VOL. 12, NO. 18, SEPTEMBER 2017 ISSN 1819-6608
ARPN Journal of Engineering and Applied Sciences

www.arpnjournals.com

REUSE OF LIQUID WASTE FROM TEXTILE DYEING WITH NATURAL DYES GAMBIER (UNCARIA GAMBIR ROXB.) FOR COTTON YARN DYEING

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ABSTRACT

Gambier is a kind of sap that comes from compressed extract of leaves and twigs of gambier plants (*Uncaria gambir* Roxb.). The use of gambier is being developed for natural dye of textile products such as weaving yarn. Previous research showed that solution of gambier 5% had been good enough to be used as a textile dye. Gambier residual dyeing solution is liquid waste that still contains active ingredients namely tannins and catechins which still can be reused. This study was aimed to see whether the waste can be reused for dyeing and to see the quality of products. The study was conducted using cotton yarn and mordant Calcium Oxide (CaO) with 2 treatments namely dyeing time (5, 15, and 25 minutes) and kind of liquid waste (1st reuse and 2nd reuse). The utilization of fresh gambier solution was done as a control. The results showed that gambier liquid waste of dyeing still contain tannins up to 52.14% and still can be used as a textile dye. The dyed yarn quality using 1st and 2nd reused dye waste showed that for all treatments, color fastness on washing 40 °C as well as on dry ironing heat were good to excellent (4-5) averagely. The results were not significant compared to yarn quality which was dyed with fresh gambier solution. Color fastness on sunlight in dyeing with 1st and 2nd reuse were moderate until fairly (2-3). The results were lower compared to dyeing with fresh gambier solution that had an average value > 4 (good). Dyeing treatment affected the color strenght value. Dyeing with 1st and 2nd reuse generated a darker color than dyeing with fresh gambier solution. Dyeing in 2nd reuse had the highest color strenght with K/S value 13,810-16,871.

Keywords: reuse, gambier, cotton yarn, tannin, liquid waste.

INTRODUCTION

Gambier is one of prime commodity from Province of West Sumatra Indonesia. It is a potential plant which is widely cultivated to produce tannins and catechins [1], [2]. Gambier is usually traded in form of raw gambier in market which is extracted from leaves and twigs of gambier plant. West Sumatra is the largest producer of gambier in the world. Production of gambier in West Sumatra Provinve amounted to 17,390.77 tonnes in 2015 [3].

The requirement of 80% world gambier come from West Sumatra which is exported to Bangladesh, India, Pakistan, Taiwan, Japan, South Korea, France, and Switzerland [4]. The main content of gambier is catechins and tannins. Tannins in gambier including of unhydrolized tannins, which will polymerize when heated. This principle is used to generate high tannin content [5].

Raw gambier from each farmer has a different tannin content. The highest levels of tannins content is in the product from Siguntur gambier Centre South Pesisir District (25.51%), followed by Halaban Centre (17.49%) and Mungka Centre (16.01%) in Lima Puluh Kota District. All the districts are in West Sumatra Province Indonesia. The amount of tannin content produced is influenced by the amount of the twigs or leaves, extraction time, process method, and drying process of the extract [6].

Tannin in gambier is an active compound that can functionate as natural dyes for textile products. Tannic catechu acid is an anhydrous tannins from catechin in amorphous shape which is soluble in cold water. When it is evaporated will leave a brownish red powder and solution will give a green color with Fe^{+3}. This characteristic shows that tannin content in the sap of gambier is derivatives of phenol.

Failisnur and Sofyan (2014) conducted study about properties of fastness and color strength of silk fabrics with gambier on different of dyeing condition and kind of mordant. The result showed that by using mordants ($Al_2(SO_4)_3$, $CaO_,$ and $FeSO_4$) were found variative colour direction on silk fabrics started from yellow, golden yellow, brownish red, brown, moss green until blackish green. Mordant CaO produced color strenght higher than others [7].

Effect of temperature and dyeing time of cotton yarn on natural dyeing with gambier extract was conducted In another study by Failisnur and Sofyan (2016). The study used three mordants ($Al_2(SO_4)_3$, $CaO_,$ and $FeSO_4$). The results showed that temperature and dyeing time did not effect tensile strength, elongation, and color staining on polyester and cotton, however affected intensity, color brightness, and color change in washing 40 ^{0}C significantly. Mordant CaO gave the highest color intensity than other mordants [8].

