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Study of Cotton Fabric Dyeing Process With Some Mordant Methods By Using Gambier (*Uncaria gambir* Roxb) Extract

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***Abstract*— Natural dye that was re-extracted from raw gambier is used to dye cotton fabrics. Aluminum sulfate, calcium oxide, and ferrous sulfate were used as mordants. Dyeing had used four different mordant methods which namely pre, simultaneous, post, and combined (pre and post) mordant. The chemical components in dry gambier extract were tested by using X Ray-Diffraction. The dyed cotton fabrics were evaluated by their color strength (K/S), color difference values (L*, a*, and b*), fastness to washing, rubbing and light. The results have shown that the main components of gambier were anhydrous catechins, catechins, and pyrocatechol. The use of post mordant and combined mordant methods with calcium oxide mordant had produced higher color strength (K/S) than others. The fastness to washing and rubbing values were in a range from good to excellent, while the average of the fastness to light was in a range from moderate to good. The amount of mordant metal that was bound to the fabric was between 15-40%.**

***Keywords*— natural dye; gambier; cotton fabric; the method of mordant; color properties**

I. INTRODUCTION

The color of textiles has an aesthetic and a functional value that can determine the identity of the user. Most of the dyeing materials used the synthetic dyes because they have a wide color range, more stable, and can be repeated accurately [1]. In recent years, the limitation of chemical use, awareness of environmental safety, and tendencies of natural lifestyle have generated significant interest in the use of natural dyes [2].

Natural dyes are eco-friendly and have many advantages compared to synthetic dyes. Applications of natural dyes are increased because of the biodegradability properties and high compatibility with the environment [3]. Fabrics with natural dyes have soft, low toxic, hypoallergenic colors, and antibacterial potential [4]–[6].

Plant of colorants source that grows in Indonesia are quite abundant, but they were not all explored yet. The colors produced are depended on the type of plant, and mordant used [6]–[8]. Gambier (Uncaria gambir Roxb) is one of the potential industrial crops and are produced in various regions especially in West Sumatra by the form of raw gambier. The tannin compound in Gambier with the form of catechu-tannic acid is the main component (20-55%) [9]. Catechu tannic acid is an anhydrate of catechins [10]. The raw material, the way of processing, and the presence of impurities during the processing determine the amount of tannin content [5], [10]. Tannins are easily bonded with protein and cellulosic fibers because they contain some hydroxyl groups. The H atom in the hydroxyl group is highly reactive and may form the hydrogen bonds, potentially which is being used as a textile dye.

The dyeing process of textiles with gambier requires the mordant metal to raise its color. It can also improve the fastness of the dyed fabric color. Some ecologically safe mordants and more eco-friendly impacts among others are aluminum sulfate, calcium oxide, and ferrous sulfate [11]–[13]. The types and methods of mordant will result in a variety of colors, brightness, and coloring properties of different dyed cotton fabrics [14]–[16].

Research on the utilization of gambier and gambier liquid waste as textile dyes has been widely applied [13], [17]–[22]. However, there is no information about the explanation of the effect of some mordant methods in increasing the color strength and fastness of the dyed cotton fabrics by using gambier dye. This study was aimed to determine the components in gambier by using X Ray-Diffraction and the characteristics of the dyed cotton fabric with some methods and eco-friendly types of mordant.

II. MATERIAL AND METHOD

A. Material

The main material was Gambier. It was raw Gambier that came from the local farmer. The cotton fabric had passed through a degumming process that came from the textile industry PT. Primisima, Yogyakarta in Indonesia. Three types of mordant which namely aluminum sulfate ($Al_2(SO_4)_3$), ferrous sulfate ($FeSO_4$), and calcium oxide (CaO) came from a supplier CV. Bratachem.

Dyeing and the mordant process had used dye bath with temperature control and time. Spectrophotometer Premiere Colorscan SS 6200 which used to color coordinate CIELab (1976/D65) L*, a*, b* and color strength (K/S) from a dyed sample with D65 illuminant at 10-degree observation. The analysis of the color-fastness on washing and rubbing had used the laundry meter and the crock meter.

B. Reextraction of Raw Gambier

Re-extraction of raw gambier used the water solvent to obtain tannin. Elimination of residue from the extract had used 400 mesh filter [10]. The next step was precipitation for 8 hours to diffuse tannin compound optimally into solution, and to precipitate the catechin compound. The supernatant was used as fabric dyes. A part of the supernatant was dried for analysis of chemicals components by using X-Ray Diffraction (XRD).

