Exploration and Standardization of Coconut Fiber Waste Utilization in Batik Dyeing Process

Nita Kusumawati¹, Samik², Supari Muslim³
¹,²Department of Chemistry, Universitas Negeri Surabaya, Ketintang, Surabaya, 60231, Indonesia
³Department of Electrical Engineering, Universitas Negeri Surabaya, Ketintang, Surabaya, 60231, Indonesia
E-mail: nkusumawati82@yahoo.com

Abstract. Coconut fiber, is a by-product of coconut commodities which can be further processed into batik dye material, due to the tannin content it has. However, it is necessary to evaluate the intensity and color fastness produced. In this study an evaluation of the results of the cotton fiber dyeing using coconut fiber material water extract. Utilization of alum mordant and a number of fixers is done to further optimize color quality. The results show the appearance of black, light brown and dark brown shades in the respective cotton fibers for dyeing process which are fixed with iron (II) sulfate, alum and lime. The intensity of the resulting color is in the range 67.83% -86.29%, with the fixer iron (II) sulfate as the producer of the highest intensity and alum as the lowest. The resulting color fastness is on the staining scale 3-4 and 4. This value indicates the great potential of coconut fiber material to be developed as a natural dyeing material for batik.

1. Introduction

Indonesia is a tropical country with an abundance of high natural resources, one of which is coconut commodity. With a total plantation area of 3,544,393 ha, 2,868,686 tons of coconut were produced in 2017 [1]. This abundance has both positive and negative impacts on sustainable development in Indonesia. With a weight that reaches 35% of the total weight, 1,004,040 tons of coconut fibers have been produced. This condition is worrying because most of this material is only used as fuel for cooking, even though coconut fibers can be processed so that they can be of more economic value, one of which is as a natural dye for batik.

In the fresh coconut fiber material contained 3.12% tannin. Tannins are polyphenol compounds that have complex structures. This compound belongs to the quinone pigment group, which is a colored compound with chromophore that consisting of two carbonyl groups conjugated with two carbon-carbon double bonds [2, 3]. As a dyeing agent, tannins will show brown or brown color [4-6]

Although it has many advantages, especially in supporting sustainable development in Indonesia, the use of natural dyes still faces a number of obstacles, especially in the effort to maintain color stability during processing and storage so that color degradation can be avoided. Several factors that affect color stability are temperature, oxygen content, pH and light [7-8]. According to Ref.[9-10], natural dyes are not permanent so they quickly fade when exposed to detergent or sunlight. Therefore, to get good fastness, mordanting and fixation process are needed [10-13].

In natural dyeing, mordanting is needed as an intermediate compound for optimal reactions formed between fibers dominated by cellulose which tend to be negatively charged with natural dyeing compounds that have the same charge tendency. Metal mordants with more than one valence can be
used for this purpose [14, 15]. This study involved the use of K2SO4.Al2(SO4)3 (alum) as a mordanting agent.

The fixation process will allow the complex formation reactions between fibers with dyes and fixers [16]. The presence of this fixer compound also decreases the reactivity of the dye to the material in the surrounding environment, which can potentially change the shades and reduce the intensity and color fastness produced. For this purpose, in this study three types of fixers were used, including K2SO4.Al2(SO4)3 (alum), CaO (lime), and FeSO4 (tunjung). Furthermore, to evaluate the success of utilizing coconut fiber waste as a natural dyeing material, this study will evaluate the effect of the use of alum mordant and the three types of fixers on the color shades as well as the intensity and color fastness that results against wet washing.

2. The material and method

2.1. Material
All chemicals were of analytical grade. The materials used in this study include: (1) the fabric material used is cotton fiber purchased from Lima Jaya store (Surabaya, Indonesia); (2) material for the preparation of cotton fibers, namely Turkish Red Oil (TRO, ≥70%) purchased from CV. Dunia Kimia (Surabaya, Indonesia), alum (Al2(SO4)3.18H2O, ≥17%) purchased from PT. Brataco Chemistry (Surabaya, Indonesia) and soda ash (Na2CO3, ≥48%) purchased from CV. Water (Surabaya, Indonesia); (3) material for cotton fibers dyeing, namely coconut fibers obtained from the local market in Surabaya-Indonesia; (4) materials for fixation, namely iron (II) sulfate (FeSO4.7H2O, d 2.84 g/cm3) and calcium oxide (CaO, ≥90%) which were purchased from PT. Nusa Indah Megah and Mitra Water (Surabaya, Indonesia).