Textile industries is developing rapidly today generally use synthetic dyes. It produces a lot of waste water which contains a number of pollutants including toxic contaminants. Most of dyes used in textile finishing derived from petrochemicals. These organic compounds are hazardous and can cause skin allergies and cancer diseases moreover release toxic and harmful by-products during their synthesis [9].

The use of natural colorants in textile dyeing has been a revival of interest. Moreover as a response to current allergic and toxic reactions related to synthetic

dyes, many countries have imposed stringent environmental standards. Thus, there is an increasing interest for the production of natural dyes from plant renewable resources. Gambier is one of natural dye source which is produced and cultivated throughout the year [7].

Gambier has been developed nowadays as natural dye for textile products such as weaving yarn and batik (traditional cloth in Indonesia). From previous research, solution of gambier 5% has been good enough to be used as textile dye. Residual of dyeing solution from gambier is liquid waste that still contains active ingredients namely tannins and catechins which still can be reused.

The use of gambier as dye for textile products will generate residual of gambier solution as liquid waste which can cause pollution. It is required efforts to utilize the waste generated become a useful resource to create an environmental friendly industry. The gambier solution of residual dyeing still contain active ingredients visually still be used for dyeing. There are no studies that examine the effectiveness of utilization of the liquid waste whether they can be used as a dye or not.

MATERIALS AND METHODS

Material and equipment

The following material were used in this study: liquid waste of textile dyeing from gambier, raw gambier, cotton yarn, mordant CaO, soap. The process of dyeing and mordants used equipment dye bath. Testing of color strenght used UV-Visible spectrophotometer (Premiere Colorscan SS 6200, CIELAB (1976)/D65, Navi Mumbai, India). Observation of color fastness used crockmeter, laundry meter, grey scale, and staining scale.

a) Bleach process of yarn

Yarn was soaked in soap solution for bleaching process and removes starch or wax before dyeing process with the dye gambier. The yarn was then washed until all the soap was gone, then dried, and ready to be dyed with a dye gambier.

b) Reextraction of raw gambier

Reextraction of raw gambier solution 5% was carried out for dyeing in fresh gambier solution. The reextraction was conducted through the stages: refining, dissolving with hot water ±90 ^{0}C, decantation, and filtration. The filtrate was used as dyeing solution.

c) Liquid waste preparation

Remaining of fresh gambier solution 5% which had been used in the initial dyeing process was liquid waste for dyeing 1st reuse. The liquid waste was used after 2 weeks of storage. The 1st reuse residual waste solution was used for 2nd reuse dyeing. The liquid waste was used after storaging for 2 months. Liquid waste was heated up to 80 °C before using, then allowed to room temperature and ready to be used for dyeing.

d) Preparation of CaO mordant

CaO Mordant solution was made by dissolving 50 g/L and precipated, the filtrate was used for mordant.

e) Dyeing process and mordant

The study was done by using liquid waste from yarn dyeing using natural dye gambier. Liquid waste was heated up to 90 ^{o}C and then allowed to room temperature before using. Yarn was bleached before using. The bleached yarn was dyed in liquid waste in accordance with dyeing time treatments (5, 15, and 25 minutes). The dyed yarn was dried under a shade place. Dyeing process was done 3 times. The dried dye yarn was mordant using CaO mordant. Mordant process was done by post mordant method for ± 10 minutes.

f) Finishing process

Finishing process was conducted through soaking the dyed fabric in hot water 60 ^{0}C for 10 minutes and continued with water washing repeatedly until the faded disappear.

g) Evaluation/Observation

1) Color Intensity: The evaluation was conducted using Minolta Spectrophotometer CM 3600d (Tokyo, Japan) on iluminan D65 observer standard position 10^0. Quantitative value of color intensity was obtained by measuring the percent of reflectance (% R) at the same wavelength and then converted to the value of K/S using table of K/S Kubelka-Munk (equation 1).

$$K/S = \frac{(1-R)^2}{2R} = aC \quad (1)$$

K is the absorption coefficient (light absorbed), S is the diffused light coefficient, C is the hue value, and R is the percent reflectance value at a specific wavelength (λmax).

