

Enhancement of the Dyeability of Ecoprint Fabric by Chitosan Biomordant Dyeing Process

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Abstract: Ecoprinting is an environmentally friendly printing technique used in the textile industry that utilizes plant-based dyes. Ecoprinted products often lack vibrant color because some natural dyes lack the active groups necessary to form chemical bonds with fabric fibers. Consequently, a mordant is required to facilitate bonding between the natural dye and the fibers. This study investigated the effects of chitosan mordant concentration and mordanting duration on ecoprinted fabrics dyed with teak leaves. Color intensity was evaluated both visually and using a chromameter. The results demonstrated that the chitosan mordanting process enhances color intensity and improves the definition of the teak leaf patterns. Optimal mordanting conditions were achieved with a chitosan concentration of 0.3% and a mordanting time of one hour. FTIR analysis revealed interactions between the active groups of the chitosan and the active groups of the fibers and dye pigments within the ecoprinted fabric.

Keywords: Ecoprint; Teak Leaf; Chitosan; Mordant.

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I. INTRODUCTION

Ecoprinting is a textile industry technique that is gaining popularity and expanding rapidly in Indonesia. It is widely favored due to the relative ease of the process, the use of eco-friendly materials, its significant economic value, and the simplicity of the required equipment. In addition to utilizing natural dyes, ecoprint fabrics possess unique and modern artistic and design qualities [1].

Ecoprinting is a natural printing technique that utilizes materials such as leaves, twigs, and flowers. It serves as an innovative approach within "ecofashion" to create environmentally friendly products. However, the results of ecoprinting are often considered suboptimal; the technique tends to yield less vibrant fabric colors because it relies on natural dyes [2]. This occurs because certain natural dyes lack the active groups necessary to form chemical bonds with fabric fibers, resulting in less-than-ideal color outcomes. Consequently, the production process requires the addition of a mordant [3]. A mordant is a substance that acts to fix and enhance color. Its use serves to bind the dye to the fibers,

improve the fabric's absorption quality, intensify the color, and enhance color fastness [4].

Fabrics suitable as a base for ecoprinting are those made from natural fibers, such as cotton, which is derived from natural cellulose fibers. Cotton is widely used because ecoprinting relies on plant-based dyes, requiring a fabric capable of absorbing these natural pigments [5]. However, most natural dyes exhibit low substantivity, meaning they fade easily and often fail to produce optimal color vibrancy on the fabric. This occurs because natural dyes lack the active groups necessary to form chemical bonds with the fibers of the fabric. To ensure optimal color absorption, an additional process known as mordanting is required; this can be performed either before or during the dyeing process [6].

Conventionally, metal salts such as aluminum sulfate and iron(II) sulfate are widely used as the primary raw materials in the process. However, metallic substances like aluminum and iron have adverse effects on the environment and human health [7]. To address this issue, many studies have utilized biomordants derived from organic materials as substitutes for metal salts [8].

Chitosan is a natural copolymer composed of glucosamine and N-acetylglucosamine units [9]. It is obtained through the deacetylation of chitin. Chitin is the most abundant biopolysaccharide after cellulose and is widely found in the shells of crustaceans, such as shrimp shells, crab shells, and lobster shells.

In practice, chitosan can serve as a raw material for an effective biomordant solution in the dyeing of fabrics with natural dyes. Chitosan is a type of natural carbohydrate possessing cationic amine groups. These cationic groups can bond with the active groups of other natural carbohydrates, which are predominantly anionic [8].

Cotton fabric is composed of cellulose fibers containing numerous hydroxyl (–OH) groups, rendering them reactive and capable of interacting with other substances. In the ecoprinting process using teak leaves (**Tectona grandis**), the resulting colors are derived from natural pigments such as anthocyanins, flavonoids, tannins, and quinones [10]. These pigments bind to the cellulose fibers through hydrogen bonding and physical interactions, allowing the color to adhere to the fabric. The resulting color variations—ranging from red to purple—are influenced by environmental conditions, such as pH, and the specific treatments applied during the ecoprinting process. The presence of –OH groups on the cellulose plays a crucial role in this process, serving as the primary site of interaction between the fibers and the natural pigments [11].

In this study, cotton fabric serves as the primary raw material for producing ecoprint fabric, while teak leaves are used as the natural dye source. The steaming technique was selected because it yields more durable colors. A chitosan-based biomordant is employed during the production process to enhance color vibrancy. The study examines the effects of chitosan concentration and mordanting duration on the characteristics of the resulting ecoprint fabric.

