

## EXISTENCE OF ALUM ON THE EFFECTIVENESS OF EXTRACTED MIANA LEAVES ON BATIK COLORING

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### Abstract

The purpose of this study was to investigate the effect of existence of alum on batik coloring application. As a model of batik coloring agent, miana leaves extract was used. In short of the experimental procedure, extracted miana leaves were dipped into the cotton fabrics that were initially soaked into the alum solution. To understand the effects of alum on the coloring phenomena on fabrics, the process was completed with the analysis of various pH conditions. The experimental results revealed that existence of alum was effective for binding extracted miana leaves into the fabrics, confirmed using several characterization analyses. The main idea for the successful coloration is due to the existence of aluminum ion ( $Al^{3+}$ ) that is from the dissolution of alum (i.e.,  $Al^{3+}$  ions).  $Al^{3+}$  ions then easily penetrate to the deepest position in the fabric fibers. The penetrated ions react with the extracted miana leaves to form large-size complex salt. As a consequence, dye could not be removed easily from the fabric fibers. Therefore, understanding the existence of alum has the potential to be applied in the batik industry to improve the natural dye coloration.

Keywords: Alum, Batik, Miana leaves extract, Natural dye.

## 1. Introduction

Coloring batik is one of the important parameters in creating beautiful batik artworks. To color batik, it is not difficult since the process is putting only coloring agent into the fabric material. The batik coloring agent can be obtained easily in commercially available markets and stores in Indonesia, and even it can be obtained from the companies that sell batik materials and tools. One of the most used dyes for coloring agent is chemical-based dyes (known as textile dyes). Although these chemicals are largely available in various colors, problems in the waste management are found since they cannot be easily decomposed.

To against this condition, natural dyes are typically used as one of the alternative coloring agents because the material is largely available, environmental friendly, and inexpensive [1, 2]. Natural dyes are usually obtained by extracting parts of plant, such as bark, leaves, roots, fruit, seeds, and flowers that contain tannins, flavonoids, and quinonoids [3, 4]. Each plant and/or the part of plant can produce different types of colors [4]. The color produced by natural dyes is also softer, shinier, and calmer for eyes [2]. Since the natural dyes were extracted from the parts of plant, they are environmental friendly and harmless [2].

Examples of the plants and/or the parts of plant that can be used as the sources of natural dyes are rosella petals (*Hibiscus sabdariffa L*; for producing red, orange, purple and blue colors); mangosteen peel (for creating red color); teak leaf extract (for getting blue color at high pH and red colors at low pH); Kesumba seed extract (for providing yellow and red colors); and secang wood extract (*Caesalpinia Sappan L*; for giving red color) [4].

Although the natural dyes can be used for altering the use of chemicals, they have limitations in the number of colors. This makes them to be less practical [5]. One of the methods for solving the problems is using the fixation process [2]. The fixation process or commonly called the mordanting process can be done in three ways, namely pre-mordanting (before coloring), simultaneously mordanting (by mixing fixation agents with natural dyes), and post-mordanting (after coloring) [2]. Various types of fixation agents can be used, such as iron sulphate [6, 7], alum, aluminium sulphate [2], aluminium potassium sulfate, iron sulphate, copper sulphate, stannous chloride [8], natrium sulphate, magnesium sulphate, [9], and tin chloride [10].

Although above chemicals can be used for the fixation process, they must be used in the specific steps in the fixation process. According to Gupta et al. [10], increasing color sharpness with the post-mordanting method gives good results when using copper or iron sulphate, while the pre-mordanting method should be done using tin chloride or alum. Although the methods are effective for improving the number of colors gained from natural dyes, applications of the methods in the realistic conditions in Indonesia are still limited.

This study aims to investigate the effect of the existence of alum on batik coloring application with miana leaf extract (*Coleus atropurpureus [L] Benth*; as a model of tannin source). Understanding this study will be potential for further developments in batik industries in Indonesia, in which this will become the most important engineering aspect in this study.

The main idea for the use of miana leaf extract is because this part of plant is largely available in tropical countries such as Indonesia, while, until now, the use of miana leaves as the source of natural dyes is still rare. Marpaung et al. [11]

reported that the miana leaves have flavonoids, tannins, triterpenoids, steroids, and essential oils. Specifically, for tannin, this compound is the most important chemical because the phenolic hydroxyl groups of tannins can form crosslinking with various types of fibers [12]. The use of tannin as natural dyes is effective to be applied to fabric fibers that have low affinity [13].