2) Color Fastness: Test of color fastness on washing 40 ^{0}C is according to Indonesia National Standard (SNI) 0276-2009, ISO 105-C06-2010, A02, 2010; A03, 2010. Resistance on day light is according to ISO 105-B01-2010; A02, 2010. Resistance on heat suppression is according to ISO 105-X11, 2010.

RESULTS AND DISCUSSIONS

a) Analysis of raw gambier, fresh gambier solution 5% and liquid waste of dyeing

The anaysis of raw gambier, fresh gambier solution 5%, and liquid waste of dyeing were used in this study as shown in Table-1.

Table-1. Analysis of raw gambier, fresh gambier solution 5% and dyeing liquid waste.

No.	Sample	Results			
		Ph		Density	Tanin (%)
		Initial	Final		
1	Raw gambier	-	-	-	28.51
2	Fresh gambier solution 5%	3.21	4.91	1.02	50.20
3	Liquid waste of dyeing (1st reuse)	4.30	5.58	1.02	50.11
4	Liquid waste of dyeing (2nd reuse)	4.82	4.94	1.03	52.14

Tannin content in raw gambier is 28.51%. Tannin is a compound that can funcionate as a dye. Tanin content in Fresh gambier solution 5% increased became 50.20%. The content was higher than raw gambier. This was caused by the heating of the solution before dyeing. The characteristic of tannin is soluble in hot water. Heating will cause tannic catechu acid will plymerize into tannin. Tannin content in 1st and 2nd reuse liquid waste of dyeing was still high 50.11% and 52.14%. The high of tannin content caused this liquid waste still can be used for dyeing.

Gambier solution is acidic because it contains tannic catechu acid. Initial pH was the condition before dyeing and final pH was the condition after dyeing. The pH solution after dyeing increased due to the influence of the yarn used. Yarn was bleached and washed using soap before dyeing. This treatment caused the yarn was alkaline.

b) Liquid waste in dyeing process with gambier

The following data is explained the amount of liquid waste which was generated from a dyeing process of yarn with gambier. For dyeing 285 grams cotton yarn in 5000 ml gambier solution 5% was generated 4562 ml liquid waste. The average amount of gambier solution 5% which was absorbed by 285 gram yarn about 438 ml. The average amount of gambier solution 5% which was absorbed by 1000 grams yarn about 1540 ml averagely. The remaining gambier solution 5% was an effluent which could be reused as a dye.

c) Color coordinate and color directions

Color directions of dyed yarn for all treatments were similar visually brown as shown in Table-2. The color intensity was measured using the CIE Lab scale. Scale a* indicates the direction of red color, while scale b* indicates the direction of the yellow color.

Table-2. Coordinate values and color shades of cotton yarn at color system cie 1976 on various treatments.

Treatments	Dyeing Time (min)	L*	a*	b*	Color Shades
Fresh gambier solution 5%	5	48.372	17.356	28.709	
	15	48.958	17.452	29.606	
	25	49.364	17.604	30.076	
Liquid waste of dyeing 1st Reuse	5	45.881	18.098	30.066	
	15	47.062	18.550	31.436	
	25	47.218	18.598	31.896	
Liquid waste of dyeing 2nd Reuse	5	40.650	17.900	27.550	
	15	40.710	17.570	26.800	
	25	38.780	17.990	27.340	

Colors produced in this study are brown with different color intensities. Mordant has importan role in determining the direction of color produced. Mordant CaO and gambier produced a reddish-brown color [7]. One of the problems in the use of natural dyes for textiles is limited shades produced [10], [11].

Data in Table-2 shows that the treatment of dyeing using waste affected the color brightness (lightness) of yarn produced. Results of color brightness values can be seen from the value of L*. Dyeing with fresh gambier solution or initial immersion had color brightness value higher than dyeing in 1st reuse or 2nd reuse. The color brightness was between 48.372 - 49.364. It means that the resulting brown color lighter than other treatments. Dyeing in 1st reuse had color brightness value between 45.581 - 47.218 and dyeing in 2nd reuse had the lowest brightness color between 38.780 - 40.710.