2.2. Washing
Washing procedure is done to reduce the level of contamination of cotton fibers that will be dyed. The success of this procedure will increase the intensity and color fastness produced. This procedure is carried out by washing cotton fibers using TRO. For this purpose, 2.57 g of cotton fiber immersed in 2 g/L TRO for 6 h. The washing procedure at this stage ends with flushing cotton fibers 3 times to remove the remaining TRO and drying in the open air for 24 h [11].

2.3. Mordanting
Mordanting is needed to provide a connecting compound for the reaction between fibers and tannin compounds in coconut fiber extracts. As is true in the washing procedure, the success of the mordanting will increase the intensity and color fastness produced on cotton fibers. Mordant solution was prepared by dissolving 8 g of alum and 2 g of soda ash in 1 L of distilled water. To ensure solution homogeneity, stirring was carried out using a magnetic stirrer for 5 min. The mordant solution is then heated to 100 °C. Mordanting is done by immersing cotton fiber in a 100 °C mordant solution for 1 h. Optimization of the reaction between the mordant and cotton fibers is done by immersing for 24 h without heating. Mordanting is ended by rinsing 3 times using distilled water to remove the remaining mordant residue and drying in the open air for 24 h without squeezed. To further optimize the quality of colors produced, fiber orientation is uniformed through ironing [11].

2.4. Dyeing
The dyeing procedure is carried out to give color to the cotton fibers by supplying specific chromophores by tannin dyes in coconut fiber extracts. Dyeing is done using the pre-mordanting method. The extract of the dye solution was prepared by extracting 100 g of coconut fiber using 1,000 mL of water solvent. The extraction procedure is carried out at a temperature of 100 °C until a concentration twice as concentrated as before. The application of color extracts to cotton fibers follows the previously published procedures [11]
2.5. Fixation
Fixation are performed to reduce the color potential loss from cotton fibers by colors locking. Fixation at this stage was carried out using three fixer compounds, namely iron (II) sulfate, alum and lime. The preparation was carried out by dissolving 50 g of each fixer in 1 L of distilled water. The solution homogeneity is ensured by stirring using a magnetic stirrer for 5 min. The homogeneous solution is stored for 24 h to get the transparent part. The fixation is carried out by immersing 0.85 g of cotton fiber in a 30 mL fixer for 10 min. To ensure the cotton fiber free from the fixer residue, rinsing is done 3 times with each using 100 mL of distilled water. The fixation ends with drying in the open air [11].

2.6. Characterization
To ensure the dominance of tannin in the cotton fibers dyeing with coconut fiber extracts, an electromagnetic radiation absorption analysis was carried out using Pharmaspec UV-1700 UV-Visible Spectrophotometer. The characteristic optimum absorption at a maximum wavelength of 747.5 nm will ensure this dominance. Furthermore, to determine the intensity and color fastness resulting from the application of each fixer, an analysis was performed using Shimadzu UV-2401-PC Diffuse Reflectant Ultraviolet (DRUV) Spectrophotometer. In general, high reflectance values indicate high color intensity [11] to determine the color fastness that is produced, an analysis is carried out using the gray staining scale method, with the interpretation of the assessment as shown in Table 1.