C. The process of Mordant and Dyeing

The mordant process was conducted by the method of pre, simultaneous, post, and combined (pre and post) mordant. The pre mordant is for the soaking of the cotton fabric before the dyeing process with coloring extract. The simultaneous mordant is a mordant solution mixed with dye extract, and the fabric is dipped into it simultaneously. The post mordant is for the soaking of the cotton fabric in mordant solution conducted after the dyeing process [23]–[25]. The combined mordant method is a combination of pre and posts mordant. The mordant concentration was 3% with ratio 1:20 (cotton fabric weight/dye volume) [23].

Optimization of the dyeing process had used the gambier natural dyes with a concentration of gambier 5% at ratio weight/volume 1:30. Dyeing was conducted at temperature 70^oC for 30 minutes [13], [26]. The cotton fabric was then dried in a shady place because some mordants would be very sensitive to light [27].

D. Washing Process

The dyed cotton fabric was then immersed in hot water (± 60^0C) for approximately 5 minutes, and after that washed with the water. This process was aimed to improve the color fastness, especially in washing treatment.

E. Observation

1) Identification of Chemical Components: Identification of components had used X-Ray Diffraction PANalytical X'pert Pro. The resulting X-ray was reflected by the material at a particular wavelength at a reflection angle of 2θ so that the reflected intensity could be detected. The test was aimed to confirm the presence of tannin components based on X-Ray diffraction measurements at wavelengths from 2000-6000 cm^1, the position of goniometer angle 2θ.

2) Evaluation of Color Strength: The dyed cotton fabrics with gambier extract which processed in this study were subjected to the reflectance of the color measurements by using a visible Spectrophotometer method CIE-Lab (1976)/D65) with the light reflection technique of "Premiere Colorscan" SS 6200. The quantitative value for the color strength was obtained by measuring the percent value of reflectance (% R) at the same wavelength and then converted to K/S value with K/S table assistance by Kubelka-Munk theory (equation 1). Reflectance value is the reflection of light amount reflected by objects that contain color.

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

K is the absorption coefficient (absorbed light); S is the spread light coefficient; R is the percent value of the reflectance (λmax) [1], [28].

The color coordinates of the CIE-Lab L* a* b* system and their position in the color space using the Hunter Lab Color Flex have shown the color difference values. The color coordinate values L* for color brightness (brightness, 100 = white, 0 = black), redness (+a*), greenness (-a*), yellowness (+b*), and blueness (-b *) [28], [29].

3) Colorfastness Properties: To evaluate the dyeing performance, the color-fastness on washing had use SNI ISO 105-C06-2010, rubbing SNI ISO 105-X11, 2010, and light SNI ISO 105-B01-2010. The test used the Laundry meter and the Crockmeter instrument. The color-fastness value was evaluated by comparing with the grey scales standard for color changes and the staining standard for the color staining on another kind of fabric.

4) Observation of The Absorption of Metal Mordant: Test of the absorption of the mordant metal by using SNI 19-2896-1992. The test was aimed to know the amount of the absorption and bounded mordant metal as a multiplex reaction of fiber-mordant-tannin in the dying process. The test was done by the spectrophotometric method to the fabrics before and after the dyeing process.

For analysis, the absorption of the mordant metal had used the Atomic Absorption Spectrophotometer (AAS) GBC 932 AA at wavelength 396.2 nm for the aluminum metal, 422.7 nm for the calcium metal, and 372 nm for the iron metal. The amount of the absorbed metal was expressed by the weight (mg) of the metal per weight (g) that the cellulose fiber used.

Preparation of the sample had used the dry destruction in a muffle furnace with temperature $400\text{-}800^0C$. There were some stages for the test which namely preparation of the standard, treatment preparation, selecting of resonance line, optimization of instrument condition, absorbance reading of standard solution, absorbance reading of treatment solution, and absorbance interpolation of treatment solution on a linear curve.

III. Results and Discussion

A. Identification of Gambier Dyes Components

The components of gambier dyeing powder were identified by using X-Ray Diffraction (XRD) (Fig 1). Each peak had a different intensity. The XRD pattern had shown several angles with the high intensity at 2θ = 16.5^0; 19^0; 23.5^0; 26^0; 27.5^0 and 28.5^0 which identified as catechin ($C_{15}H_{14}O_6$), anhydrous catechins (tannins) ($C_{15}H_{14}O_6.H_2O$) and pyrocatechol ($C_6H_6O_2$). The highest intensity had occurred at position 23.5^0, which identified the anhydrous catechin (tannin) compound.

