constant (Farooq et al. 2013). Based on the data shown in Table 4, it is known that the dyeing time does not crucially affect the color evenness, as visually seen or based on the obtained standard deviation of the L* value. Thus, the optimized dyeing time is selected based on the optimal L* value, i.e. 30-minute of dyeing time.

3.4 Effect of Material to Liquor Ratio (MLR) in the post-mordanting process

As seen in Table 5, the effect of MLR on the color depth of the dyed fabrics (L* value) is not incredibly notable. It can be said that by using the lowest MLR, good color depth can be produced. On the other hand, MLR has a significant effect on color evenness. The greater the MLR, the smaller the standard deviation of the L* value, and the better the color evenness achieved. High MLR causes a higher possibility of contact between the fabric and the fixative solution. There is a similarity in the standard deviation of the L* value in the MLR of 1:30 and 1:40. Considering the material efficiency, the MLR of 1:30 is seen as the optimal one.

3.5 The quality of dyed fabric with optimized parameters

The quality of cotton fabric dyed with various natural dyes derived from Tegeran, Merbau, Tingi, Jambal, and Jolawe at optimized dyeing parameters was analyzed according to the color coordinates, and the color strength, the washing fast-

Samples		L*	
	Alum	Lime	Iron (II) sulfate
Tg_A_1:10_28_20	64.91 ± 0.255	68.63 ± 0.313	35.27 ± 0.341
Tg_A_1:10_40_20	64.26 ± 0.237	68.61 ± 0.299	34.55 ± 0.350
Tg_A_1:10_60_20	64.35 ± 0.256	68.42 ± 0.234	34.51 ± 0.333
Tg_A_1:10_70_20	65.01 ± 0.223	68.99 ± 0.243	35.73 ± 0.304
Me_A_1:10_28_20	62.95 ± 0.275	61.78 ± 0.500	43.52 ± 0.550
Me_A_1:10_40_20	62.38 ± 0.283	61.23 ± 0.504	43.33 ± 0.531
Me_A_1:10_60_20	62.45 ± 0.309	61.41 ± 0.493	43.12 ± 0.545
Me_A_1:10_70_20	63.53 ± 0.246	61.77 ± 0.472	43.72 ± 0.517
Ti_A_1:10_28_20	50.93 ± 1.205	40.84 ± 1.728	30.72 ± 0.642
Ti_A_1:10_40_20	50.32 ± 1.196	40.87 ± 1.747	30.83 ± 0.617
Ti_A_1:10_60_20	51.00 ± 1.177	40.35 ± 1.759	30.64 ± 0.590
Ti_A_1:10_70_20	50.79 ± 1.165	41.61 ± 1.684	30.70 ± 0.601
Ja_A_1:10_28_20	47.89 ± 0.537	43.06 ± 0.637	34.63 ± 0.528
Ja_A_1:10_40_20	47.45 ± 0.532	43.11 ± 0.641	34.26 ± 0.503
Ja_A_1:10_60_20	47.78 ± 0.504	43.00 ± 0.622	34.24 ± 0.479
Ja_A_1:10_70_20	47.99 ± 0.499	43.57 ± 0.630	34.70 ± 0.485
JL_A_1:10_28_20	74.13 ± 0.556	67.77 ± 0.704	36.15 ± 0.421
Jl_A_1:10_40_20	73.96 ± 0.538	67.59 ± 0.691	36.21 ± 0.400
Jl_A_1:10_60_20	73.64 ± 0.529	67.55 ± 0.710	36.16 ± 0.396
JL_A_1:10_70_20	74.02 ± 0.493	67.74 ± 0.697	36.23 ± 0.405