Table 1. Assessment Interpretation Of The Color Fastness Analysis Results Using The Gray Staining Scale Method

| Color Fastness Value | Color Fastness Interpretation |
|----------------------|-------------------------------|
| 1                    | Bad                           |
| 1.2                  | Bad                           |
| 2                    | Not good enough               |
| 2.3                  | Not good enough               |
| 3                    | Fair                          |
| 3.4                  | Good enough                   |
| 4                    | Good                          |
| 4.5                  | Good                          |
| 5                    | Very good                     |

3. Results and discussion
The appearance of maximum absorbance at wavelength 747.5 m, has verified the dominance of tannin in the dyeing by coconut fiber extracts. Specific absorption at these wavelengths triggers $\pi \rightarrow \pi^*$, $\pi \rightarrow \pi^*$ dan $n \rightarrow \sigma^*$ electronic transitions by the C=C and C=O chromophores, which give rise to the tannin characteristic brown color. Visual color of coconut fiber extract at various levels, presented in Figure 1.

Figure 1. Visual color of coconut fiber extract with concentration: (a) 1; (b) 2; and (c) 4 times more concentrated.
Figure 2. Illustration of natural dyeing applied in research

Figure 2 shows an illustration of a series of processes to get good quality natural dyeing. Contamination-free cotton fiber will ensure the optimal presence of mordanting agent as a bridge compound for the reaction with tannin compounds. The molecular structure of cotton fibers dominated by negatively charged hydroxide groups will inhibit the entry of natural dyeing compounds which are also generally dominated by the same charge. This condition will be bridged by the presence of a positive charge with a valence of three provided by alum. The quality of dyeing produced will be greatly influenced by many or at least anionic functional groups of dye compounds which are bound to the Aluminium metal. This is actually the background of the high use of metal chromium complexes with higher valence (six) as mordanting agents for textile and textile products dyeing, even though the negative impacts on health and the environment are very high [17-20].

Fresh coconut fiber contains 3.12% tannin. Tannin as a dyeing agent will cause brown or brown color, where the shades of this brownish color will increase with increasing concentration of the extract. This compound is a quinone pigment with a chromophore content which includes two carbonyl groups conjugated with two carbon-carbon double bonds (see Figure 3). The mechanism of reaction that occurs between cotton fibers, alum mordants and tannin dyes has been published previously [11].

Figure 3. The molecular structure of tannin
The application of three types of fixers in dyeing process has resulted in the appearance of brownish color gradations in cotton fibers, as shown in Figure 4. Meanwhile, in Figure 5 the color shades produced from the dyeing and leveling fixation conditions follow the order of alum-lime-iron (II) sulfate fixation. Dyeing fixation using alum tends to form a light brown color, while the application of lime and iron (II) sulfate successively produces a darker brown color. Compared to the results of dyeing which are not accompanied by fixation, dyeing which is accompanied by the fixation process shows the appearance of different color shades. This shows the influence of specific chromophore group content in each fixer used, namely C=C, S=O and C=N in alum, Ca=O in lime, and S=O in iron (II) sulfate.

In addition to influencing the color shades formed, the use of different fixers will also determine the strength of the resulting color. The results of the color intensity analysis showed the highest color intensity (86.29%) in dyeing that was fixed using iron (II) sulfate, while the lowest (67.83%) was produced from dyeing that was fixed using alum. The size of alum molecules is predicted to be both strengths and weaknesses of this fixer. The variable chromophore content in the large molecular structure of alum has strengthened the appearance of tannin's distinctive color, while at the same time triggering specific obstacles to the reactions that occur between alum and tannin. However, these non-
ideal conditions have proven to be better able to minimize color loss triggered by wet washing operational conditions (Table 1). Even though it shows the lowest color intensity, the use of alum fixers has proven to be able to produce higher compared to lime fixers. This is natural if it is associated with a lower chromophore content in lime and the low inhibition of the dyes interaction with the surrounding environment produced by the lime fixer is thought to be the cause of this.

### Table 2. The Intensity And Color Fastness Of Brown On The Cotton Fibers Resulting From The Use Of A Variety Of Fixers

| Cotton Fiber Sample | Fixer Type | Color Intensity (%) | Color Fastness (scale) |
|---------------------|------------|---------------------|------------------------|
| CF-1                | Alum       | 67.83               | 3-4 (Good enough)     |
| CF-2                | Lime       | 70.49               | 3 (fair)               |
| CF-3                | Iron (II) sulfate | 86.29           | 3-4 (Good enough)     |

The gray scale consists of nine standard pairs of gray plates. Each pair represents a difference in color or color contrast according to the fastness rating with a number on the gray scale. The assessment of color fastness and color change accordingly done by comparing the differences in the samples that have been tested with an original example of the difference in standard color changes illustrated by the gray scale.

