Colorimetric properties of batik fabrics colored using gambier liquid waste

F Failisnur1, S Sofyan1 and S Silfia1

1Institution of Research and Standardization of Industry - Padang
Jl. Raya LIK No. 23 Ulu Gadut, Padang, Indonesia

*failisnur@gmail.com

Abstract. The process of batik and colouring on cotton and silk using gambier liquid waste, through the wax removal process in hot temperature affects the color characteristic and fastness properties of the fabric. The purpose was to study the effects of the batik process on the color and fastness properties the dyed fabrics. The study was conducted with the use of soda ash as the wax removal, and CaO, FeSO₄, Al₂(SO₄)₃ as mordant with post mordanting method on cotton and silk fabrics. The results showed that the batik process affects the can provide varying color strength (3.7 - 16.7) with color coordinates in the range of yellowish (7.7 - 19.9) to reddish (5.7 - 24.8) and fairly good color fastness to excellent against washing, rubbing and light effects. The highest color strength was obtained when using mordant FeSO₄ which was applied to silk.

1. Introduction
Gambir (Uncaria gambier Roxb.) is cultivated plants and locally in West Sumatera Province and several other areas such as Aceh, North Sumatera and South Sumatera. The leaves and twigs of extraction of young gambier called raw gambier. This extraction process is carried out through boiling, pressing, settling, draining, molding and drying. Raw gambier is a semi-finished product and it requires a further process to be applied for various purposes. This product furthermore is reprocessed into a multipurpose finished product.

Catechins and tannins are the main compounds in gambier with ± 32.12% and 51.14% content [1]. Catechin are also generally used as raw materials for medicines, cosmetics and food [2,3], while tannins are widely used for textile dyes [4-6], food coloring [7] and tanners [8,9].

The process of raw gambier produces a by product in the form of about 25 percent liquid waste and it is brown to blackish brown. The waste is disposed around the production area and it has not been utilized. The waste has an acidic odor and their pH are 3-4. It has also the potential to pollute the surrounding [10,11]. In the open air, the waste is damaged by aerobic bacteria identified as black Aspergillus niger, green Aspergillus fumigatus and Penicilium sp [12].

Liquid waste from gambier processing contains tannin and there are quite high up to 52.14%, and they also potential to be used as textile dyes [6]. In weaving yarn coloring, gambier liquid waste provides a higher intensity compared to raw gambier [5].

Some research on the use of gambier coloring for batik have also been carried out [13-15]. The coloring process produces a fairly high color intensity, good color fastness, and higher tear strength
compared to those without coloring [15]. However, studies of inhibitory effects on several potential microbes and the tensile strength of batik fabrics colored using gambier have not been found.

Batik is a product of Indonesia's unique cultural heritage and is in demand by various groups to foreign countries. The trend in the use of batik has so far increased sharply supported by the Indonesian government's policy regarding the use of batik as formal outfit in government. This product is also often used at receptions, businesses and other important events. The dyes used are mostly sourced from synthetic and are suspected to be toxic and carcinogenic [16,17]. Development of eco-green batik products with natural dyes is feasible to be developed because it has high economic value, has a soft color, is more natural, is health-friendly for the wearer, and is environmentally friendly [18].

2. Methodology

2.1. Material and Tools

The material used in the study was gambier liquid waste from Nagari Siguntur, Pesisir Selatan Regency, West Sumatra. The mordant such as CaO, FeSO4, Al2 (SO4) 3, soda ash and starch are produced by Bratacho Chemical, cotton fabrics is obtained from PT. Primissima, Yogyakarta, silk fabrics, and batik wax are from IKM, Yogyakarta). The chemicals for tannin testing are folin ciocalteu reagents and ammonium tungstate from Merck.

The equipment includes a 400 mesh filter, cruiser, homogenizer, oven, gray scale, staining scale, UV-Visible spectrophotometer (Premiere colorsan SS 6200, CIELAB (1976) / D65, Navi Mumbai, India)

2.2. Research procedure

2.2.1. Preparation of gambier liquid waste. Gambier liquid waste is filtered to remove residues and other impure material. Waste is heated at 90-100°C for ± 1 hour. The purpose of heating is to kill aerobic microbes that cause damage and odor. The amount of waste needed is in accordance with the dyeing plot at a ratio of 1:20. For 1 kg of fabric need20 liters of gambier waste.