II. MATERIALS AND METHODS

➤ Materials

Cotton fabric from PT Kesmatex Pekalongan, Indonesia. Teak leaves are obtained from trees in the Ngalihan Teak plantation in Semarang, Indonesia. The chitosan used was industrial-grade, supplied by PT. Biotech Surindo, Indonesia (Deacetylation Degree: 83%). Sodium carbonate, Aluminum sulfate.

➤ Preparation of Materials

Teak leaves were cleaned of adhering impurities using distilled water. The cotton fabric was washed with soap and then air-dried for one day. Subsequently, the cotton fabric was soaked in a solution mixture of 14 grams of aluminum sulfate and 6 grams of sodium carbonate in 1 liter of distilled water. The fabric was soaked for one hour at a temperature of 80°C, followed by continued soaking for 24 hours. Finally, the fabric was removed from the solution and air-dried.

➤ Creating Ecoprint Fabric

Once dried, the fabric is ready for the ecoprinting process. This study employs a steaming technique. First, the fabric is lightly misted with water until damp. Next, teak leaves are

arranged on the spread-out fabric. The layer of fabric and leaves is covered with plastic, then the fabric is rolled up and tightly bound with a string. The rolled fabric is steamed for two hours. After steaming, the bindings are removed, and the leaves are lifted from the fabric surface. The fabric is then air-dried indoors until evenly dry. Once dry, it proceeds to the mordanting process.

➤ The Mordanting Process

The first step is the preparation of the mordant solution (chitosan solution) at concentrations corresponding to the experimental variables. The mordanting process involves soaking the fabric in the solution, ensuring it is fully submerged for durations of 30 minutes, 1 hour, 1.5 hours, and 2 hours, as dictated by the variables. The fabric is then dried by leaving it on a tray for one day. Once the ecoprint fabric is dry, the analysis of the product's characteristics is conducted.

➤ Analytical Methods

Color intensity testing was conducted using a Konica Minolta CM-5 chromameter. Color measurements were taken to obtain L*, a*, and b* parameter values based on the CIELAB (Commission Internationale de l'Éclairage) color system. The L* parameter indicates the level of color intensity, where lower values signify darker and more intense colors, while higher values indicate lighter colors. The a* parameter indicates the color tendency between red and green, with positive values leaning towards red and negative values towards green. The b* parameter indicates the color tendency between yellow and blue, with positive values leaning towards yellow and negative values towards blue. The CIELAB system is widely used in textile color evaluation because it is capable of quantitatively representing human visual perception (Kumah et al., 2019).

Visual testing was conducted by directly observing the ecoprint fabric with the naked eye. The chemical functional groups present on the eco-printed fabric were analyzed using Fourier Transform Infrared Spectroscopy (FTIR). FTIR spectra were recorded using a Shimadzu IR Prestige-21 spectrophotometer within the wavenumber range of 500–4000 cm⁻¹.

III. RESULTS AND DISCUSSIONS

➤ The Effect of Chitosan Concentration on the Color of Ecoprint Fabrics

This study analyzes the effect of varying chitosan concentrations on the color intensity of ecoprinted cotton fabric. Chitosan was applied as a mordant to enhance the color intensity of the ecoprinted fabric; it was introduced during the mordanting stage following the ecoprinting process. The study was conducted under constant operating conditions, specifically a mordanting time of one hour and the use of teak leaves. Color changes resulting from different chitosan concentrations were observed. The results of the chromameter analysis for the ecoprinted fabric at various chitosan concentrations are presented in Figure 1.

Fig 1 shows that the L* value decreases as the chitosan concentration increases, indicating an increase in color intensity. The a* value shows a trend of increasing redness intensity, peaking at a chitosan concentration of 0.3%.

Meanwhile, the b^* value tends to shift towards a darker tone, reaching its lowest value at a chitosan concentration of 0.3%.

For teak leaf ecoprint fabric, the desired outcome is a deep reddish-purple hue with clearly defined leaf structures. In the CIELAB color system, this reddish-purple characteristic is defined by a relatively low L^* value, a high positive a^* value, and a low b^* value (approaching zero) [12]. The test results in

Figure 1 show a decrease in L^* values as chitosan concentration increases, indicating that the ecoprint fabric becomes darker and more intense in color. The consistently positive a^* values across all concentration variations indicate a predominance of red tones, while the relatively low b^* values suggest minimal yellow contribution, resulting in a color that leans towards reddish-purple.

