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# Utilization of Tamarind Seeds Extract as a Natural and Sustainable Fabric Dye

Astri Nur Istyami[1,2], Muhammad Arif[1], Muhammad Ilham Azzindi[1], Meiti Pratiwi[1,2], Sangono Adisasmito[2], Nuning Yanti Damayanti[3], Agus Tendi Ahmad Bustomi[2], Jenny Rizkiana[1,2,*]

[1]Department of Bioenergy Engineering and Chemurgy, Faculty of Industrial Technology, Institut Teknologi Bandung, Bandung, 40132, Indonesia
[2]Department of Chemical Engineering, Faculty of Industrial Technology, Institut Teknologi Bandung, 40132, Indonesia
[3]Faculty of Art and Design, Institut Teknologi Bandung, Bandung, 40132, Indonesia
[4]Center for Catalysis and Reaction Engineering (CaRE), Institut Teknologi Bandung, Bandung, 40132, Indonesia

[*]*Corresponding author: rizkiana@itb.ac.id*

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**Abstract**

This research focuses on the use of tannin components in tamarind seed coats as a mordant and natural dye in cotton fabrics. Tannins were extracted from the tamarind seed coat by boiling method and then the tannin content was determined. The tannin extract was then used as a natural mordant with the addition of metallic copper sulfate ($CuSO_4$) mordant. Tannin extract is also used as a dye on fabrics with the addition of sodium sulfate ($NaSO_4$). The color strength of the tannins in the fabric was analyzed using a spectrophotometer from the rinse water. The results showed that the cloth that had been given the mordant had a stronger color strength than the cloth without the mordant. The use of mordant was varied at concentrations of 5, 10, 15, 20, and 25%-owf. The results of the analysis showed the most optimum tannin concentration at 15%-owf. The concentration of tannin used in the coloring process was also varied at concentrations of 5, 10, 15, 20, and 25%-owf. The results of the analysis show that the concentration of tannin used in the dye does not affect the strength of the color, but only affects the brightness of this color.

*Keywords: Tamarind seed coat; Tannin; Mordant; Dye; Color strength*

## INTRODUCTION

Indonesia marked a historical event in 2009 after UNESCO as the United Nations Educational, Scientific, and Cultural Organization designated batik as the original Indonesian cultural clothing. This spurred the domestic batik industry to produce batik not only for domestic needs but also for export. The high production of batik is in line with the need for batik-making materials, one of which is dye materials. The majority of dyes used in the batik industry are synthetic dyes. Synthetic dyes are chosen due to the availability of the material in large quantities. However, synthetic materials certainly present many shortcomings to the health of the surrounding environment.

Some common dyes used in the textile industry include black, red, and golden yellow remazol. In the dyeing process, the compound can only be used for about 5% while the rest will be wasted as liquid waste. Due to its remarkable stability, this dye resists natural degradation, making it challenging to break down (Dewi & Rahmayanti, 2022). When Batik wastewater, which contains the remazol dye, is released into the environment, it can persist for an extended period and gradually build up to a specific concentration (Bijang, Nurdin, Latupeirissa, Aziz, & Talapessy, 2022; Pambudi, Prayogo, Nadjib, & Ediati, 2021). At high concentrations, this compound is harmful to the environment because it can increase Chemical Oxygen Demand (COD) and Biological Oxygen Demand (BOD) levels in water (Sutisna et al., 2017).

Batik liquid waste generally comes from dyeing processes, removal of dyestuffs from fabric fibres, and rinsing, most of which are discharged into nearby water discharges without proper treatment. Some previous reseacrches (Birgani et al., 2016; Mukimin, Vistanty, Zen, Purwanto, & Wicaksono, 2018) explained that some other non-biodegradable pollutants, including iron mordant (II) sulfate ($FeSO_4$), alum ($K_2SO_4$. $Al_2$ $(SO4)_3$.24 $H_2O$), calcium oxide (CaO), and wax were also found contained in this wastewater (Kusumawati, Rahmadyanti, & Sianita, 2021).

The impact of the use of synthetic dyes is felt by several neighborhoods around the batik industry. Kusumawati et al. (2021) stated that there was a heavy metal content in the Bengawan Solo River water caused by liquid waste from the surrounding batik industry (Kusumawati et al., 2021). River water pollution in Solo City due to batik industry liquid waste has exceeded the quality standard threshold. Two rivers, Premulung and Jenes, are even included in the category of heavy pollution. This assessment is based on the analysis of the results of water quality parameters, such as BOD and COD levels, heavy metal content, color, smell, and taste. In addition to Solo City, other batik-producing cities or regions such as Yogyakarta, Pekalongan, and Cirebon are also affected by the same things.