The anhydrous catechins are condensed tannins with amorphous structures and provide a reddish color [30]. The chemical structure of tannin is in Fig. 2. The pyrocatechol is a derivative of catechins, which is usually used in medicine as therapeutic.

B. Color Measurement of Cotton Fabric

Color strength measured by the K/S value is the amount of dye attached to the dyed cotton fabrics. The more fiber bound the dyes, the more absorption of light by the dye particles. On the contrary, the amount of light reflected and captured by the detector (R) is getting smaller [31]. From the observation, the color strength and coordinate value of the dyed cotton fabric as shown in Table 1. The large K/S value represents the high color strength in each treatment. The color strength of an object can be compared with each other if it is on the same wavelength spectrum [32].

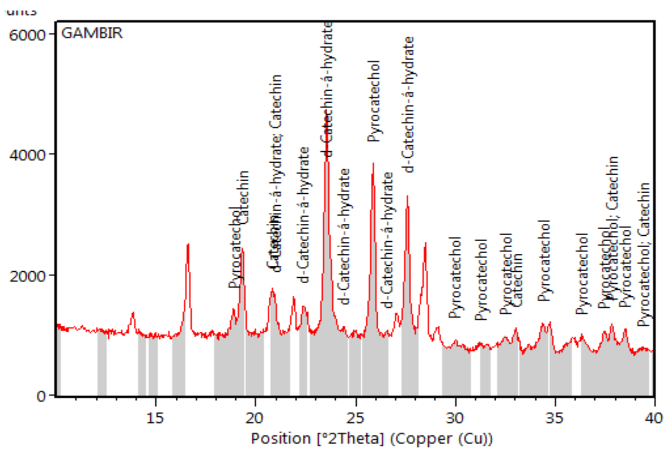

Fig. 1 Analysis of x-ray diffraction of gambier extract

TABLE I
RESULTS OF DYEING WITH GAMBIER WITH TREATMENT OF MORDANT METHOD AND MORDANT KIND

Mordant Method	Mordant Kind	K/S	Results of Dyeing				
			L*	a*	b*	Color Obtained	Color Name [a]
Pre Mordant	$Al_2(SO_4)_3$	2.98	66.82	7.97	24.39		Moderate Red 5R 5/4
	CaO	1.86	68.76	9.12	16.73		Light Brown 5YR 5/6
	$FeSO_4$	5.85	44.42	3.26	11.09		Grayish Red 5R 4/2
Simultaneous Mordant	$Al_2(SO_4)_3$	2.48	67.88	7.62	24.05		Light Brown 5YR 6/4
	CaO	2.44	62.96	3.97	17.53		Moderate Brown 5YR 4/4
	$FeSO_4$	9.87	3635	2.41	11.35		Brownish Black 5YR 2/1
Post Mordant	$Al_2(SO_4)_3$	2.21	68.24	8.12	21.06		Light Brown 5YR 6/4
	CaO	15.15	45.03	18.66	33.27		Moderate Reddish Brown 10R 4/6
	$FeSO_4$	6.78	41.69	2.50	11.30		Grayish Brown 5YR 3/2
Pre and Post Mordant	$Al_2(SO_4)_3$	2.87	64.13	7.73	22.09		Light Brown 5YR 6/4
	CaO	20.38	36.89	20.47	28.89		Dark Reddish Brown 10R 4/3
	$FeSO_4$	13.85	31.39	3.55	10.72		Brownish Black 5YR 2/1

[a]) Color name is based on Munsell Soil Color Chart

The use of $Al_2(SO_4)_3$ provides the lowest color strength for all mordant methods. Aluminum metal forms weak coordination complexes with dye compared to iron metal [24]. Iron metal produces the highest color strength for the pre and simultaneous mordant methods, while CaO optimum is used in the post and the combined mordant methods. Strong coordination tends to increase the interaction between the fiber and the dye so that the color absorption increases [24]. Dyeing process with the post and the combined mordant methods have given a higher color strength value averagely because of a strong bond of complexity that had formed between the dye-mordant-cellulose fibers. It connects the bond of dyestuff with the fibers so that the dyestuff affinity may protect against the fibers to increases. The diffused dye that goes into the fiber core bound by a chemical bond that is the hydrogen bonding of the carboxyl group of fabric fibers, the ionic bond between anionic and cationic in the tannin group (carboxyl group) or covalent bond by an interaction between tannin and reactive group in the fibers [29]. The mechanism of complexity that occurs among the cotton fiber, mordant, and dye as in Fig. 3.