In the experiment, to extract dyes from miana leaves, we used combinations of drying, grinding, and maceration process. Drying and grinding process is used to make the miana leaves to be easily stored and used whenever they are needed. Combinations of drying, grinding, and maceration were used to solve the current problems in the traditional batik industry. The traditional batik industry typically implemented multistep processes for extracting natural dyes. In short, the traditional batik industries get the dyes using the following procedures: soaking a certain amount of parts of plants in water for several days, which is followed by immersing the cloth into the soaked parts of plants in 1-3 days. This makes the production process less efficient.

To improve colors gained from miana leaf extract, alum is used as a fixation agent. Alum is one of the most-used fixation agents [6, 7]. In this study, alum was used in the pre-mordanting step. To understand the effect of alum on the coloring phenomena on the fabrics, the process was completed with various pH conditions. To identify the compounds in miana leaf that acts as a coloring agent, UV-Visible (UV-Vis) spectrophotometry and Fourier transform infrared (FTIR) analysis were carried out. To determine the morphology of the fabrics after going through the coloring as well as washing processes, analysis using a scanning electron microscope (SEM) was conducted. Based on the results of the analysis, tannin from extracted miana leaves is effectively binding aluminium ions ( $Al^{3+}$ ), forming large complex compounds to make the natural dyes trapped in the fabric fibers.

## 2. Experimental Method

Miana leaves (obtained from Bandung, Indonesia) were sliced into small sizes and then dried. Dried Miana leaves were ground and mashed to get powders with a specific size using our grinding apparatuses [14, 15]. The powder was then put into the maceration method with soaking time of 24 hours. The maceration was carried out using ethanol 95% as a solvent. Detailed maceration apparatuses were explained in our previous studies [16-18]. The product was filtered and then evaporated for 90 minutes with a rotary evaporator to get the extracted dyes.

The extracted dyes were diluted with solvent and put with the fabrics (obtained from Bandung, Indonesia), in which this step is named as the coloring process. Prior to adding into the coloring process, the fabrics were fixated with alum. For a standard comparison, some fabrics are put into the coloring process without alum fixation. In the case of fabrics that fixated by alum, fabrics were soaked in alum solution (1 wt%) for 10 minutes. Next, the soaked fabrics were dried in room temperature overnight. Fabrics with alum fixation and without alum fixations were then soaked in extracted miana leaves for 1, 2, and 3 hours.

To determine the effect of immersion time on the structural properties of the chemical compounds in the fabrics, an FTIR (FTIR-4600, Jasco Corp., Japan) was used.

To investigate the effects of pH and the type of solvent on the chemical composition as well as the color produced from Miana leaf extract, miana leaf

extract was dissolved in different pH values (from 3 to 7). Then, the dissolved extracts were tested using a UV Visible spectrophotometry (Vis mini 1240, Shimadzu Corp, Japan). In addition, for gaining solutions with pH of 3, 4, and 7, acetic acid (Bratachem, Indonesia) were used. For obtaining the pH of 5, ethanol (95%, Bratachem, Indonesia) was used.

To determine the morphology of the fabric fiber before and after the coloring process, samples with various soaking processes were tested using a SEM (JSM-6360LA; JEOL Ltd., Japan).

### **3. Results and Discussion**

Figure 1 presents the photograph image of samples after additional extracted miana leaves. The figure showed different colors for the fabrics. Initial fabrics were white, whereas after additional extracted miana leaves resulted in the yellowish green colored fabrics.

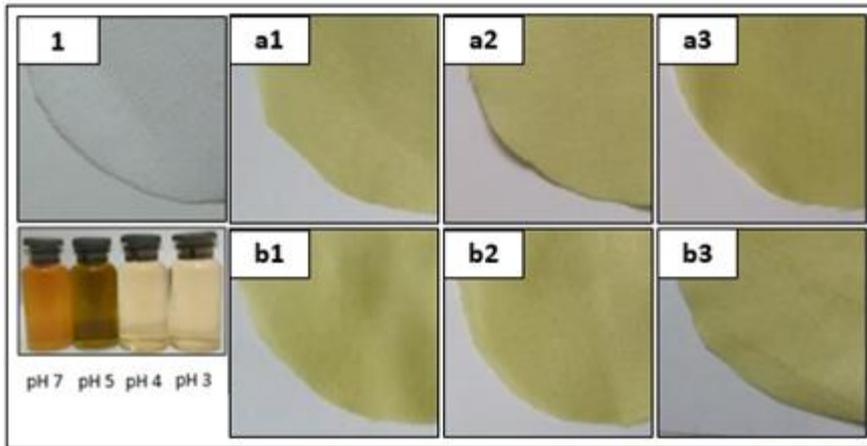
The sharpness of the color was affected by several factors as shown in Fig. 1. The factors investigated in this study were the existence of alum fixation and soaking time of fabrics. Figure 1(1) shows the initial fabrics. Figures 1(a1), (a2), and (a3) are the fabrics with the alum fixation process that were soaked at 1, 2, and 3 hours, respectively. Figures 1(b1), (b2), and (b3) are the fabrics without alum fixation process that were soaked at 1, 2, and 3 hours.