2.2.2. Waxing (the proces of giving batik motifs). In the batik production process, the first step is pasting a color resist in the form of wax on a fabric sheet. The purpose of waxing is to create a motif using wax, therefore the color cannot penetrate the fabric within the motif. There are several method to include wax on the sheet such as by hand using canting (written batik), stamped using metal or copper stamp (stamped batik), a combination of written and stamped (combination batik) and by using screen printing in factory printing machine called printed batik [19].

The community considers the written batik has a higher value than stamped batik because the manual in the written batik requires high precision and persistence. The process takes time and more complicated, therefore the selling price is relatively expensive, depending on the complexity of the manufacturing process. Batik wax consists of materials including Resina colophonium, Shorea javanica, paraffin, animal fat, kote and micro wax. Resina colophonium, Shorea javanica, micro wax and paraffin as the basic wax-making materials are difficult to remove without hot temperature, therefore in releasing the wax, it requires enough heat [20].

2.2.3. Fabric coloring. Silk and cotton patterned with batik wax are then soaked in water and dried. In the dry state, the fabrics are dipped in a gambier waste solution for ± 15 minutes and it further dried. This process is repeated up to three times to obtain the strong color and more evenly distributed

2.2.4. Post mordanting. The mordant solution making. Mordant are chemicals substance in the form of metal salts and it improve the dyeing performance. The mecanism of its action is through bridging the reaction between the dyestuff and the textile fiber to the complexity of the dyestuff-mordant-textile fibers. In addition the mordant increases the intensity and fastness colors from the result of dyeing.
The making of mordant CaO 50 g/L, FeSO₄ 30 g/L and Al₂(SO₄)₃ 70 g/L is adjusted to the amount of water and weight of the fabric to be mordanted. The amount of mordant solution used to soak batik fabric is at a ratio of 1:2. Mordant substances are dissolved with clean water and homogenized until all ingredients are dissolved. The solution is left for ± 24 hours and filtered. The filtrate is taken as a mordant solution, while insoluble precipitate is removed.

**Post mordanting process.** The post mordanting method was chosen in this coloring because it gives a higher color intensity compared to the pre and simultaneous mordanting [4]. Dyed Fabric with gambier color is carried out the mordanting process to evoke the color by dipping in mordant solution. This process is called fixation. To increase color evenness, Soaking in mordant solution for ± 15 minutes Is needed

2.2.5. **Wax Removal.** The wax removal process aims to remove the wax layer on the batik therefore the motifs and colors are clearly visible. The wax removal and sticking of color are two distinguish process from other natural coloring. In this process, wax removal used can dissolve wax on batik. The materials commonly used are soda ash (Na₂CO₃), sodium silicate (Na₂SiO₃) a and starch, because it is able to dissolve the wax in a hot temperature. Wax removal with soda ash is carried out by dissolving 500 grams of soda ash in 30 L of water. Furthermore, it is heated to boiling and put the fabric into the solution until the wax is completely dissolved. Fabric without wax is then washed thoroughly

2.3. **Analysis**

2.3.1. **Analysis of fabric dyed.** The colored batik was observed for its antimicrobial inhibition against potential bacteria Escherichia coli and Staphylococcus aureus using the measurement of the clear zone formed. The fabric Tensile strength after the coloring process refers to Indonesian standard of SNI 08-0276-2009. The the color intensity (K / S) and the color coordinates of CIE L*, a*, b* are measured from the reflectance λ_max on each color using the Spectrophotometer premcan colorscan SS 6200 on the D65 illuminance, the reflectance results are converted to the Kubelka-Munk table based on equation 1).

The test was carried out at Bandung Polytechnic of Textile Technology. Lightness (L *) or color brightness was at values of 0-100 (100 = white, 0 = black), a * reddish (+) and greenish (-), b * yellowish (+) and bluish (-).

\[
\frac{K}{S} = \frac{(1 - R)^2}{2R}
\]

(1)

Note: K is the absorption coefficient (absorbed light); S is the spread light coefficient; R is the percent value of the reflectance (λmax)

2.3.2. **Color fastness of fabric dyed.** Evaluation of color fastness on dyeing fabric is carried out on washing, rubbing and light referring to BSN, 2010: SNI ISO 105-C 06), (BSN, 2008: SNI 0288-2008) and (BSN, 2010: SNI ISO 105- B01.