The lower brightness color values the darker resulting brown color. The color of 1st and 2nd Liquid waste of dyeing darker than Fresh gambier solution 5% visually. Tannin content might be affect the color. The increasing of tannin content in liquid waste caused color darkness of yarn was higher than dyed yarn in Fresh gambier solution 5%. The color of dye solution also influenced the color darkness. The 1st and 2nd reuse liquid waste of dyeing had darkness color more than fresh gambier solution 5% visually as shown in Figure-1. The darkness of color caused by repeat heating before dyeing. Repeat heating caused tannin in solution would be

polymerized. The 2nd reuse liquid waste was heated more and had color darkness more than others.

From the data, the color difference tolerance values ± 1, the result that none of the samples met the criteria received or passed on measurement of color differences with the absolute value system CIE 1976 color differences (AE a*b*). With these systems, different color values are calculated as a resultant from color coordinates in color space, L* (lightness), and a* & b* (color direction) as indicated in Table-2.

Dyeing time gave a significant effect on the color brightness. It can be seen from data L* as shown in Table-2. The value of L* increased with the raising of dyeing time in each treatment, but the increase was not too far. The longer the dyeing time the more dye which was absorbed into the yarn fibers [8].

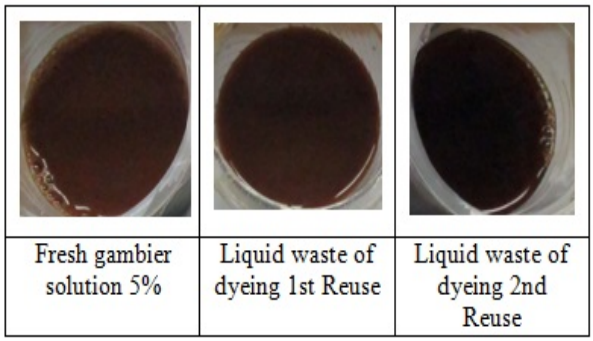

Figure-1. Color darkness of dyeing solution.

d) Color strength

Measurement of color strenght was done by using iluminan D65 at standard position observer 10". All samples were analyzed at a wavelength 420 nm. Determination of maximum wavelength was obtained from statistical analysis. The light intensity was measured by percent reflectance value (% R) which indicated the amount of light reflected on the modification process of light by sample.

Color intensity in the form of color darkness was measured by K/S value. % R value was converted to the K/S value according to Kubelka-Munk theory to provide a quantitative value of colors darkness on the same wavelength. % R value and K/S form (uncolored yarn) was measured as a control, and to give the K/S value of dye without light refraction by the material. The K/S values are displayed in Table-3.

The test results showed that dyeing treatments affected color strenght value of dyed yarn. Dyeing in 1st reuse and 2nd reuse produced a darker color than initial dyeing using fresh gambier solution 5%. The darker color the higher K/S value. The data as represent in Table-3 shows that from all the dyeing treatments, the 2nd reuse dyeing had color the highest strenght value with K/S value between 13,810-16,871. Dyeing in 1st reuse and fresh gambier solution had K/S value between 10,723 – 15,958 and 7,718-9,227 respectively.

Table-3. Color streght value (k/s) of cotton yarn at various treatmens.

Dyeing time (min)	K/S		
	Fresh gambier solution 5%	1st Reuse	2nd Reuse
5	8,295	10,723	14,544
15	7,718	14,889	13,810
25	9,227	15,958	16,871

Dyeing time also influenced color strenght in each treatment. It raised generally with the increasing of dyeing time.

e) Color fastness on washing 40 ºC

The analysis of color fastness on washing 40 °C can be seen in Table-4. The results show that the value of color change in Fresh gambier solution 5% dyeing generally good [7]. Dyeing in 1st reuse and 2nd reuse appeared the increase of the value which was 4-5 (good to excellent) averagely. Color fastness on washing (color change) of dyed yarn in 1st reuse and 2nd reuse was higher than fresh gambier solution. Dyeing time did not gave significantly effect on color fastness on washing. All the values are similar for each time treatment as indicated in Table-4.

Table-4. Color fastness on washing 40 ^{0}C of cotton yarn at various treaments and dyeing time.