To overcome this, several solutions are offered, one of which is the use of natural batik dyes. It will present more positive sides to the environment and consumers. Some articles explained in more detail the positive sides of the use of natural dyes, such as biodegradable, non-toxic, does not cause allergies, environmentally friendly, creating jobs, and utilizing the land for plantations (Arora, Aggarwal, & Gupta, 2017; Samanta & Konar, 2011)

After knowing the positive impact of the use of natural batik dyes, this study offered a natural batik dye derived from tamarind seeds (*Tamarindus indica* L.). Tamarind seeds contain tannins that can be used as dyestuffs and mordant (Das, Islam, Faruk, Ashaduzzaman, & Dungani, 2020). Tannins from tamarind seeds can be extracted using a water solvent (De Hoyos-Martínez, Merle, Labidi, & Charrier – El Bouhtoury, 2019). Tamarind seeds can be used as an option as a natural source of dyes because in everyday life tamarind seeds are wasted from food processing and tamarind is easy to find anywhere (Petchidurai et al., 2019; Prabhu & Teli, 2014; Sarkar, Asif, Rahman, Islam, & Rahman, 2020). So that the use of tamarind seeds will reduce waste while creating natural dyes for batik.

## METHODOLOGY

### Materials and Instrumentals

The main ingredient used in this research is tamarind seeds. The supporting materials used are water as a solvent, Folin-Ciocalteu reagent for the analysis of tannin content, sodium carbonate, gallic acid, mordant, copper (II) sulfate solution, sodium sulfate, and detergents to test the color strength of fabrics. The sample solution was tested for tannin absorbance using a UV-Vis Spectrophotometer with a wavelength of 760 and 500 nm (color strength). The mordanting process is carried out using a cotton cloth with a size of 10x10 cm.

### Tamarind Seed Extraction

Tamarind seeds are dried using a stove for 30 minutes. The tamarind seed coat is separated from the seed by manually crushing it so that the seed coat and core separate. The tamarind seed coat that has been separated from the seed core is ground into powder. Tamarind seed powder is sifted to separate particles that have not been completely ground. 1000 grams of Tamarind seed coat powder was extracted with 2 Liters of water solvent for 2 hours at boiling point (100 ºC). After the extraction process, the solution is cooled. The solution is filtered with a fine cloth and the filtrate is collected separately. The residue remaining on the filter was further extracted three times under the same operating conditions to complete the extraction. The total extract is heated to boiling and left overnight. The clear filtrate is concentrated in a water bath and treated with a saturated salt solution. The brown-colored precipitate obtained is filtered with a paper filter and dried to produce tamarind seed coat tannins in powder form.

### Analysis of Tannin Content in Tamarind Seed Extract

The tannin content in tamarind seed extract was tested using the Folin-Ciocalteu method. 1 mL of tamarind seed extract is put into a test tube and then added with 5 mL of Folin-Ciocalteu reagent which was diluted 10 times and 5 mL of Sodium carbonate with a concentration of 7.5%. The sample solution was incubated for 20 minutes at a temperature of 20 $^{o}$C then compared its absorbance with the standard solution using a UV-Vis Spectrophotometer at a wavelength of 760 nm. The standard curve of the solution is prepared by mixing Folin-Ciocalteu reagent which was diluted 10 times, 5 mL of Sodium Carbonate with a concentration of 7.5%, and gallic acid with a concentration of 25, 50, 100, 200, 500 μg/mL of gallic acid.

### Mordanting Process

In the mordanting process, variations are carried out without mordant and with the addition of mordant at concentrations of 5, 10, 15, 20, and 25% mordant on the weight of the fabric (owf). Tannins from tamarind seed are prepared according to variations, then the tannins are dissolved in water with the ratio of water to fabric set at 30:1. Cotton cloth with a size of 10x10 cm is dipped in a mordant solution at a temperature of 95 $^{o}$C for 45 minutes. Furthermore, the

fabric is dipped in a solution of Copper (II) sulfate with a concentration of 1% on the weight of the fabric at a temperature of 85 $^{o}$C for 45 minutes. The fabric is then lifted for later dye the fabric.