3. **Result and Discussion**

3.1 **Wax Removal Process**

This process aims to dissolve batik wax using removal material in hot surrounding. After the wax is removed, the batik motif can be seen. Wax removal affect the fastness and degradation colors of batik. Based on observations, it can be seen that the use of Na₂CO₃ wax removal material on average gives a stronger color intensity compared to before the wax removal process (Table 1). In the type of FeSO₄ mordant, the process gives color changes to be darker, Al₂(SO₄)₃ mordant changes color from yellow to reddish brown. This is due to the significant influence of the alkaline Na₂CO₃, therefore the
coloring with gambier becomes reddish. However, in the use of CaO mordant, the color intensity tends to decrease, perhaps the alkaline nature of Na$_2$CO$_3$ is lower than the CaO metal salts. Based on the research results, it was found that the wax removal process using Na$_2$CO$_3$ at 90°C for 10 minutes produced dissolved wax of about 96.2%. According to [20], the wax removal process is successful if all the wax can dissolve and does not affect the color and strength of the fabric. Based on the research of [19], the average release of wax in the removal process with sodium silicate and Na$_2$CO$_3$ at 80°C for 5 minutes and without wax removal material was 83.83%, 84.04%, and around 69.51%, respectively. Furthermore, [20] stated that by adding the time became 15 minutes with sodium silicate at 80-90°C, there was an increase in the percentage of dissolved wax to 97.68%.

Table 1. Effect of the wax removal process on the results of batik colored using gambier dyes

| Fabric Type | Treatment            | CaO       | FeSO$_4$ | Al$_2$(SO$_4$)$_3$ |
|-------------|----------------------|-----------|----------|-------------------|
|             | Before Wax Removal   | Before    | Before   | Before            |
| Cotton      | After Wax Removal    | After     | After    | After             |
| Silk        | Before Wax Removal   | Before    | Before   | Before            |
|             | After Wax Removal    | After     | After    | After             |

3.2 Photometric characteristics of coloring results

The color strength and coordinate values of batik dyed with gambier liquid waste are shown in Table 2. All treatments using mordant type show higher color intensity (K/S) on silk batik than cotton. Silk fabric consists of protein fibers containing the COOH and NH$_2$ groups which are amphilic. In certain circumstances, this group can automaton to produce a highly reactive amide (NH$_3$+) group (lack of electrons). Meanwhile, natural dyes are rich in electrons (OH-). This condition causes these two compounds to be easy to react, therefore dyes are widely absorbed into the fiber [1].

Based on the type of mordant used, the highest color strength was found in FeSO$_4$ and followed by CaO and Al$_2$(SO$_4$)$_3$. Among the three mordants, the use of CaO gives a more reddish color direction than other mordans. The resulting color direction can be seen its position in the color space coordinates of the CIE Lab system. In addition to bridging the bond between the dye and fiber, the use of mordant can also change the color of certain dyes. Different mordants used in the same dye can change the final color of the dyed fiber [21].

Table 2. The color strength and difference values of cotton and silk batik fabrics colored with gambier liquid waste

| Fabric Type | Mordant | K/S   | L*    | a*    | b*    |
|-------------|---------|-------|-------|-------|-------|
| Cotton      | CaO     | 8.995 | 45,516| 18,708| 24,883|
The colorimetric properties of dyed fabrics such as lightness (L*), redness-greenness (a*), yellowness-blueness (b*), and color strength (K/S) are highly dependent on the chemical properties of mordant and fiber, as well as the ability of metal ions to form complex between dyes and fibers [22]. Color coordinate values will distinguish the quality of the color brightness, such as red, pink (light) or dark red (dark). The commonly used levels are nine levels starting from the brightest level which is white, then gray to the darkest level which is black. White has the highest lightness while black has the lowest lightness.