Treatments	Dyeing Time (min)	Color fastness on Washing 40^{0}C		
		Color change	Color staining	
			Polyester	Cotton
Fresh gambier solution 5%	5	4	4-5	4-5
	15	4	4-5	4-5
	25	4	4-5	4-5
1st Reuse	5	4-5	4-5	4-5
	15	4-5	4-5	4-5
	25	4-5	4-5	4-5
2nd Reuse	5	4-5	4-5	4
	15	4-5	4-5	4
	25	4-5	4-5	4

Problems in using of natural dyes is inadequate of color fastness [10], [11]. The problems can be overcome by using a metal salt or a mordant which produces affinity between fabric and dye [12], [13], [14]. The use of mordant CaO in this study was very important in producing good color fastness.

Color staining on polyester had the same value for all treatments 4-5. Color staining on cotton yarn appeared the difference in treatment 2nd reuse which was an average 4. This value was lower than the dyeing in fresh gambier solution 5% and 1st reuse with an average value of 4-5.

f) Color fastness on ironing

The analysis results of color fastness on ironing (direct color change, colors change after 4 hours, and staining on cotton) as indicated in Table-5.

Table-5. Color fastness on ironing (dry) at various treatment and dyeing time.

Treatments	Dyeing Time (min)	Color fastness on ironing		
		Direct color change	Color change after 4 hours	Staining on cotton
Fresh gambier solution 5%	5	4-5	4-5	4-5
	15	4-5	4-5	4-5
	25	4-5	4-5	4-5
1st Reuse	5	4-5	4-5	4-5
	15	4-5	4-5	4-5
	25	4-5	4-5	4-5
2nd Reuse	5	4-5	4-5	4-5
	15	4-5	4-5	4-5
	25	4-5	4-5	4-5

The values were good to excellent (4-5) for all treatments. This indicated that dyeing treatment in 1st reuse and 2nd reuse did not affect the color fastness on ironing. Dyeing time did not gave significantly effect on color fastness on ironing. All the values are similar for each time treatment as indicated in Table-5.

g) Color fastness on sunlight

Testing of color fastness on sunlight in 1st reuse and 2nd reuse dyeing on average 2-3 as shown in Table-6. This values decreased and were lower than dyeing in fresh gambier solution 5% with average value higher than 4. This indicated that the use of liquid waste gambier affected the color fastness on sunlight. This might be caused by complex reaction of mordant and dye did not perfect. Dyeing time did not influence color fastness on sunlight. Dyeing time did not gave significantly effect on color fastness on sunlight. All the values are similar for each time treatment as indicated in Table-6.

Table-6. Color fastness on sunlight of cotton at various treatments and dyeing time.

Dyeing time (min)	Color fastness on sunlight		
	Fresh gambier solution 5%	1st Reuse	2nd Reuse
5	> 4	2-3	2-3
15	> 4	2-3	2-3
25	> 4	2-3	2-3

Natural dyes generally have low color fastness on sun lighting [15], [7], [16], [17]. Degradation of natural dye chromophore component By photo-oxidative, producing small molecules which losses color on textile [18]. Mordant process will form complex which protect chromatophore from photolitic degradation and improve color fastness on sunlight [16].

CONCLUSIONS

Liquid waste of gambier dyeing still contained tannins up to 52.14% and could still be used as a dye for textile products. The analysis results of color fastness on washing indicated that the color change in fresh gambier solution 5% were generally good. The color fastness on washing increased in treatments 1st and 2nd reuse with an average value 4-5 (good to excellent). The color fastness on ironing (direct color change, color change after 4 hours, and staining on cotton) averagely was good to excellent (4-5) for all treatments.

Color fastness on sunlight in 1st and 2nd reuse dyeing was 2-3 averagely. This value was lower than dyeing in fresh gambier solution 5% with average value more than 4. This indicated that the use of liquid waste affected the color fastness on sunlight. Dyeing in fresh gambier solution 5% had a brightness value higher than in 1st or 2nd reuse. The treatments affected the value of color darkness of dyed yarn. Dyeing in 1st and 2nd reuse produced a darker color than dyeing in fresh gambier solution 5%. It can be seen from the highest value of K/S in 2nd reuse which had the highest color darkness.

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