Experimental results showed that soaking for 3 hours is the most optimum condition for coloring process. The different colors obtained with the process with 2 and 3 hours were not significant, and further longer soaking processes did not bring more effects on the coloring process. For this reason, we limited the observation of soaking time to 3 hours. In addition, fabrics that were fixated in the pre-mordanting step had better coloring sharpness in comparison with that without alum fixation process.

In the case of sample for getting pH of 5, ethanol was used. Then, to understand the effects of pH on the coloring process, acetic acid was used to get pH of 3 and 4. To get pH of 7, more water dilution to the ethanol solvent was used. The photograph image of the dyes produced with various pH conditions is inserted in the left-bottom of the Fig. 1.

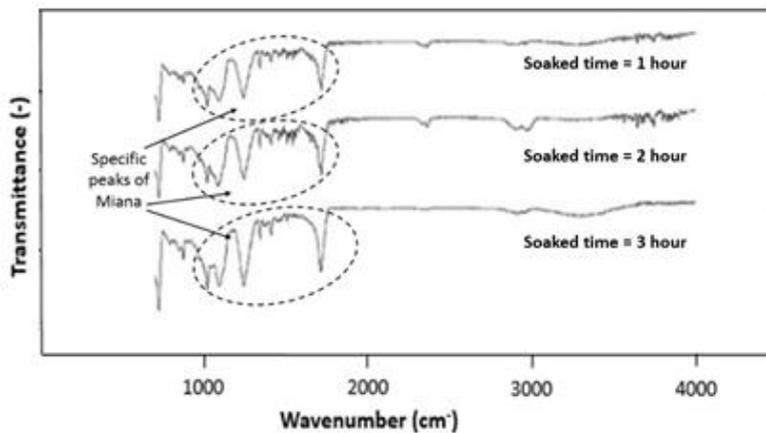
Extracted miana leaves could be used as natural dyes because it had chemical content that produced color and could be bound to fabric fibers. Figure 2 is the result of FTIR analysis on colored fabrics with extracted miana leaves with various soaking times. To ensure the effect of soaking time, FTIR curves were classified as the top, the middle, and the bottom for the sample that was soaked for 1, 2, and 3 hours with alum fixation, respectively.

The appearance of the extracted miana leaves using 95% of ethanol is shown in the paneled image in the bottom left in Fig. 2. The results showed several peaks were identified, in which this is in a good agreement with the tannin-related components. For example, from the results of the analysis in accordance with the results of FTIR analysis on tannins [19], there were peaks at wavenumbers of 1040 and 1115  $\text{cm}^{-1}$ , indicating participation of  $-\text{CO}$  and  $-\text{CH}$  groups. Also, peaks in the wavenumbers of 1370-1380  $\text{cm}^{-1}$  indicated the  $-\text{OH}$  and  $-\text{CH}$  -related groups, informing that the produced extract solution can be dissolved in water and ethanol. Detailed information about the peak in the FTIR is shown in Table 1.



**Fig.1.** The photograph images of fabric after additional extracted miana leaves. Figure (1) is the sample before the coloring process. Figure (a) is the samples that are dyed and fixated by alum with soaking time (1, 2, 3) 1, 2, and 3 hours, respectively. Figure (b) is the samples that are dyed without alum fixation with soaking time (1, 2, 3) 1, 2, and 3 hours, respectively. The insert image in the bottom left is the extracted miana leaves with various pH conditions.

Figure 3 is the UV-Vis spectrophotometric analysis of the extracted miana leaves in various pH conditions. The detailed information about the correlation of peaks with the potential compound in the solution is shown in Table 2. Several peaks were obtained, especially at wavelength of 250, 350, and 650 nm. This confirms the potential extracted solution for producing different colors and chemical compounds. Indeed, this informs that producing a specific color has a correlation to the additional treatment, such as controlling pH solution.



