Effect of fixator on color performance of bark extract from three tropical wetland species for fabric dye

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Abstract. Fixation is a stage in dyeing fabric or batik fabric with natural dyes to obtain the desired color and bind the color so that it does not fade quickly due to sweat, washing, sunray, and rubbing. Three fixators used as color binders were alum (Al₂(SO₄)₃), lime (CaCO₃), and tunjung (FeSO₄). Each material showed a different color on each extract. This activity aimed to observe the color performance of bark extracts from three wetland species applied to batik fabric with 1 or 2 combinations of fixators. The results showed the color performance of batik fabric depended on the bark of tree species and the kind of fixator. The performance of bark extract with fixators produced darker and sharper colors than without the use of fixators. The application of lime on Rhizophora apiculata Bl. bark extract produced dark moderate orange color, while the combination of lime and tunjung produced very dark orange (Brown tone). The combination of lime and tunjung on Acacia mangium Willd and the Terminalia catappa L. barks gave a dark moderate orange color with different intensities. In contrast, the combination of alum and tunjung on Terminalia catappa L. bark extract produced a very dark greyish orange color.

Keywords: bark extracts, natural dyes, fixator, color diversity.

1. Introduction

Indonesia has been recognized worldwide as the owner of batik patent since 2 October 2009 by the United Nations Educational, Scientific, and Cultural Organization (UNESCO). Export of batik and woven fabrics to overseas countries already reached the US $ 58.6 million in 2019 [1]. Specifically, batik products exports increased 20% in 2020 compared to the exports in 2019, with significant marketing to Japan, the United States, and Europe [2]. Although the demand for batik from Indonesia is increasing, the materials for the dye are still imported from foreign countries. Imported batik and weaving dyes are generally cellulose synthetic dyes [3]. These synthetic dyes are demanded a lot by the batik crafters, because their color is durable and resistant against leaching and rubbing/ironing actions. In their uses, those pigments should be added with sodium hydroxide (NaOH) solution in order to become soluble in water [4]. The NaOH-added synthetic dyes can be carcinogenic, thereby endangering human health and pollute the environments.

Indonesia is endowed with huge potency as the sources of natural renewable dyes, either from forest plants or from plantation estates. As many 71 of 4000 tree species as those in natural forests [5], which several researchers in fact have investigated could afford remarkable potency of natural dyes for batik fabrics [6]. The superiority of natural dyes compared to synthetic dyes besides being renewable...
is that it can create specific and smooth colors, which are unable or difficult to imitate by synthetic dyes. Nowadays, batik market segments, which use natural dyes either in domestic or in overseas countries, are still limited. This is possibly hindered by the prices of natural dyes, which are still expensive, because the raw materials for natural dyes have not been produced in mass quantities or industrial scales.

The fabrics that will be colored with natural dyes necessitate the binding agents or called fixators. The fixation of fabrics after being dyed represents one of the processes to bind the pigments from plants (natural dyes), so that the pigments are firmly attached to the fabrics and therefore will not easily leach out due to the rubbing, washing, and drying under a direct sun ray [7]. Besides functioning as color binding, the fixators could also distribute the color evenly throughout the fabric surface and intensify the color of dyed fabrics [8]. Three kinds of fixators commonly used by the traditional batik crafters which use natural dyes are alum (Al$_2$[SO$_4$]$_3$), lime (CaCO$_3$), and tunjung (FeSO$_4$), where each fixator exhibits its own color-altering performance differently.

The hindrances frequently encountered by the batik crafters, especially those residing at coastal areas, are mainly the limited color diversities. The use of one kind or more of fixators in combination expectedly could add the color variation to natural dyes. This paper presents the results of the application of natural dyes of bark extracts from *Acacia mangium*, *Rhizophora apiculata*, and *Terminalia catappa* with two combinations of fixators on batik patterned fabric, where those three plants come from wetland species.

2. Materials and Methods

2.1. Materials

The materials as natural dyes for batik fabrics in this research were extracted from three barks species, i.e. *A. mangium* Willd, *R. apiculata* Bl., and *T. catappa* L., each originated from wetland species. Meanwhile, the solvent used for the bark extracts was water. Kind of fabric that would be dyed with natural dyes was mori or primisima. The fixators (dye-binding agents) consisted of three kinds, i.e. alum (Al$_2$[SO$_4$]$_3$), lime (CaCO$_3$), and tunjung (FeSO$_4$). The chemical agent used to wash the fabrics (before being dyed with bark extracts) was tipol. The chemical used to cover the particular parts of the fabrics so that those parts could not be touched with the extracts was malam (kind of wax). Caustic soda (NaOH) was used to leach out malam.