Samples	L*				
	Alum	Lime	Iron (II) sulfate		
Tg_A_1:10_28_10	65.13 ± 0.200	69.25 ± 0.354	36.14 ± 0.301		
Tg_A_1:10_28_20	64.91 ± 0.237	68.63 ± 0.299	35.27 ± 0.350		
Tg_A_1:10_28_30	64.42 ± 0.212	67.57 ± 0.337	33.76 ± 0.383		
Tg_A_1:10_28_40	64.35 ± 0.258	67.33 ± 0.365	33.48 ± 0.416		
Me_A_1:10_28_10	64.06 ± 0.198	62.59 ± 0.336	45.11 ± 0.655		
Me_A_1:10_28_20	62.95 ± 0.283	61.78 ± 0.504	43.52 ± 0.531		
Me_A_1:10_28_30	61.25 ± 0.418	60.72 ± 0.655	42.29 ± 0.479		
Me_A_1:10_28_40	60.89 ± 0.440	60.47 ± 0.781	41.53 ± 0.420		
Ti_A_1:10_28_10	52.76 ± 1.007	41.67 ± 1.321	33.13 ± 0.548		
Ti_A_1:10_28_20	50.93 ± 1.196	40.84 ± 1.747	30.72 ± 0.617		
Ti_A_1:10_28_30	49.44 ± 1.485	39.00 ± 1.900	29.09 ± 0.895		
Ti_A_1:10_28_40	48.92 ± 1.503	38.53 ± 1.989	28.76 ± 0.882		
Ja_A_1:10_28_10	47.43 ± 0.510	44.21 ± 0.485	34.97 ± 0.516		
Ja_A_1:10_28_20	47.89 ± 0.532	43.06 ± 0.641	34.63 ± 0.503		
Ja_A_1:10_28_30	45.02 ± 0.762	41.84 ± 0.971	33.55 ± 0.848		
Ja_A_1:10_28_40	44.92 ± 0.818	40.63 ± 1.134	33.28 ± 0.942		
JL_A_1:10_28_10	74.33 ± 0.451	68.00 ± 0.685	36.40 ± 0.443		
JL_A_1:10_28_20	73.96 ± 0.538	67.59 ± 0.691	36.21 ± 0.400		
JL_A_1:10_28_30	73.15 ± 0.555	66.63 ± 0.773	34.29 ± 0.782		
JL_A_1:10_28_40	72.84 ± 0.560	66.64 ± 0.780	33.75 ± 0.864		

TABLE 4. Effect of dyeing time on the L* value.

ness, and the light fastness. The dyeing process was conducted in the same procedure for all natural dyes used, starting from pre-mordanting the cotton fabric with alum and soda ash solution, dyeing it with natural dyes, and finishing in the post-mordanting process using a fixative solution. The pre-mordanting process overcomes the problem of low affinity between natural dyes and cotton fabrics. The metal ions of the mordant solution can act as a bridge between the dye and the fabric so that the dye can bond to the fabric. The post-mordanting process determines the color coordinates of the dyed cotton fabric. The application of several fixative solutions (alum, lime, and iron (II) sulfate) in the dyeing process for the same natural dye can produce a variety of shades. The color variation is due to different interactions between the metal ions in the fixative solution and the dye molecules. Thus, the metal ions of the mordant act as color determinants and bridge between the dye and the fiber.

Natural dyes are mainly composed of polyphenolic compounds, such as tannins (Merbau, Tingi, Jambal, and Jolawe) and morins (Tegeran), as seen in Figure 1. In the solution, alum (KAl(SO_4)₂ 12H₂O) dissociated into Al³⁺ ions providing



FIGURE 1. Chemical structure of tannin (left) and morin (right) (Kavitha and Kandasubramanian 2020; Wei et al. 2015).

3s, 3p, and 3d orbitals as Lewis acid sites to bind the -OH and -C=O functional groups of the dye molecules and the -OH functional groups of the cellulose (Lewis base sites/ligands), forming metal-ligand complexes (Ding and Freeman 2017). In lime (CaCO₃) solution, ionized Ca^{2+} ions provided 3d, 4s, and 4p orbitals, while Fe^{2+} ions in iron sulfate ($FeSO_4.7H_2O$) solution provided 4s, 4p, and 4d orbitals as binding sites for dye molecules and fibers (Pamungkas et al. 2021). The wide range of colors of the dyed fabric from various fixative solutions is distinguished by the metal-ligand complex formed. The color of natural dyes results from the absorption of light by the chromophore functional group (e.g., -C=O) at a specific intensity and wavelength. By the metal, ions-chromophore interaction causes a shift in the absorbed wavelength, hence the variation of color achieved. As visually seen in the fabric and the resulting color coordinates, for the alum fixative solution, each dye produced a yellow color for Tegeran, brown for Merbau, reddish-brown for Tingi, light reddish-brown for Jambal, and light beige for Jolawe. For the lime solution, each dye gave off the same color in darker shades, except for Tegeran which created a light yellow color. Meanwhile, for the iron (II) sulfate solution, the dyes constructed dark greenish-brown for Tegeran, dark grayish brown for Merbau, dark brown for Tingi and Jambal, and dark gray for Jolawe.