**Fig. 2.** The FTIR analysis of alum-fixed fabrics with various soaked times.

**Table 1.** Functional group of Miana leaf extract in FTIR.

Functional Group	Wavenumber (obtained from experiment) (cm <sup>-1</sup> )	Wavenumber (from references) (cm <sup>-1</sup> )
=CH (alkena)	996	995-710
C-O alcohol	1239	1050-1260
C-H (at CH <sub>3</sub> )	1337	1300-1475
C-H (at CH <sub>2</sub> )	1412	1300-1475
C=C	1717	1620-1680
C-H aliphatic	2911	2800-3000

The differences in the pH conditions caused different peaks appeared in the UV-Visible spectrophotometry analysis (see Fig. 3), in which these results are in a good agreement with the changes in colors (see Fig. 1). Using four pH conditions, all samples have the same highest peak at wavelengths of 265-268 nm, in which this peak corresponds to flavonoids [20]. The results showed a major peak at the wavelength 400 nm, which is identified tannin compounds in extracted miana leaves.

At the sample using pH of 5, a slight different result was obtained. There are three different peaks at wavelengths of 411, 536, and 608-665 nm, corresponding to auron, anthocyanidin, and condensed groups, respectively (see Table 2) [20-22]. These compounds make different types of color on different pH conditions. Also, these compounds are used to make green color.

The changes in color are due to the change of tannin stability. At the pH condition of 3, tannins will form oxonium or stable flavillium salts. At higher pH condition, oxonium in the solution will be hydrolyzed, producing pseudobasa components, in which this component can undergo tautomeric and there is an equilibrium condition between keto- and enol-form [23].

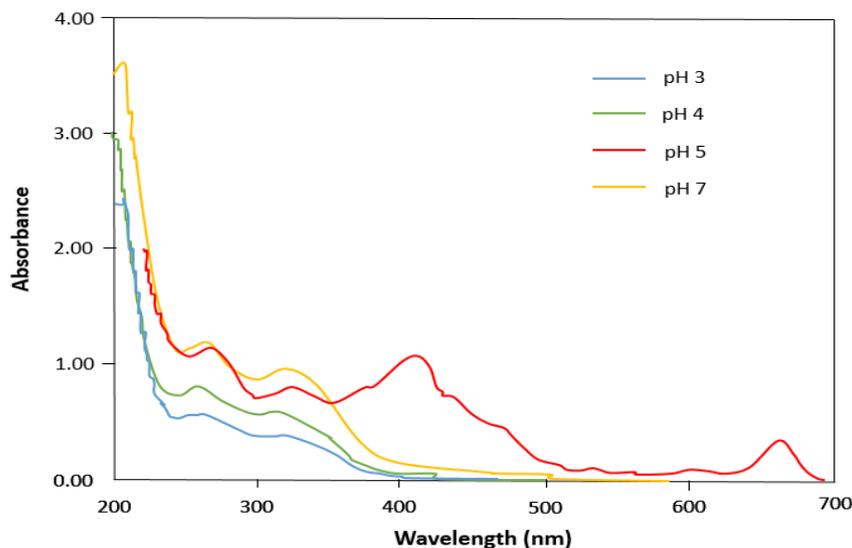


Fig. 3. The results of UV Visible spectrophotometry analysis of extracted miana leaves in various pH conditions.

Table 2. Visible spectrum of miana leaf extract in various pH conditions.

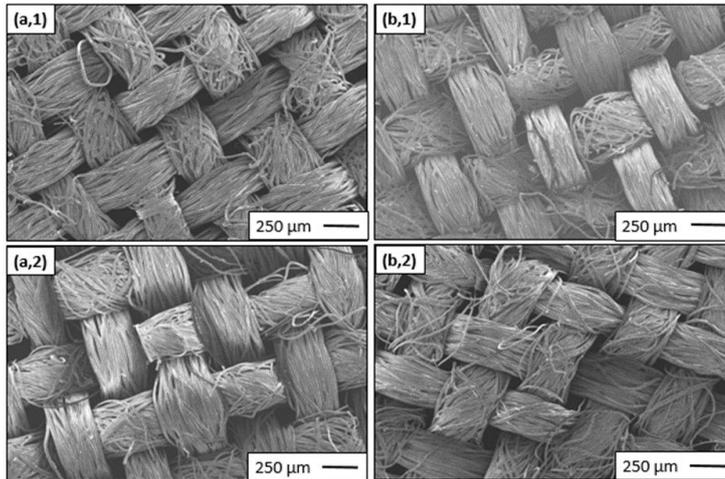
pH	Peak	Compound
3	207	ellagic acid
	265	flavonoid
	314	flavon
4	265	flavonoid
	315	flavon
	671	condensed group
5	268	flavonoid
	327	flavon
	411	auron
	536	antosianidin
	608	condensed group
	665	condensed group
7	208	ellagic acid
	265	flavonoid
	321	flavon