**Dyeing Process**

After the mordanting process is carried out, the fabric is dipped in a dye solution varying at a concentration of 5, 10, 15, 20, and 25% (owf). Tannins are prepared according to variations, then the tannins are dissolved in water with the ratio of water to fabric set at 30:1. Dye solution is added with Sodium Sulfate with a concentration of 5 g / L. The prepared cloth is dipped in a dye solution at a temperature of 85 $^{o}$C for 45 minutes. The fabric is removed and then dried.

**Evaluation of Color Strength**

Fabrics that have been previously dyed are tested for fastness by the standard method of fastness testing ISO 105-C06. Detergents with a concentration of 5 g /l are used for washing fabrics. The fabric is soaked in a detergent solution for 45 minutes at a temperature of 50 ºC with a water-to-cloth ratio of 50:1. The fabric washing water was then analyzed for absorption using a UV-Vis Spectrophotometer with a wavelength of 500 nm. Fabric absorbance is compared with the standard solution. The standard curve of the solution is made using tamarind seed tannins with a concentration of 1, 3, 5, 7, 9, and 11 g/L of tannins.

## RESULTS AND DISCUSSION

**Analysis of Tannin Content in Tamarind Seed Extract**

Tannins are polyphenol compounds that contain a phenolic-hydroxyl group, the tannin content in tamarind seed extract can be calculated using the Folin-Ciocalteu method. During the reaction, the phenolic-hydroxyl group reacts with the reagent Folin-Ciocalteu, forming a phosphotungstate-phosphomolybdate complex of blue color (Rahayu & Inanda, 2015). An increasingly concentrated blue solution indicates the increasing amount of phenol compounds contained in the solution. The addition of $Na_2CO_3$ aims to form an alkaline atmosphere during the reaction. The results of the experiment can be seen in Figure 1.

The solution was then analyzed using a UV-Vis Spectrophotometer at a wavelength of 760 nm and the concentration was calculated using the gallic acid standard curve. Gallic acid can be used as a measurement standard for phenol level analysis because gallic acid is a derivative of hydroxybenzoic acid which is classified as a simple phenol acid and is stable (Lee, Kim, Lee, & Lee, 2003).

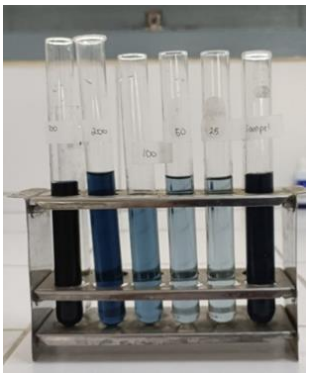


Figure 1. The result of the reaction of phenol compounds with Folin-Ciocalteu reagents

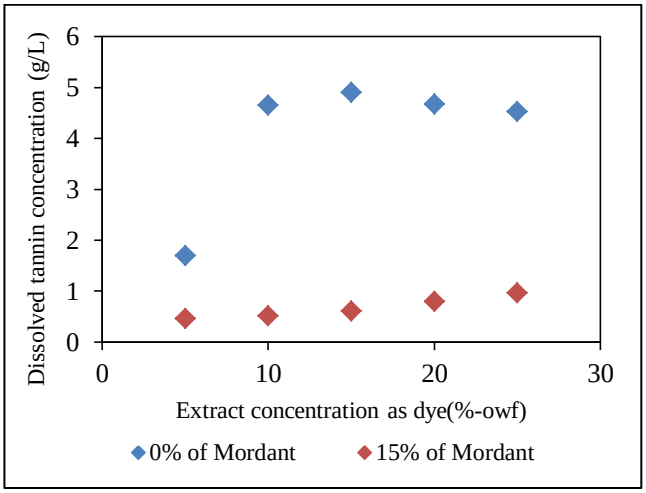
The measurement results of tannin samples resulted in an absorbance value of 2.1497. Using the standard curve regression equation of gallic acid, the sample's tannin concentration of 394.9 µg/mL was obtained. Then the total level of tannins in the extract is estimated by Equation 1.