For Tegeran, Merbau, and Jambal, the fixative solution affected the K/S values (see Table 6); K/S values of lime < alum < iron (II) sulfate. This indicates that Fe²⁺ ions in the iron (II) sulfate solution have a higher affinity for the dye than other fixative solutions. It was slightly different for Tingi and Jolawe, where the K/S values of the dyed fabrics in various fixative solutions were alum < lime < iron (II) sulfate, respectively. The smaller the orbital energy difference and the higher compati-

Samples	L*				
	Alum	Lime	Iron (II) sulfate		
Tg_A_1:10_28_30	64.42 ± 0.212	67.57 ± 0.337	33.76 ± 0.383		
Tg_A_1:20_28_30	64.77 ± 0.151	67.76 ± 0.194	32.53 ± 0.309		
Tg_A_1:30_28_30	64.63 ± 0.072	67.62 ± 0.132	32.54 ± 0.246		
Tg_A_1:40_28_30	64.81 ± 0.069	67.45 ± 0.134	33.26 ± 0.251		
Me_A_1:10_28_30	61.25 ± 0.418	60.72 ± 0.655	42.29 ± 0.479		
Me_A_1:20_28_30	60.90 ± 0.236	60.44 ± 0.491	42.87 ± 0.403		
Me_A_1:30_28_30	61.08 ± 0.165	60.88 ± 0.232	42.21 ± 0.295		
Me_A_1:40_28_30	61.13 ± 0.155	61.06 ± 0.199	42.08 ± 0.282		
Ti_A_1:10_28_30	49.44 ± 1.485	39.00 ± 1.900	29.09 ± 0.895		
Ti_A_1:20_28_30	51.00 ± 0.997	40.03 ± 0.852	30.24 ± 0.587		
Ti_A_1:30_28_30	49.61 ± 0.443	39.41 ± 0.558	29.73 ± 0.421		
Ti_A_1:40_28_30	50.03 ± 0.381	39.23 ± 0.501	29.66 ± 0.410		
Ja_A_1:10_28_30	45.02 ± 0.762	41.84 ± 0.971	33.55 ± 0.848		
Ja_A_1:20_28_30	45.36 ± 0.560	42.33 ± 0.405	33.79 ± 0.444		
Ja_A_1:30_28_30	45.47 ± 0.302	42.60 ± 0.309	33.74 ± 0.277		
Ja_A_1:40_28_30	45.72 ± 0.322	42.81 ± 0.283	34.98 ± 0.195		
JL_A_1:10_28_30	73.15 ± 0.555	66.63 ± 0.773	34.29 ± 0.782		
Jl_A_1:20_28_30	74.51 ± 0.393	67.08 ± 0.479	34.54 ± 0.392		
JL_A_1:30_28_30	74.25 ± 0.224	67.76 ± 0.385	35.01 ± 0.262		
JL_A_1:40_28_30	74.94 ± 0.219	67.90 ± 0.403	35.23 ± 0.246		

bility of the coordinate position between the empty orbital of the metal ion and the electron-containing orbital of the dye molecule, the easier it is to form metal-ligand complexes.