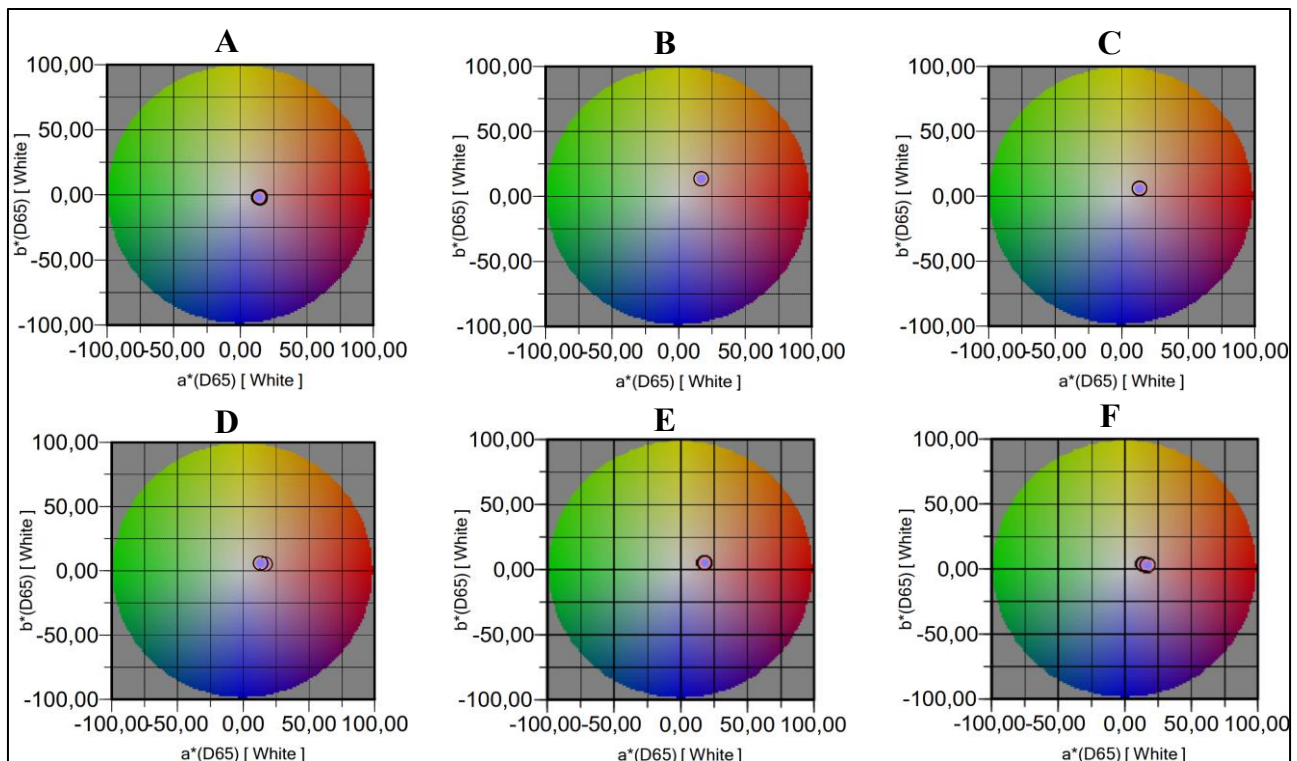


Fig 1 Chromameter Test Results for Ecoprint Fabric at Various Chitosan Concentrations (A) 0% Chitosan B) 0.05% Chitosan C) 0.1% Chitosan D) 0.2% Chitosan E) 0.3% Chitosan F) 0.4% Chitosan).

Research findings indicate that increasing the chitosan concentration up to a certain limit enhances fabric color intensity, evidenced by lower L^* and b^* values and higher a^* values. This demonstrates that the resulting color becomes darker and more saturated as chitosan content increases [13]. This effect arises from chitosan's ability to form a transparent, homogeneous film on the fiber surface, leading to more uniform pigment distribution and enhanced color visibility [14], [15]. However, at a chitosan concentration of 0.4%, color intensity decreases, and the fabric becomes stiffer. This indicates that excessively increasing chitosan concentration does not necessarily improve color quality. High concentrations cause chitosan molecules to accumulate on the fiber surface, forming a layer that hinders the bonding between the dye and the fibers, thereby reducing color intensity [16]. Furthermore, excessive chitosan concentration results in a denser chitosan layer, increasing fabric stiffness due to stronger intermolecular bonds forming between the chitosan and the fabric fibers. This condition restricts fiber mobility and reduces flexibility [17].