The examination of the coloring fabrics was carried out using a washing method using a detergent (see SEM images in Figs. 4 and 5). The results of this analysis were analyzed by SEM at 50x (Fig. 4) and 2000x magnification (Fig. 5). The result showed the patch of the tannin extract on the surface of the fabrics.

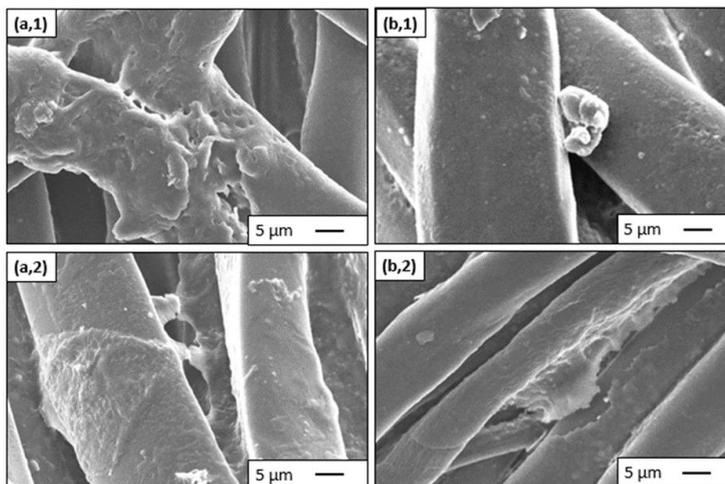
Figure 4 shows the SEM images of the samples after dipped into extracted miana leaves. Comparison of samples before and after the washing process (using detergent) was to obtain the effect of alum fixation on the fabrics. The result showed no different in the SEM observation.

Since the low magnification of SEM images in Fig. 4 was not clear enough to confirm the patch of tannin color (from extracted miana leaves), analysis using high-magnified SEM image is important (Fig. 5). As shown in the figure, the patch of material (which was identified as a complex of tannin) was clearly obtained when using high magnification of SEM images (Fig. 5). We found that the additional alum fixation had the impact to the more patched materials on the surface of fabrics. The larger components attached to the fabrics confirmed that the coloring agents cannot enter and penetrate into the deepest position in the fabrics. Indeed, this results in the release of this component after several times of washing process. However, when using the fixation process, the small components were produced, making them to penetrate into inside of the fabrics.

Based on the above results, the existence of alum was effective for binding tannin (gained from extracted miana leaves). This was confirmed by several characterization analyses shown in the above. The main idea for the successful coloration is due to the existence of aluminum ions ( $Al^{3+}$ ) that are from the dissolution of alum.  $Al^{3+}$  ions then easily penetrate to the deepest position in the fabric fibers. The penetrated ions react with the extracted miana leaves to form large-size complex salt that is bound inside the fabric fibers. The large complex was confirmed by the existence of attached material into the fabrics (see SEM images in Fig. 5). As a consequence, dye could not be removed easily from the fabric fibers.



**Fig. 4.** The results of low-magnified SEM analysis of samples with various fixations of alum. Figures (a, 1) and (b, 1) are the samples without and with alum fixation before washing process. Figures (a, 2) and (b, 2) are the samples without and with alum fixation after washing process.



**Fig. 1.** The results of high-magnified SEM analysis of fabrics under various fixations of alum. Figures (a, 1) and (b, 1) are samples without and with alum fixation before washing process. Figures (a, 2) and (b,2) are samples without and with alum fixation after washing.

Based on the above results, understanding the existence of alum has the potential to be applied in the batik industry to improve the natural dye coloration. However, further analyses must be done to improve the coloring batik.

#### 4. Conclusion

The investigation of the effect of existence of alum on batik coloring application has been presented in this study. As a model of batik coloring agent, miana leaf

extract was used. The experimental results showed that existence of alum is effective for binding extracted miana leaves on the fabrics, which was confirmed using several characterization analyses. Control of the coloring phenomena can be achieved by controlling pH condition. Some parameters must be considered to make better penetration of alum.

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