In addition to producing a wider range of colors, fixative solutions are used to improve the fastness properties of the dyed fabric. The cotton fabrics dyed in Merbau, Tingi, Jambal, and Jolawe extract solutions have reached the minimum standard of washing fastness on textile products according to SNI 0051:2008, with a scale of 3-4 (fairly good) as seen in Table 6). The good fastness properties of the dyed fabrics indicate that the metal-ligand-cellulose complex (Figure 2) produced by the interaction of $\mathrm{Al}^{3+}, \mathrm{Ca}^{2+},$ and Fe^{2+} ions with dye molecules possesses great stability and high bond energy. The washing fastness of Tegeran-dyed cotton fabric is on a scale of 3 (fair), meaning that it does not meet the standard. The formed complexes of Al^{3+} , Ca^{2+} , and Fe^{2+} ions with ligands from the dye molecules of Tegeran bark extract are seen to be less stable than the other dyes with metal-ligand complexes. Furthermore, the hydrophilicity of Tegeran dye may be due to its low washing fastness.



FIGURE 2. Formation of a coordinate complex between cellulose fiber and dye (Ding and Freeman 2017)

The scales of light fastness for all dyes were between 4 (good) and 5 (excellent). According to Pamungkas et al. (2021), the presence of a metal-ligand complex causes the delocalization of electrons which stabilizes the energy absorbed by the chromophore and the metal-ligand complex. This stability of absorbed energy reduces the possibility of dye degradation. No natural dyes used have surpassed the washing fastness rating of reactive dyes (four to five). The reactive dyes exhibit excellent washing fastness due to their stable covalent bonds with cellulose fiber. However, the widely used azo reactive dyes only have fair light fastness (rating scale of 3) due to unstable electronic arrangement (Clark 2011). The natural dyes used in this study are proven to have overcome these shortcomings. The resulted excellent light fastness is an attractive subject for further studies on these natural dyes.

4. CONCLUSIONS

The optimization of dyeing parameters must be carried out to generate reproducibility of the dyeing results. The results of this study show that the dyeing parameters such as dye concentration, dyeing time and temperature, and material-to-liquor ratio (MLR) in the post-mordanting process influence the quality of the resulting color on the cotton fabrics. The optimized dyeing conditions for all natural dyes used were the code A extract concentrations (0.0113 g/mL of Tegeran; 0.0115 g/mL of Merbau; 0.0204 g/mL of Jambal; and 0.0582 g/mL of Jolawe), dyeing at 28°C, dyeing time of 30 minutes, and MLR of 1:30.

NOTATIONS

1. K = light absorption coefficient

Dyes	Optimized parameter	Optimized condition	Fixative	Color coordinates		Color coordinates		Washing fastness	Light fastness
				L*	a*	b*			
Tegeran	[Dye]	0.113 g/mL	Alum	64.63	14.71	45.49	2.99	3	4–5
	Temperature	1.38							
	Time	1.86	Lime	67.62	11.84	25.71	0.87	3	4–5
	MLR*	1:30	Iron (II) sulfate	32.54	6.68	20.02	5.56	3	4–5
Merbau	[Dye]	0.012 g/mL	Alum	61.08	11.64	19.56	2.32	4–5	4–5
	Temperature	28°C							
	Time	30 min	Lime	60.88	14.27	22.55	1.24	3–4	4
	MLR*	1:30	Iron (II) sulfate	42.21	8.77	14.51	3.60	4	4–5
Tingi	[Dye]	0.020 g/mL	Alum	49.61	23.48	36.34	2.07	4	4–5
	Temperature	28°C							
	Time	30 min	Lime	39.41	26.21	29.53	3.50	4	4
	MLR*	1:30	Iron (II) sulfate	29.73	6.07	18.63	4.27	4	4
Jambal	[Dye]	0.010 g/mL	Alum	45.47	24.09	35.30	3.44	3–4	4–5
	Temperature	28°C							
	Time	30 min	Lime	42.60	25.36	28.68	3.02	4	4
	MLR*	1:30	Iron(II) sulfate	33.74	9.41	14.58	3.33	4	4
Jolawe	[Dye]	0.058 g/mL	Alum	74.57	3.15	17.27	0.06	4	5
	Temperature	28°C							
	Time	30 min	Lime	67.88	4.84	18.75	0.16	4–5	4–5
	MLR*	1:30	Iron (II) sulfate	35.48	1.23	0.89	2.28	4	4–5

TABLE 6. The dye concentration of the extracts.

*Post-mordanting process.

- 2. S = light scattering coefficient
- 3. R = percentage of reflectance

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