The relationship between increasing chitosan concentration and changes in L^* , a^* , and b^* values does not follow a linear pattern, as it is influenced by the non-homogeneous distribution of natural pigments and limitations

in bond formation on the fabric fibers. Furthermore, the formation of increasingly dense cross-links on the fiber surface can enhance light scattering, thereby affecting the color values measured by the chromameter. Consequently, evaluating the color of ecoprinted fabric requires considering the combination of L^* , a^* , and b^* values simultaneously to achieve a more accurate color interpretation [18].

Based on the test results, varying chitosan concentrations resulted in differing effects on the color intensity and characteristics of teak-leaf ecoprinted fabric. At a chitosan concentration of 0.3%, the resulting reddish-purple color appeared more intense. At higher concentrations, color intensity tended to decrease, and the fabric exhibited increased stiffness. Therefore, a chitosan concentration of 0.3% yielded the most balanced results regarding color quality and physical fabric characteristics in this study.

➤ The Effect of Mordanting Time on Color in Ecoprint Fabrics

Color changes resulting from varying mordanting times were observed at a chitosan concentration of 0.3% using teak leaves. The results of the chromameter analysis of the ecoprint fabric at various mordanting times are shown in Figure 2.

The effect of mordanting time on the color of ecoprint fabric, as determined by chromameter analysis, is shown in Figure 2. At 0 hours (before the mordanting process), the L^* , a^* , and b^* values were 44.78, 14.82, and 2.59, respectively, indicating a slightly faded reddish-brown color. At 30 minutes, the L^* , a^* , and b^* values were 40.53, 24.95, and -0.26, respectively, indicating a fairly intense brick-red color. At 1 hour, the L^* , a^* , and b^* values were 26.14, 22.52, and 1.76, respectively, indicating an intense purplish-red color. At 1.5 hours, the L^* , a^* , and b^* values were 34.02, 22.02, and 1.15, respectively, indicating a somewhat faded dark red color. At 2 hours, the L^* , a^* , and b^* values were 46.80, 23.07, and -3.99, respectively, indicating a faded purplish-red color.

Figure 2 also shows that the L^* -value before mordanting was 44.78; the value subsequently decreased as the mordanting time increased, then rose again after the one-hour mark. This decrease in the L^* -value indicates an increase in the color intensity of the ecoprint fabric. The a^* value shows a trend of increasing redness intensity, peaking at the 30-minute mark. The b^* parameter—which indicates the color tendency between yellow and blue—fluctuated, shifting from 2.59 at 0 hours to -0.26 at 30 minutes, before rising to 1.76 at the one-hour mark