3.3 Color fastness
Color fastness evaluation is based on the color change of the dyed fabric or the desecration in the accompanying white fabric. The low color fastness value is due to the lack of strength of complexity bond formed by the metal mordant between the dye and fiber. Color fastness tests were carried out on washing, rubbing and light with the results as in Table 3.

Table 3. The color fastness of cotton and silk batik fabrics with gambier liquid waste dyes on washing, rubbing and light

| Fabric Type | Mordant  | Washing Color change | Color desecration | Rubbing | Light |
|-------------|----------|----------------------|-------------------|---------|-------|
| Cotton      | CaO      | 4                    | 4-5               | 4-5     | 4     |
|             | FeSO₄    | 3-4                  | 4-5               | 4       | 4-5   |
|             | Al₂(SO₄)₃| 3-4                  | 4-5               | 4-5     | 4-5   |
| Silk        | CaO      | 4                    | 4-5               | 4-5     | 4     |
|             | FeSO₄    | 3-4                  | 4-5               | 4-5     | 4-5   |
|             | Al₂(SO₄)₃| 4                    | 4-5               | 4-5     | 4     |

The color fastness value on rubbing and light is generally good to very good (4-5). It means that there is no significant color change in all treatments. Whereas for washing, there is no color desecration on other fabrics (4-5), but there is slight color change in the use of FeSO₄ mordant both on cotton and silk batik (3-4). Overall, coloring of cotton and silk batik using gambier liquid waste as a dye provides good average color fastness (4).

Nearly 50% of natural dyes which are derivatives of flavonoid compounds such as tannins have low color fastness to light [23]. The evaluation results of the color fastness to light on the coloring of cotton fabric using gambier obtained an average value of less (2-3) [4]. However, the wax removal process in making batik can increase the color fastness value to light (4-5).

4. Conclusion
The process of wax removal on batik fabrics and the use of mordant when dyeing with gambier liquid waste gave a different color direction and color strength. The highest color strength was obtained when mordant FeSO₄ was applied to silk fabric. The color fastness of both cotton and silk batik fabrics did not make a significant effects to the process of washing, rubbing, and light exposure.
Acknowledgments
The authors are grateful to the Institution of Research and Standardization of Industry-Padang and Ministry of Industry for funding and facilities assistance during the research. The authors are also grateful to Marlusi and Sulastri for their assistance and cooperation during the research.