### 4. Conclusion

Exploration and standardization of natural batik dyes from coconut fiber waste has been carried out. To obtain stable and homogeneous dyeing quality, with high intensity and color fastness, standard operating conditions have been obtained including stages of washing, mordanting, dyeing and fixing. The results showed the appearance of color shades in the form of brown gradations on cotton fibers after going through the fixation using iron (II) sulfate, alum and lime. Very different color visuals appear when multilevel dyeing and fixing are applied to cotton fiber, where the results of 12 times dyeing by successive fixation procedures using alum, lime and iron (II) sulfate even produce black shades. Dyeing of cotton fibers using extracts of coconut fiber material has produced varying color intensities in the range of 67.83%-86.29%, with the highest intensity produced by iron (II) sulfate and the lowest using alum fixer. Slightly different from the intensity, the color fastness produced by alum fixers is actually higher than that of lime, but still lower than iron (II) sulfate.

### References

[1] Fitriyah H, Ciptandi F 2018 e-proceeding of Art and Design 5 2534
[2] Setiawati E, Haryanti N, Rachmawati A 2013 Pengaruh Usia Sabut Kelapa dan Variasi Metoda Ekstraksi Terhadap Hasil Pencelupan Kapas dan Sutera (Bandung:Sekolah Tinggi Teknologi Tekstil)
[3] Elnoor A A M, Mirghani M E S, Musa K H, Kabbashi N A, Alam M Z 2015 Health Science Journal 12 1
[4] Prayitno E K, Nurimaniwati 2003 Proses Ekstraksi Bahan Pewarna Alam dari Limbah Kayu Mahoni (Yogyakarta: Puslitbang Teknologi Maju BATAN)
[5] Ammayappan A, Mosess J 2007 Man-made textiles in India 50 293
[6] Ali S, Saleemm H, Hussain T 2010 Asian Journal of Chemist 22 7065
[7] Suparmi L L and Prasetyo B 2009 Sains Medika 1 81
[8] Kamel M 2009 AUTEX Research Journal 9 1
[9] Paryanto P A, Kwartiningsih E, Mastuti E 2012 Jurnal Rakayasa Proses 6 26
[10] Samanta A K 2011 Dyeing of Textiles with Natural Dyes (University of Calcutta)
[11] Kusumawati N, Santoso A G, Sianita M M, Muslim S 2017 International Journal on Advanced Science, Engineering, Information Technology 7 878
[12] Ashrafi N 2018 Progress in Color, Colorant, and Coating 11 79
[13] Moniruzzaman M D, Mondal M S, Hossain Md N 2018 The Influence of Mordant and Mordanting Techniques on Ecofriendly Dyeing of Cotton Fabric by Extracted used Tea.
[14] Kusumawati N, Samik S, Santoso A B, Wijiastuti A 2018 Advances in Engineering Research 171 50
[15] Kusumawati N, Mufida L, Annisa R A 2018 Pewarna alami untuk tekstil (Surabaya:Unesa University Press)
[16] Shofwan A 2015 Studi tentang pewarnaan alami bunga belimbing sayur pada kain serat nanas menggunakan fiksator tunjung, tawas dan kapur tohor Skripsi (Malang:Universitas Negeri Malang)
[17] Haar S, Schrader E, Gatewood B M 2013 Clothing and Textile Research Journal 31 97
[18] İslam Ö E and Yıldırım L 2019 The Impact and Prospects of Green Chemistry for Textile Technology57 82.
[19] Har B S and Bharati K A. 2018 Mordants and their applications Handbook of Natural Dyes and Pigments 18 28.
[20] Prabhu K H and Teli M D 2014 Journal of Saudi Chemical Society 18 864

Acknowledgment
We would like to thank the Directorate of Research and Community Service of the Ministry of Research and Technology, Higher Education of the Indonesia Republic for the financial support provided.