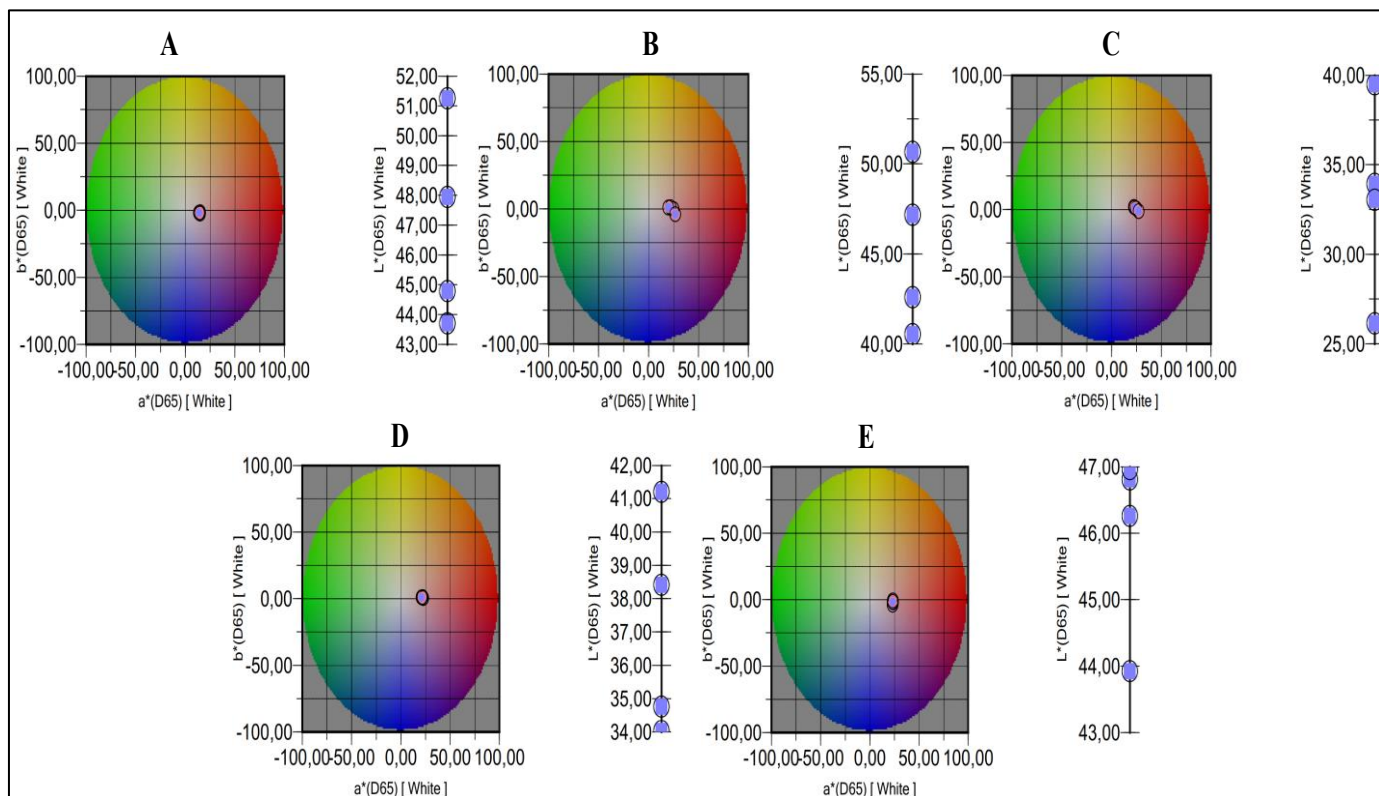


Fig 2 Chromameter Test Results for Ecoprint Fabric at Various Mordanting Times (A) 0 Hours B) 0.5 Hours C) 1 Hours D) 1.5 Hours E) 2 Hours).

➤ FTIR Analysis

Fourier Transform Infrared (FTIR) analysis was conducted to compare the functional group characteristics of ecoprint fabric samples—one without chitosan mordanting and the other with 0.3% chitosan mordanting for one hour. This analysis aimed to identify changes in chemical structure and detect interactions between functional groups that contribute to the stabilization of the material's structure. Figure 4.3 presents the FTIR analysis results for both samples.

The FTIR spectrum of the chitosan-mordanted fabric exhibits absorption characteristics indicating the presence of amide groups, a hallmark of chitosan. The absorption band appearing around 1600 cm^{-1} falls within the Amide I region;

however, this region may also encompass contributions from Amide II, associated with N–H and C–N bonds [19]. These amide groups serve as a key indicator that chitosan has been deposited on or has interacted with the fabric fiber surface [20]. In addition to the amide region, a broad absorption band at 3330 cm^{-1} was observed in the chitosan-mordanted fabric sample, indicating the presence of hydroxyl groups. The broadening of this band suggests bonding between the hydroxyl groups—found in the cellulose fibers and the color pigment—and the amine or amide groups of the chitosan. The band at 2895 cm^{-1} indicates C–H bonds, while the band in the $1150\text{--}1000\text{ cm}^{-1}$ region is associated with C–O–C bonds, which are characteristic of polysaccharide structures [21].