The tools which were involved in this research, among others metal pan, stove, container for immersing the fabrics (to be colored) in the dyeing solution as well as for leaching the malam, gawangan used to hook the fabric sheets in order to facilitate more easily in imparting the batik patterns on the fabric, canting used to make batik patterns (e.g. writings, scratches, and carvings) with wax on the fabric, weighing scales, and color-measuring instrument.

2.2. Methods

2.2.1. Bark extraction

This activity was an application of the results of the previous best research, whereby the extraction method used water as a solvent with barks to water ratio of 1: 4 (w/v). The barks were then boiled for 2 hours at 70 °C temperature [8]. Afterward, that the solution was cooled and then filtered.

2.2.2. Imparting of batik patterns and dyeing of batik-patterned fabrics

The mori fabrics before being imparted with the batik motive should be washed in the water which was mixed with 1% tipol (v/v). Afterward, the fabrics were rinsed and then dried. Batik motives on fabrics depend on the taste. The patterned fabrics were then covered with malam by canting, so that the part of the fabric that was not being colored was not exposed to a dye solution at the time of dyeing. The dyeing of the already batik-patterned fabrics was performed by immersing the fabrics in the bark extract solution with 5-6 times of immersion, depending on the strength of the desired color, whereby each immersion took about 7 minutes.
2.2.3. Dye fixation
The fixation process could be performed by immersing the color-imparted fabrics for 15 minutes in hot water, to which was already added 2-3% fixator (w/v). If using two types of fixators, then after doing the first fixation, the fabric must be soaked again in a dye solution (bark extract), then re-fixated again using another type of fixator.

2.2.4. Leaching of fabrics
After the dyeing process and fixation treatment, batik fabric was soaked in hot water that was added with a small amount of caustic soda/NaOH to dissolve the malam. After that, the batik fabrics should be dried.

2.3. Color test and color quality assessment
The testing of the color difference in each type of extract and in one only or two combinations of fixators was conducted using a color-measuring instrument [9]. On the color gauge was obtained color values “L”, “a”, and “b”. The value “L” indicates the brightness of the color from 0 (black) to 100 (white). The value “a” indicates a color from 0 (green) to 80 (red) and the value “b” indicates a color from 0 (blue) to 70 (yellow). The determination of the color values "L, a, and b" for each treatment was performed on 5 location points at each of the fabric sheets. The color change/difference (ΔE) between two fixator treatments could be calculated as a mathematic function of ΔL, Δa, and Δb, in the following formula:

\[
\Delta E = f(\Delta L, \Delta a, \Delta b) = \left( [\Delta L]^2 + [\Delta a]^2 + [\Delta b]^2 \right)^{0.5}
\]

where \(\Delta E\) = color change value, \(\Delta L\) = difference in L values between the compared samples (dyed fabrics with 1 or 2 fixators vs dyed fabrics without fixators or control), \(\Delta a\) = difference in “a” value between the compared samples (the same); and \(\Delta b\) = difference in “b” value between the compared samples (the same).

From the obtained \(\Delta E\) values, the classification of color change and the change criteria, could then be decided according the related criteria (Table 1)

| Class | Color change (\(\Delta E\)) values | Criteria |
|-------|-----------------------------------|----------|
| I     | \(\Delta E < 0.2\)              | Invisible change |
| II    | \(0.2 < \Delta E < 2.0\)         | Very small change |
| III   | \(2.0 < \Delta E < 3.0\)         | Small changes (color changes visible by high-quality filter) |
| IV    | \(3.0 < \Delta E < 6.0\)         | Medium (color changes visible by high-quality filter) |
| V     | \(6.0 < \Delta E < 12.0\)        | Big (distinct color changes) |
| VI    | \(\Delta E > 12.0\)              | Different color |

Source: [9]

3. Results and Discussion
3.1. Results
Table 2 shows the color parameter of dyed and fixed batik cloth, and Table 3 shows the Application of natural coloring on batik cloth.
### Table 2. Color parameter of dyed and fixed batik cloth.