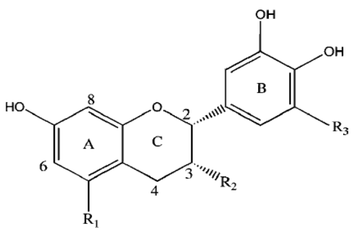

Fig. 2 Chemical structure of tannin [33]

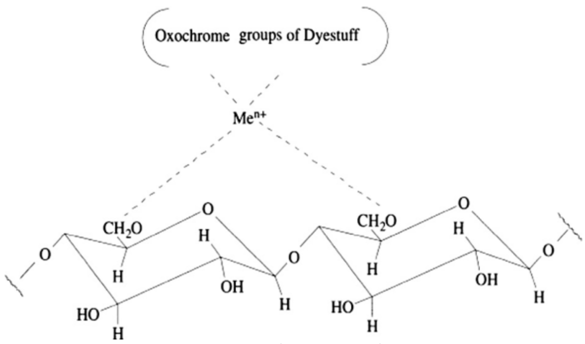

Fig. 3 The complex mechanism of cellulose fibers and dyes with Fe ion metal [34]

Based on the color measurement for cotton fabric, the maximum wavelength is at 400 nm (Fig. 4). It means that the maximum light absorption in the visible spectrum is at wavelength 400 nm. Visually the overall treatment has different color direction characteristics (Table 1). However, the maximum absorption value of light is at the same wavelength spectrum (400 nm) (Fig. 4). The direction of color is in the same shade. The difference is only in the color strength that is lighter, medium, or darker.

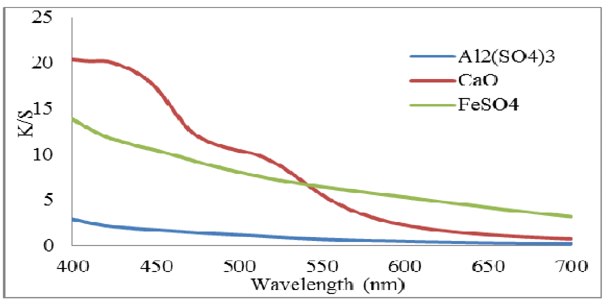

Fig. 4 K/S value of cotton fabric with the combined mordant method

The visual color of the research is three colors which namely yellowish, reddish brown, and darker brown. Treatments that used $Al_2 (SO_4)_3$ mordant, color measurement with the spectrophotometer produces color direction in color coordinates (a,b) with the dominant value in b. It shows that the color strength of the treatment is yellowish. The interaction between the aluminum metal cations of $Al_2 (SO_4)_3$ with dye molecules produces yellowish color.

The use of Al metal as a mordant produced yellowish brown color in the cotton fabric. The same result of a dye of *Juglans regia* L [16], chestnut leather [35], mangrove bark [36], and *mangosteen* peel [3]. Dyeing with orange peel [37], and jackfruit peel [38] has given a yellowish color. The dye of *Bixa Orellana* produces a yellowish red color [2]. The highest color strength of Al metal usage is obtained in the pre-mordant method.

In the treatment of used CaO mordant, the result of the color measurement by spectrophotometer had shown the visual color at coordinate (a, b) with dominant value in b. The fabric color of treatment is more reddish than others. This result in the previous research using the *mangosteen* peel [3], while dying of jackfruit peel produces yellowish color direction in fabric [38]. The highest color strength of Ca metal usage was in the combination followed by the post-mordant method.

In the treatment that used $FeSO_4$ mordant, the color of fabric results in dark brown with the lowest brightness level. It was due to the presence of iron cations that interactive with the dying is high enough. Consequently, when used as a mordant, the intensity of the color is stronger. The evidence from the high absorption of $FeSO_4$ metal used as in Table 4. The result of the color was dark green (blackish green) when visually viewed. The condensed tannin will give the blackish green color when reacts with Fe metal ions [39]. Compared to other natural dyes such as *Eucalyptus* and *Mimosa Tenuiflora* are produced in grayish brown [34]**.** The highest color strength of Fe metal usage obtained in combination is followed by the simultaneous mordant method.