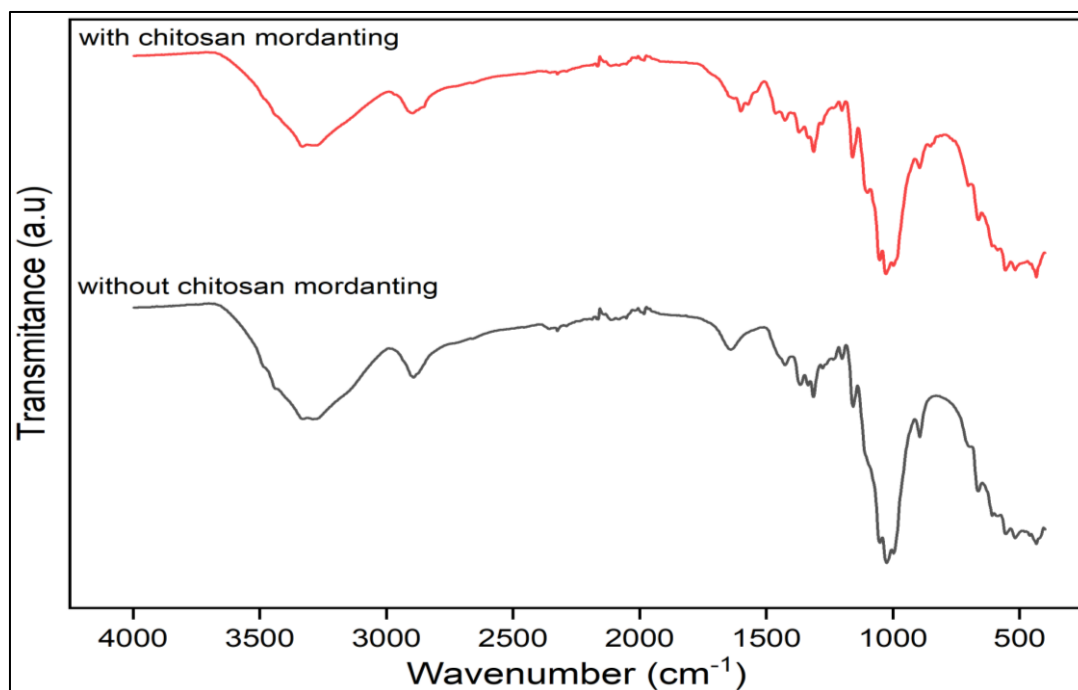


Fig 3 FTIR Spectra of Ecoprint Fabric with Mordanting and Without Mordanting Under Optimum Conditions.

In contrast, the FTIR spectra of the fabric without chitosan mordanting did not exhibit the characteristic bands of amide groups. In the spectra of the fabric without chitosan addition, the observed absorption band corresponded to the hydroxyl group (at 3290 cm^{-1}), a characteristic feature of the cellulose fiber structure [22]. Furthermore, no absorption bands were detected in the range characteristic of amide groups—specifically the $1500\text{--}1600\text{ cm}^{-1}$ region. This indicates that the fabric without chitosan addition does not contain amide groups [21].

hours to -0.26 at 30 minutes, before rising to 1.76 at the one-hour mark

➤ The Effect of Chitosan Concentration on the Visualization of Ecoprint Result

Visualization of the ecoprint results was conducted to directly observe changes in color intensity and the sharpness of the leaf patterns. This observation was performed qualitatively based on photographic documentation obtained for each treatment. Subsequently, the visual observation results were compared with quantitative data obtained from color measurements using a chromameter to determine the correspondence between visual perception and the numerical values of color parameters. Figure 4 shows the visual appearance of the ecoprint results at various chitosan concentrations.

At a concentration of 0%, the resulting color appeared palest, with less distinct leaf patterns. This likely occurred because the natural dye's binding capacity for cellulose fibers

was limited, relying solely on interactions with the -OH groups on the cotton fabric [23]. At concentrations of 0.05% and 0.1%, color intensity increased, and the patterns became more visible. This indicates that the amine groups ($-\text{NH}_2$) of the chitosan began to play a role in enhancing dye affinity through electrostatic interactions and hydrogen bonding [24].

A further increase in color intensity was observed at a concentration of 0.2%, where the color appeared deeper and more evenly distributed; meanwhile, the 0.3% concentration yielded the highest color contrast and the sharpest leaf details, indicating optimal color fixation. However, at a concentration of 0.4%, although the color remained deep, there was a tendency toward uneven color distribution caused by the formation of an excessively thick chitosan layer on the fiber surface, which hindered uniform dye penetration [16].

Visual observations align with the chromameter data. Increasing the chitosan concentration affects the fabric's color intensity. Visually, the color deepens as the concentration rises from 0% to 0.3%; this corresponds to the chromameter results, which show a decrease in the L^* value (indicating a darker color) and an increase in the a^* value. At a concentration of 0.4%, although the color remains intense, the result is not superior to that at 0.3% and tends to be less uniform; consequently, the chromameter values show no significant improvement compared to the 0.3% concentration. Therefore, a chitosan concentration of 0.3% is optimal, as it yields the most visually distinct color, a finding supported by the corresponding chromameter measurements.