| Natural dyes kind | Fixator material       | Quantitative colour system | Colour change ($\Delta E$) | Implication $^3$ |
|-------------------|------------------------|----------------------------|-----------------------------|------------------|
| A. mangium        | Control (Unfixation)   | L  55.14                   | a  10.44                   | b  23.11         | 48.00 DC          |
|                   | Lime                   | 46.23 (0.127)              | 16.77 (0.052)              | 33.93 (0.098)    | 45.60 DC          |
|                   | Lime + tunjung         | 41.33 (0.084)              | 16.01 (0.028)              | 21.59 (0.088)    | 48.43 DC          |
| T. catappa        | Control (Unfixation)   | 46.68                      | 14.06                      | 36.51            | 48.43 DC          |
|                   | Lime                   | 42.77 (0.113)              | 13.29 (0.098)              | 26.98 (0.085)    | 44.07 DC          |
|                   | Lime + tunjung         | 42.46 (0.163)              | 8.20 (0.075)               | 24.27 (0.072)    | 40.72 DC          |
| T. catappa        | Control (Unfixation)   | 42.77                      | 13.29                      | 26.98            | 40.72 DC          |
|                   | Alum                   | 42.81 (0.168)              | 12.44 (0.085)              | 29.12 (0.080)    | 49.11 DC          |
|                   | Alum + Tunjung         | 40.34 (0.153)              | 1.60 (0.080)               | 14.71 (0.072)    | 41.91 DC          |
| R. apiculata      | Control (Unfixation)   | 49.23                      | 14.28                      | 26.98            | 56.94 DC          |
|                   | Lime                   | 37.93 (0.180)              | 19.01 (0.099)              | 28.76 (0.091)    | 52.52 DC          |
|                   | Lime + Tunjung         | 26.11 (0.182)              | 6.44 (0.097)               | 18.58 (0.087)    | 52.52 DC          |

Remarks: Average from 5 location points; $^1$Refer to equation (1); $^2$Refer to Table 1 (Values in parentheses are standard deviations); DC = Different color

#### 3.2. Discussion

*Acacia mangium* (mangium), *Rhizophora apiculata* (bakau minyak), and *Terminalia catappa* (ketapang), are three species of plants producing natural dyes, which are famous enough especially for the coastal-motive batik crafters. Based on the result origins [8], the plant parts of the three species contained tannin enormously. The greatest percentage of the tannin is contained in the bark parts. Inside the barks, there are parenchyma tissues that function as food reserves, physiological activities, and specific chemical synthesis, including the tannin [10].

The “L” values of the batik fabrics colored with natural dyes without fixation were more significantly greater than those of the fabrics which were colored and afterwards being performed the fixative treatment (Table 2). This indicated that the use of fixators could intensify the color brightness of the batik fabrics, which colored with natural dyes from bark extracts. This was because the fixators could serve as a bridging intermediary between the chemicals contained in the dyes and the fabric fibers [11]. In the fixation process, the fabric fibers would attract the dye molecules and then bind them strongly such that the dye molecules were firmly adhered to the fabrics. This situation rendered the affinity of the dyeing solution (bark extract liquid) toward the fabric fibers to increase. Fabric fibers which are compatible with the natural dyes should be the cellulose fibers, such as cotton fibers and artificial-silk fibers. The lowest “L” values were exhibited by the batik fabrics colored with the natural dye from *R. apiculata* bark extracts and further treated with the lime and tunjung fixators. This occurrence indicated that the fabric colors had changed to become darker. In this case, the tunjung fixators allegedly contributed a lot toward darker fabric colors. The tunjung fixator generated green or brownish green or blackish colors due to iron (Fe+2) ions inside. Further, the tannin in the bark extracts reacted with Fe+2 ions, creating ferro tannate compounds, a complex salt, which caused the batik fabrics (immersed in bark extract solution and then treated with tunjung fixator) to exhibit blackish colors [12]. Meanwhile, due to the presence of Al+3 ions, the alum fixators aptly created the fabric colors lighter than the lime fixators, which contain Ca+2 ions (Table 2).
Table 3. Application of natural coloring on batik cloth.

| Natural dyes kind | Non-fixator\(^1\) (Control) | Fixator material | Batik pattern | Color description                          | Color code | Color composition (%) | Red    | Green   | Blue   |
|-------------------|-------------------------------|------------------|---------------|--------------------------------------------|------------|-----------------------|--------|---------|--------|
| A. mangium        | - Lime + tunjung              | - Lime + tunjung | Dark moderate orange | #8557f | 52.2 | 34.1 | 24.7 |
| T. catappa        | - Lime + tunjung              | - Alum + tunjung | Very dark grayish orange | #7d5e3c | 49 | 36.9 | 23.5 |
| R. apiculata      | - Lime                        | - Lime + tunjung | Very dark orange (Brown tone) | #9f6a41 | 62.4 | 41.6 | 25.5 |

Remark: Value for fabric color without motives\(^1\) [8]
The color change (ΔE) values of the dyed batik fabrics, for all treatment combinations which incorporated either any one kind of the fixator or either any of the combined two kinds of fixators (as disclosed in Table 2), revealed that the ΔE values were entirely greater than 12 (refer to Table 1). These phenomena implied that a color difference occurred in the dyed fabric at each of the different treatment combinations (e.g. the fabric dyeing without fixative treatment vs. the fabric dyeing with incorporating the fixative treatment) (Table 2). This occurrence could be very advantageous, because the manipulation of such treatment combinations could produce the batik fabrics with diverse colors by combining various fixators, such as lime with alum alone, alum with tunjung, lime with tunjung, and even alum mixed with tunjung and lime. Each mixture that involved more than one kind of fixators could generate specific colors of the dyed batik fabrics.