$$T = \frac{C.V}{M} \quad (1)$$

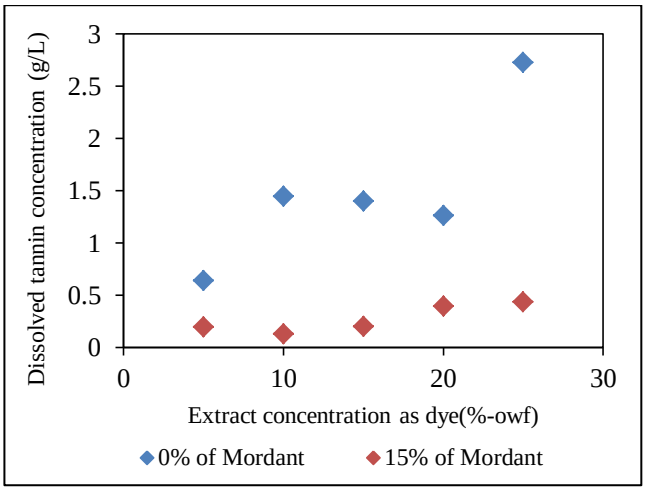
T is the level of tannins in the extract, mg/g of the extract; C is the sample concentration on the standard curve, mg/L; V represents the volume of the extract, mL; M represents the weight of the dry extract in the sample, g. From the calculation using equation 1, it is obtained that the level of tannins in tamarind seed extract was 0.394 gram tannins/g of extract. In other words, there are 39.4% tannins in tamarind extract.

**Analysis of the Effect of Tamarind Seed Extract as Mordant**

In this experiment, a process of dyeing cloth without and with the use of mordant was carried out to determine the effect of the use of mordant on the strength of color in the fabric. The color strength of the fabric is tested based on the fastness of the fabric when going through the washing process. To calculate the concentration of tannins dissolved in washing water, a standard curve of tannins is used which is derived from the absorbance value of the tannins. The results of the test on the concentrations of tannin without and with the use of mordant are shown in Figure 2.




(a)



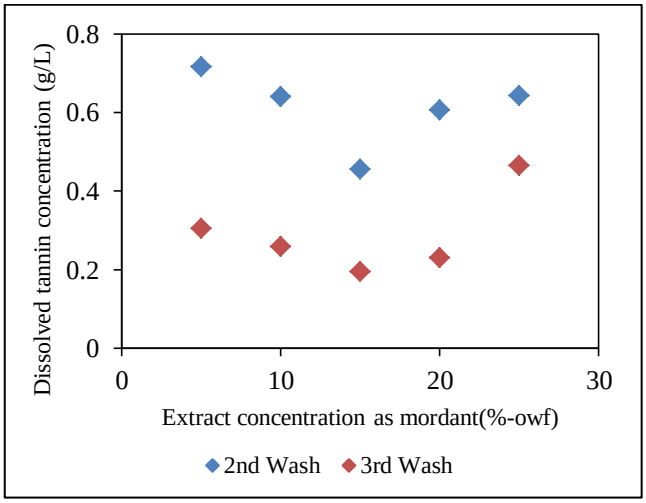

(b)

Figure 2. The effect of the addition of mordant on the strength of dyestuffs in fabrics at the (a) second wash (b) third wash

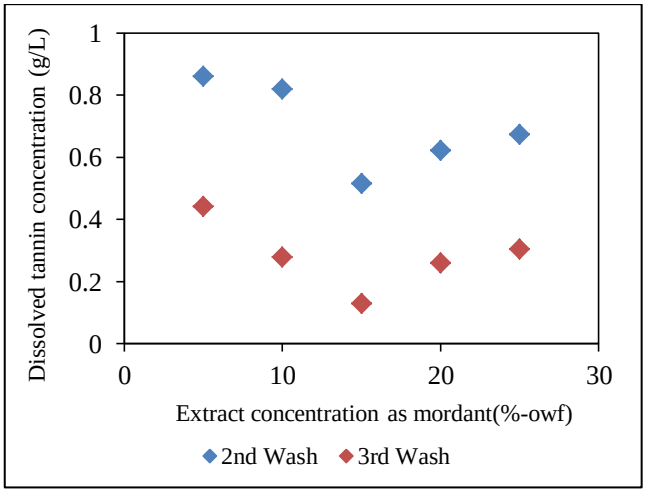
Based on the results of the experiments that have been shown in Figure 2, it can be seen that the use of mordants in fabric dyeing has a significant influence. Fabrics that are not treated with mordant have a lower fastness, this can be seen from the high concentration of tannins from the water washed by the fabric. Tannins contain a hydraulic phenol group that will form a complex with dye molecules and causes higher fixation of the dye in the fabric.