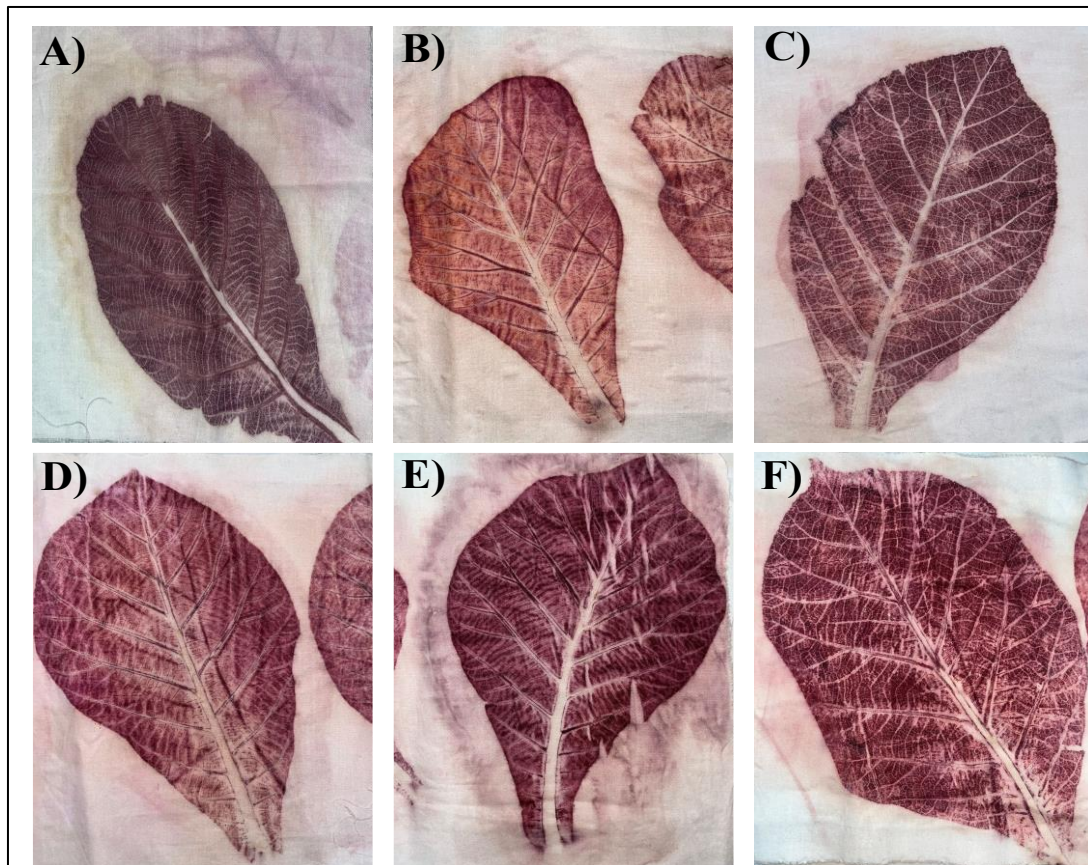


Fig 4 Results of the Visual Evaluation of Teak Leaf Ecoprinting at Various Chitosan Concentrations (A) 0% Chitosan B) 0.05% Chitosan C) 0.1% Chitosan D) 0.2% Chitosan E) 0.3% Chitosan F) 0.4% Chitosan).

➤ *The Effect of Mordanting Time on the Visual Outcome of Ecoprint*

Visual observations of ecoprint fabrics both without mordanting and with mordanting for various durations using a 0.3% chitosan concentration are shown in Figure 5.

Figure 5 shows that in the treatment without mordanting, the color appears paler and the pattern less distinct, indicating that the interaction between the cellulose fibers and the color pigment has not formed optimally [23]. At a mordanting time of 30 minutes, the color becomes more pronounced, though not yet at its maximum intensity. At a mordanting time of 1 hour, the color appears most intense, and leaf details are clearly defined. However, at mordanting times of 1.5 and 2 hours, no significant increase in color intensity is observed. Once the mordanting time reaches 1 hour, the majority of

active groups on the fibers have likely bonded with the chitosan groups, thereby limiting the number of available binding sites and causing the fibers to reach saturation. Consequently, extending the mordanting time no longer enhances the absorption or fixation of the dye onto the fabric fibers [25].