References
[1] F. Failisnur and S. Sofyan, “Karakteristik kain batik hasil pewarnaan menggunakan pewarna alam gambir (Uncaria Gambir Roxb),” Pros. Semin. Nas. Has. Litbangyasa Ind. II, vol. 2, no. 2, pp. 228–235, 2019.
[2] A. Lukas, “Formulasi obat kumur gambir dengan tambahan peppermint dan minyak cengkeh,” J. Din. Penelit. Ind., vol. 23, no. 2, pp. 67–76, 2012.
[3] G. Yeni, K. Syamsu, O. Suparno, E. Mardliyati, and H. Muchtar, “Repeated extraction process of raw gambiers (Uncaria gambier Robx.) for the catechin production as an antioxidant,” Int. J. Appl. Eng. Res., vol. 9, no. 24, pp. 24565–24578, 2014.
[4] F. Failisnur, S. Sofyan, A. Kasim, and T. Angraini, “Study of cotton fabric dyeing process with some mordant methods by using gambier (Uncaria gambir Roxb) extract,” Int. J. Adv. Sci. Eng. Inf. Technol., vol. 8, no. 4, pp. 1098–1104, 2018.
[5] Failisnur and Sofyan, “Pengaruh suhu dan lama pencelupan benang katun pada pewarnaan alami dengan ekstrak gambir (Uncaria gambir Roxb),” J. Litbang Ind., vol. 6, no. 1, pp. 25–37, 2016.
[6] S. Sofyan and F. Failisnur, “Reuse of liquid waste from textile dyeing with natural dyes gambier (Uncaria gambir Roxb.) for cotton yarn dyeing,” ARPN J. Eng. Appl. Sci., vol. 12, no. 18, pp. 5313–5318, 2017.
[7] F. Firdausni, G. Yeni, F. Failisnur, and K. Kamsina, “Karakteristik pewarna alam gambir (Uncaria gambir Roxb) untuk produk pangan,” J. Litbang Ind., vol. 9, no. 2, pp. 89–96, Dec. 2019.
[8] A. Arbain, S. Sy, and A. Kasim, “Perilaku krom dalam limbah cair penyamakan kombinasi krom-gambir dan krom-mimosa pada penyamakan kulit,” J. Litbang Ind., vol. 4, no. 1, pp. 59–66, 2014.
[9] A. Kasim, A. Asben, and S. Mutir, “Kajian kualitas gambir dan hubungannya dengan karakteristik kulit tersamak,” Maj. Kulit, Karet, dan Plast., vol. 31, no. 1, pp. 55–64, 2015.
[10] S. Sofyan, F. Failisnur, and S. Salmariza, “Pengaruh Perlakuan Limbah dan Jenis Mordan Kapur, Tawas, dan Tunjung Terhadap Mutu Pewarnaan Kain Sutera dan Katun Menggunakan Limbah Cair Gambir (Uncaria gambir Roxb),” J. Litbang Ind., vol. 5, no. 2, p. 79, Dec. 2015.
[11] Failisnur and G. Yeni, “Stabilization of gambier process wastewater and its application as silk dye,” Biopropal Ind., vol. 4, no. 1, pp. 7–16, 2013.
[12] H. Muchtar, Kamsina, and I. T. Anova, “The effect of storage condition on mold growth in gambier,” J. Din. Penelit. Ind., vol. 22, no. 2, pp. 36–43, 2011.
[13] Failisnur, Sofyan, and W. Hermianti, “Pemanfaatan limbah cair pengempaan gambir untuk pewarnaan kain batik,” J. Litbang Ind., vol. 7, no. 1, pp. 19–28, 2017.
[14] V. Atika, Farida, and T. Pujilestari, “Kualitas pewarnaan ekstrak gambir pada batik sutera,” Din. Kerajinan dan Batik, no. 7, pp. 25–32, 2016.
[15] S. Sofyan and F. Failisnur, “Gambier (Uncaria gambir Roxb) sebagai pewarna alam kain batik sutera, katun dan rayon,” J. Litbang Ind., vol. 6, no. 2, pp. 89–98, 2016.
[16] S. Islam, B. S. Butola, and A. Roy, “Chitosan polysaccharide as a renewable functional agent to develop antibacterial, antioxidant activity and colourful shades on wool dyed with tea extract polyphenols,” Int. J. Biol. Macromol., vol. 120, pp. 1999–2006, Dec. 2018.
[17] M. Yusuf, M. Shabbir, and F. Mohammad, “Natural colorants: Historical, processing and sustainable prospects,” Nat. Products Bioprospect., vol. 7, no. 1, pp. 123–145, 2017.
[18] T. Rossi, P. M. S. Silva, L. F. De Moura, M. C. Araújo, J. O. Brito, and H. S. Freeman, “Waste from eucalyptus wood steaming as a natural dye source for textile fibers,” J. Clean. Prod.,
2016.

[19] T. Haryanto and D. Suheranto, “Analisa pengaruh soda abu terhadap pelorodan lilin batik dan kekuatan tarik kain batik sutera,” Din. Kerajinan dan Batik, vol. 25, pp. 17–26, 2008.

[20] D. Suheranto, “Penggunaan natrium silikat pada proses pelorodan batik terhadap pelepasan lilin dan kekuatan tarik kain,” pp. 1–7, 2015.

[21] A. Manhita et al., “Enlightening the influence of mordant, dyeing technique and photodegradation on the colour hue of textiles dyed with madder - A chromatographic and spectrometric approach,” Microchem. J., vol. 98, no. 1, pp. 82–90, 2011.

[22] B. Tang, Y. Yao, W. Chen, X. Chen, F. Zou, and X. Wang, “Kinetics of dyeing natural protein fibers with silver nanoparticles,” Dye. Pigment., vol. 148, pp. 224–235, 2018.

[23] D. Cristea and G. Vilarem, “Improving light fastness of natural dyes on cotton yarn,” Dye. Pigment., vol. 70, no. 3, pp. 238–245, 2006.