Of those bark extracts used without fixator treatments (controls), it appeared that the bark extracts from *R. apiculate* species created brown colors of the batik fabrics, which were stronger (darker) compared to the colors of the batik fabrics dyed with the other two kinds of bark extracts i.e. *Terminalia* sp. and *A. mangium* bark extracts (also without fixators). However, compared to the colors of the batik fabrics dyed with *Terminalia* sp. bark extracts, the fabrics dyed with *A. mangium* bark extracts created darker fabric colors (Table 3). The tannin in *A. mangium* and *R. apiculata* barks are essentially a concentrated tannin or a condensed tannin, which is characterized as a typical tannin composed mainly of flavonoid polymers, which belong to polyphenol compounds [13]. According to [14], the condensed tannin in *R. apiculata* species is present a lot at the hydrophillic groups in their corresponding barks.

After being treated with the fixators, the original basic color of batik fabrics colored with natural dyes (bark extracts) sustained the noticeable changes (Table 3). As such, the original color of the dyed fabrics (without fixators), which was previously fading, changed to become sharper and more attractive, after incorporating the fixators. This was possibly due to the particular chemical components present in the fixators which further reacted with the chemical compounds in the natural dyes, thereby enhancing the beauty and sharpness of the natural dye’s color.

The changes in the colors of dyed fabrics depended on the kinds of the incorporated fixators. The dyeing of batik fabrics with the bark extracts from *T. catappa*, followed with two fixators (lime and tunjung) brought out dark moderate orange colors (Table 3). Meanwhile, the combined use of other fixators (alum and tunjung) created very dark grayish orange colors of the fabrics. The alum imparted lighter/brighter color, causing the alum combination with tunjung fixators to create grayish colors of the fabrics. Meanwhile, the dyeing of fabrics with bark extracts from *R. apiculata*, followed with the fixation using lime and tunjung brought out the fabrics with very dark orange colors, which further aptly shifted toward brown colors (brown tone). Likewise, in the use of only one kind of fixator (lime) it created dark moderate orange of the fabrics. Although the dyeing of batik fabrics with the bark extracts from *A. mangium* and the bark extracts from *T. catappa* (followed with the combined use of lime & tunjung fixators), as well as with the bark extracts from *R. apiculata* (with the use of only lime fixator) were predominantly a red color, however, its intensity differed among those three bark types (Table 3).

Referring to the standard SNI ISO 105-C06: 2010, SNI ISO 105 x 12: 2012, and SNI ISO 105 -BO2: 2014, the result of the color fading/leaching test indicated natural dyes from *R. apiculata* bark extract was more resistant to the fading/leaching [8]. The color of *R. apiculata* bark extract was also typical, which was slightly reddish or deep brown. This distinctive color was because *R. apiculata* is very rich in tannin compounds. The tannin concentration from its dried extract was also the highest (28%) compared to extracts from *T. catappa* bark (25%) and *A. mangium* (23%) on equal treatment [8].
4. Conclusion

The immersion of batik-motive fabrics in the solution of bark extracts, without fixator treatment created the dyed fabrics with “L” values greater than the values correspondingly, but treated with the fixators. This indicated that the use of fixators aptly shifted the colors of the dyed fabrics to become darker. Bark extracts from *A. mangium* trees for the dyeing of batik fabrics, followed with the combined use of two fixators (lime & tunjung) generated dark moderate orange colors at the fabrics, with the color composition of 52.2% red, 34.1% green, and 24.7% blue. Bark extracts from *T. catappa* trees, followed by combining two fixators (alum & tunjung), brought out the dyed fabrics with very dark grayish orange colors, composed of 42% red, 36.9% green, and 27.8% blue. Meanwhile, correspondingly but with the combined use of lime & tunjung fixators, it exhibited dark moderate orange colors at the dyed fabrics with the color composition of 49% red, 36.9% green, and 23.5% blue. Bark extracts from *R. apiculate* trees, followed with the combined use of two fixators (lime & tunjung) created the dyed fabrics with very dark orange colors which aptly shifted toward brown colors (brown tone) with the color composition of 37.3% red, 20% green, and 4.3% blue. Meanwhile, correspondingly but with the use of only one kind of fixator (lime) it generated dark moderate orange colors of the fabrics with the color composition of 62.4% red, 41.6% green, and 25.5% blue.

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Author’s Contribution

All authors contributed equally to this work as the main contributor.

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