**Analysis of The Effect of Tannin Concentration on Dyeing and *mordanting processes* on The Strength of Color In Fabrics**

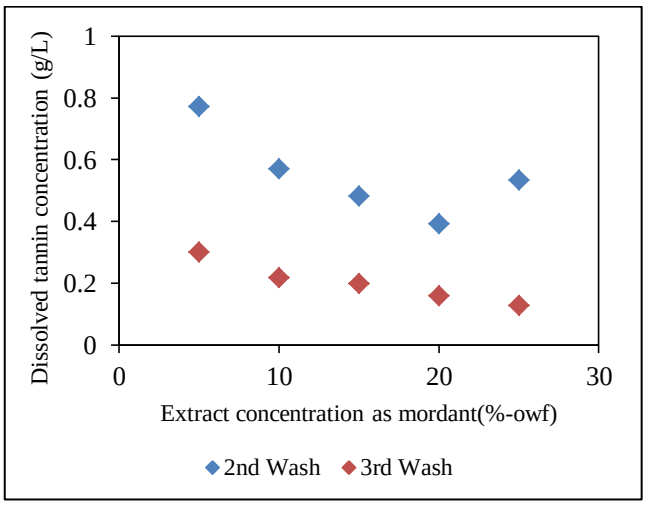
The concentration of tannins in the dyeing and mordanting processes was also analyzed using UV-Vis spectrophotometers from washed water of the fabrics. Testing the effect of mordant was carried out under 5 conditions, at dyeing concentrations 5, 10, 15, 20, and 25% owf. The test results on the concentration of tannin are presented in Figure 3.



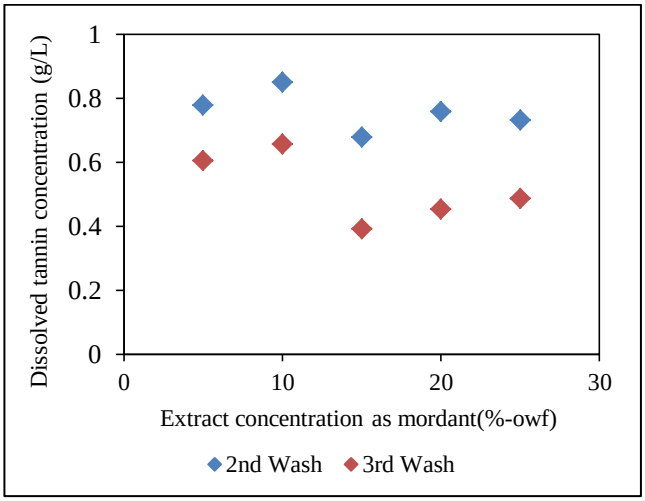

(a)



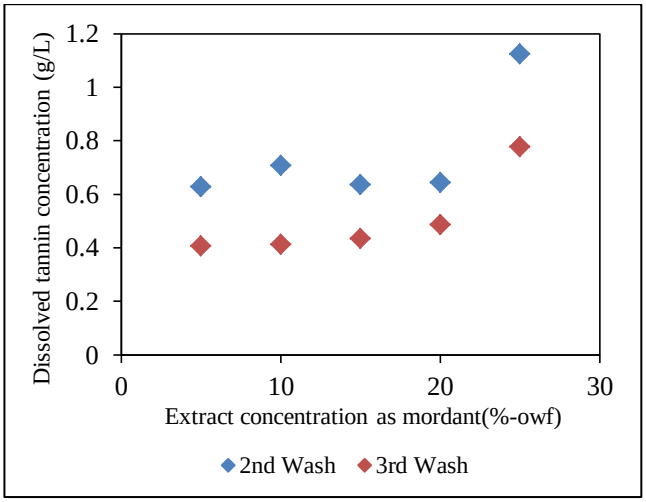

(b)




(c)




(d)




(e)

Figure 3. Effect of extract concentration as a mordant on color binding power in fabrics at staining concentrations (a) 5%; (b) 10%; (c) 15%; (d) 20%; (e) 25%

From the results of the experiments carried out, on average the best mordant concentration that produces the smallest concentration of dissolved tannins in washing water is a mordant with a concentration of 15% owf. This is due to two factors, namely the absorption of the fabric to the mordant and the quality of the mordant of tamarind seeds as a color-binding material. The absorption of fabrics to mordant is influenced by the type of fabric used, this study uses a type of cotton fabric as a test material for the dyeing process. According to Fitriani and Andriani (2019), textile materials are the most important object in determining success at the time of dyeing in the dyeing process (Fitriana & Adriani, 2019) . Linen and cotton are materials that have high absorption so they can be used as basic materials in the dyeing process. Then the next factor is the ability or quality of tamarind seeds as a mordant, the ability of tamarind seeds as a mordant has been tested so that tamarind seed mordant has good quality as a mordant.