C. Fastness Dyeing to Washing, Rubbing, and Light

1) Fastness to Washing: The results have shown that the color changes before and after washing. The color fastness in washing is from sufficient (3) to good (4) (Table 2). The value of color staining against other fabrics (cotton and polyester fabrics) for all treatments are from good (4) to very good (5). The high value of color changes has indicated that the wear off dye from the fabric is small. It is due to the tough bond of complexity that occurs in the dying and mordant process so that it is not released when the washing is finished.

The treatment of the mordant type $Al_2 (SO_4)_3$, CaO and $FeSO_4$ has given almost the same fastness value. While in the treatment of its method (pre, simultaneous, post and combined), there is a change of color fastness. The pre and the simultaneous mordant were a moderate value (3), the post and the combined mordant were good value (4). The physical bond between the natural dye and the cellulose fiber in the pre and simultaneous method is weak. Consequently, when the washing treatment is finished, the water-soluble dye can be easily detached from the fabric fiber.

The ionized cellulose polymer forms a negative charge and interacts with the mordant on the dyeing fabric. The positively charged metal seems to bridge the fiber with the dyestuff when coloring of natural dye with a negative charge in water. As a result, when obtaining mechanical treatment or washing the dyestuffs are retained in the fibers [40].

2) Fastness to Rubbing: Testing of rubbing fastness was done by using the Crockmeter. The rubbing fastness of the dyeing cotton fabrics are in a range from good to excellent (4-5) (Table 3). In the treatment using $Al_2(SO_4)_3$ and CaO mordant, the color fastness value is from good to excellent (4-5). The high color fastness value in the presence of mordant process occurs because the metal cations of the Ca, Fe with 2 or 3 valencies are reacted to form a metal complex bond with the dye into a large molecule. The larger the molecule size, the bigger the bonding force that occurs between the fibers and the dyestuffs. Consequently, when the fixed dye undergoes any mechanical power on the fabric surface, the dyestuffs will be more retained on the fabric fibers [24].

3) Fastness to Light: The fastness to light of the dyed fabric with natural dyes is less than others such as washing and rubbing because it is very easy to degrade by the influence of light. The color fastness value of the dyed cotton fabric as in Table 3.

Internal factors of the physics and chemistry of dyes such as dye concentration, properties of fiber, and mordant influenced the color fastness to light. Nearly 50% of the natural dyes are listed in the Color Index (CI), which derived from the flavonoid compounds, and the rest are *anthraquinone, naphthoquinone*, and indigo. The flavonoid compounds such as tannins have less color-fastness to light, but *anthraquinone* and indigo are good [41]. In the coloring process using *mordant*, the color fastness depends on the type and method of mordant. The complexity that occurs between the dye-mordant-fiber will provide the color stability to the light influence.

The loss of color in the dyed textiles by the influence of light can be attributed to degrade the natural dye *chromophore* components by photo-oxidative degradation, resulting in small molecules [41], [42]. Natural dyes tend to absorb the visible light. The radiation energy of these rays causes photochemical reactions that can damage the dyestuff structure resulting in color degradation.

TABLE II
FASTNESS TO WASHING OF THE COTTON FABRIC

Mordant Method	Kind of Mordant	Color Staining		Color Change
		Cotton	Polyester	
Pre Mordan	$Al_2(SO_4)_3$	5	4	3
	CaO	5	5	3
	$FeSO_4$	5	5	3
Simultaneous Mordant	$Al_2(SO_4)_3$	5	5	3
	CaO	5	5	3
	$FeSO_4$	5	5	3
Post Mordant	$Al_2(SO_4)_3$	4	5	4
	CaO	5	4	4
	$FeSO_4$	5	5	4
Pre and Post Mordant	$Al_2(SO_4)_3$	4	5	4
	CaO	4	5	4
	$FeSO_4$	5	4	4

In color degradation, there is a decrease in the concentration of *chromosphores* and some detected *hydroxybenzoic* acid. The color of the fabric becomes visually faded. This event is related to the oxidation process of C2-C3 atoms of the flavonoid compounds, followed by the breaking of C2-C3 and C3-C4 chains. Increased oxidation of flavonoids is caused by the effect of some activated radicals with the light on double bonds C2-C3 [43]. Metal salts in the mordant can also speed up the photo-oxidation on the dyed fabric in the dyeing process. Some mordants are catalyzed by the degradation of natural dyes and are very sensitive to the effects of light [27], [43].