The visual observations align with the chromameter test data. The L^* value showed a decrease up to the one-hour mark, indicating a darkening of the color along with an increase in the a^* value and a decrease in the b^* value, signifying heightened color intensity. However, beyond one hour, changes in color values tended to be insignificant, even with extended duration. Therefore, it can be concluded that the optimal mordanting time is one hour, as it yields the most intense color and a more distinct leaf motif structure.

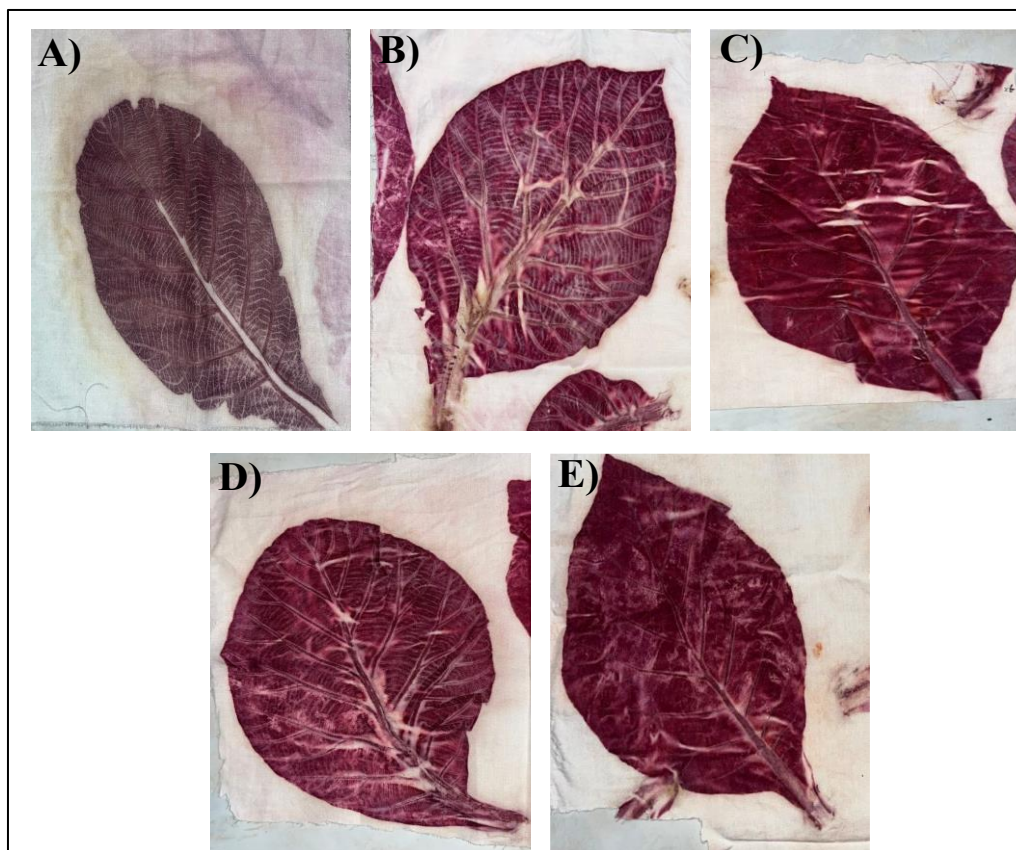


Fig 5 Effect of Mordanting Time on The Color Visualization of Ecoprint Fabric (A) 0 Hour B) 0.5 Hours C) 1 Hours D) 1.5 Hours E) 2 Hours).

IV. CONCLUSION

The research results indicate that chitosan mordanting affects the color intensity of ecoprint fabrics. Color intensity increased with rising chitosan concentrations up to 0.3% but decreased at a concentration of 0.4%. Mordanting duration also influenced the color intensity and physical properties of the ecoprint fabrics. Extending the mordanting time increased color intensity up to one hour, but no further change in intensity was observed beyond that duration. FTIR analysis revealed that the addition of 0.3% chitosan influenced color intensity. The appearance of characteristic absorption bands associated with amide, O–H, and C–O groups indicates interactions occurring between the chitosan, color pigments, and cellulose fibers of the ecoprint fabric.

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