The experimental results showed that the concentration of mordant tannins from tamarind seeds which had the best binding capacity on cotton fabrics was a concentration of 15% mordant to the weight of the fabric (owf). The concentration of mordant 20 and 25% owf from the data showed that the concentration of soluble tannins in the washing water was higher than 15% owf, this was because the absorption of cotton cloth on mordant tannins of tamarind seeds was only about 15% owf, so it cannot be completely absorbed by the fabric. This results in the tannin that is not bound to the fabric will fade during the washing process so that the value of the dissolved tannin content in the washing water is higher. In terms of cost, the concentration of mordant which is more than 15% adds a considerable cost but without a significant effect on the fabric. In addition, mordant with concentrations of 5 and 10% owf has higher dissolved tannin content in the washing water than the mordant with a concentration of 15% owf, this was because the concentration of mordant used as a binding agent was still lacking so the binding power of the mordant to the dye was not optimal. This results in dyes that are not properly bound to be dissolved in the washing water so that the dissolved tannin content in the washing water is higher.

The results of the experiment above show the most optimal mordant concentration of tamarind seed tannins is 15% owf. Furthermore, testing was also carried out at the staining stage, the staining stage varied the concentration of tannins when staining by 5.10,15,20.25% with the mordant used by 15%. The results of the experimental data can be seen in Figure 4.

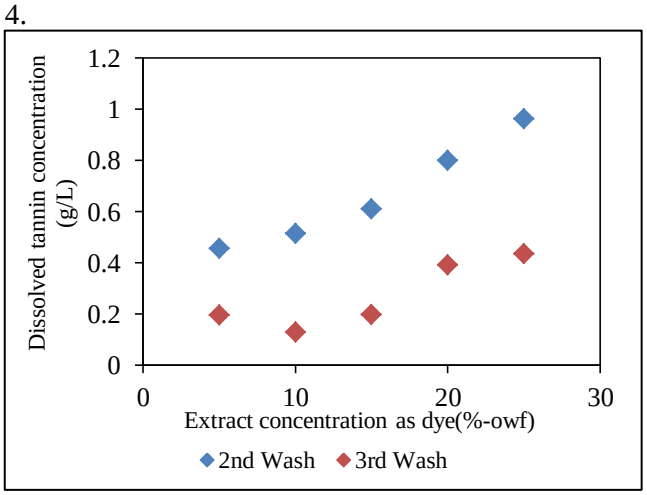



Figure 4. Influence of tannin concentration on the dyeing process

The results of the experimental data that have been shown in the graph, show that the trend of the data on the influence of coloring concentrations produces a trend that continues to increase along with the increase in dye concentration. This indicates that the variation in the concentration of dyes during the dyeing process is in no way related to the binding power of the color to the fabric. The concentration of staining affects only the desired brightness of the color. A sample of fabric staining results is shown in Figure 5.


Figure 5. Fabric dyeing samples

## CONCLUSION

Tannins are polyphenol compounds that contain a phenolic-hydroxyl group. The content of polyphenol compounds in tannins can be analyzed using the Folin-Ciocalteu method. From the results of the experiment, the tannin content in tamarind seeds was obtained by 39.4%. Tannin content makes the tamarind seed extract can be used as a natural dye for the fabric which gives the natural brown color. The strength of the color on the fabric can be increased by adding mordant. Fabrics that were mordant before the dyeing process had a smaller concentration of dissolved tannins than fabrics that did not go through the mordanting process. In this experiment, the fabric was given a mordanting treatment first before the dyeing process. From the results of the softening test on the fabric, the use of mordant with a concentration of 15% on the weight of the fabric (owf) gives the smallest average value of the concentration of dissolved tannins in washing water of the fabric. This shows that the color ties on the fabric are getting stronger.

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