TABLE III
FASTNESS TO RUBBING AND LIGHT OF COTTON FABRIC

Mordant Method	Kind of Mordant	Rubbing	Light
Pre Mordan	$Al_2(SO_4)_3$	4-5	3
	CaO	4-5	2
	$FeSO_4$	4-5	4
Simultaneous Mordant	$Al_2(SO_4)_3$	4-5	3
	CaO	4-5	2-3
	$FeSO_4$	4	> 4
Post Mordant	$Al_2(SO_4)_3$	4-5	2-3
	CaO	4-5	2-3
	$FeSO_4$	4	4
Pre and Post Mordant	$Al_2(SO_4)_3$	4-5	2-3
	CaO	4-5	3
	$FeSO_4$	4	4

D. Amount of Used Mordant Metal

The absorption of the mordant metal in dyed cotton fabrics is to evaluate the actual amount of metal ions, which has left in the fiber after the dyeing process. The result is an indication of complexity among the dyes-metal ion-fiber. Amount of the mordant metal is used to determine the color strength and the color-fastness of the dyed cotton fabrics. The test result of the mordant amount left in the fabric fibers as shown in Table 4.

Method of the post and the combined mordant averagely bind the metal more than others. The average percentage of mordant, which is absorbed, is still low (15-44%) as in Table 4. This condition indicates that the complexity that occurs among the metal ion-tannin-fiber is still low, or the number of the metal ions in the solution exceeds the quantity of complex ligament with the fibers. Another possibility is that the absorbed dyestuff is still on the fiber surface and has not penetrated in the fiber core yet, so it is easy to release into the water during the washing process.

The amount of the mordant metal contained in the dyed fabric also determines the color intensity of the resulting color (Table 1). FeSO4 in the use of CaO follows the highest amount of the used mordant metals in the combined mordant method. It corresponds to the K/S values listed in Table 1, where the high values are obtained by using of CaO and $FeSO_4$ mordant on the use of combined mordant methods.

According to [42], the increase of the mordant metal amount used in the dyeing process will not necessarily increase the metal levels in the dyed fabric. Although the mass of the metal ions in the dyeing bath is sizeable, the metal ions in the textile fibers are much lower. It is due to the unbonded metal in the fiber, which will come out with the washing after the dyeing process.

TABLE IV
THE TEST RESULT OF THE AMOUNT OF METAL MORDANT USED

Mordant Method	Mordant Kind	Metal Content (mg/L)				Mordant Used (%)
		Mordant Solution	White Fabric	Dyed Fabric	Mordant Used	
Pre Mordant	$Al_2(SO_4)_3$	980	6.99	216.25	209.25	21.35
	CaO	5607	5.78	970.24	964.46	17.20
	$FeSO_4$	8315	8.69	2364.75	2356.07	28.34
Simultaneous Mordant	$Al_2(SO_4)_3$	980	6.99	212.60	205.61	20.98
	CaO	5607	5.78	899.52	893.73	15.94
	$FeSO_4$	8315	8.69	2398.78	2390.09	28.74
Post Mordant	$Al_2(SO_4)_3$	980	6.99	219.25	212.25	21.66
	CaO	5607	5.78	2293.15	2287.36	40.79
	$FeSO_4$	8315	8.69	2390.64	2381.96	28.65
Pre and Post Mordant	$Al_2(SO_4)_3$	980	6.99	229.84	222.85	22.74
	CaO	5607	5.78	2456.05	2450.27	43.70
	$FeSO_4$	8315	8.69	2416.35	2407.66	28.96

IV. CONCLUSIONS

The main component of the *Gambier* extract identified by X-Ray Diffraction was *catechin*, *catechin anhydrous*, *pyrocatechol* including flavonoid group that could provide color with the aid of mordant. The cotton fabric was used to apply the natural dyes. Each method and type of mordant produced different and strength of color such as light brown, reddish brown to dark brown. Mordant $Al_2 (SO_4)_3$ is tended to give a lighter color than others, and high color strength obtained in the pre-mordant methods. Mordant CaO gave the color of reddish-brown, with the highest color strength on the combined and the post-mordant methods. The mordant $FeSO_4$ has produced a darker color than others have. High color intensity occurred in the combined and simultaneous mordant methods. The washing and rubbing fastness of the cotton fabric results in a range from good to excellent (4-5), while light fastness had a good enough to a reasonable value (3-4). The binding of mordant metal in coloring was still low in a range of 15-40%.

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