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Influenced factors for cotton dyeing process with mangosteen leaf extract

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ABSTRACT

This current study investigated the aqueous extracts of mangosteen leaves for dyeing cotton fabric with various mordants to develop color shadings. Freshly prepared extracts were subjected to preliminary phytochemical screening. It was found that mangosteen leaf extract revealed the presence of pH in the range of 6.0–7.6 and the content of phenolic compound, flavonoid, reducing sugar, and tannin in the range of 0.32–1.10 GAE/mg DW, 0.70–2.97 QE/mg DW, 0.91–7.15 mg/mL and 17.5–24.79 ECGC/mg DW, respectively. This might be responsible for the reduced properties. The three times repetition of 80 g/L extract with mordant was found to be the most satisfying, characterized by using colorimetric characteristics (CIE $L^*a^*b^*$). It can be concluded that mangosteen leaf extract has a high potential for fabric dyeing and might be a promising low-cost reducing agent for developing green dyeing for clothing in the near future.

Keywords: mangosteen leaves, cotton dyeing, mordants

INTRODUCTION

Natural dyes have been used since ancient times. They are currently showing a significant trend towards their popularity. Natural dyes provide more significant advantages than synthetic dyes, which cause environmental and health impacts. The application areas of the colors are numerous, including clothing, cosmetics, pharmaceutical, food industry, medicine, etc. Natural colors can be mainly obtained from minerals, animals, and plants. Most of the mineral dyes are inorganic metal salt and metal oxides. They can be categorized in terms of their colors. Animal dyes are extracted from the dried bodies of insects. Compared to the others, plant dye extracts are a high-potential source as they are commonly and abundantly found. Also, the colorants can be extracted from several plant parts, including leaves, barks, fruits, seeds, roots, etc. (Yusuf et al., 2017) by conventional and modern methods, which depend on their applications (Yadav et al., 2023).

Mangosteen or *Garcinia mangostana* L. (Thai; Mang-Khut), a small evergreen fruit tree in the family Clusiaceae, is widely grown and consumed in tropical countries at a low cost. It was reported that total phenolic, flavonoid, tannins, and saponins were rich in leaves (Suhartati et al., 2019). These components contributed to reducing properties measured using 2,2-diphenyl1-picrylhydrazyl (DPPH) reducing ability assay as indicated by Sassa-deepaeng et al. (2019). Interestingly, the extract also shows antibacterial properties against *Pseudomonas aeruginosa*, which causes various diseases, such as skin infections (Suhartati et al., 2019). Thus, the newly challenged plant might be used in this experiment as a reducing agent and dye.

As indicated by Azeem et al. (2019), it was found that mordanting treated cotton fabrics with iron (II) sulfate and tannic acid developed darker shades. To investigate the effect of mordants on color shadings, the natural materials were selected, based on reported high tannin content, for testing, such as alum, carandas-plum fruits, tea, guava leaf, raw banana peel, neem tree bark, and chemicals were also chosen such as ash calcium hydroxide, $FeSO_4$, and $CuSO_4$. Alum has been used as mordant for natural dyeing for many decades as well as our previous report that carandas-plum fruit possessed the excellent reducing properties for traditional cotton fabrics dyeing with indigo from *Strobilanthes cusia* (Nees) Kuntze (Pranamornkith et al., 2022). Tea is also used as a traditional mordant for dyeing, as reported by Triwiswara and Indrayani (2020). Guava leaf is a good source of tannin, according to the report of Mailoa et al. (2013), while raw banana peel was also reported as a tannin resource by Aroonsrimorakot and Whangchai (2019). Many publications indicated that Neem tree bark comprises tannin in the aqueous extracts (Bhagwat et al., 2020; Islas et al., 2020). For using $Ca(OH)_2$, $FeSO_4$, and $CuSO_4$, it was documented by Azeem et al. (2019) and Indrianingsih et al. (2021).

According to the information provided, the researchers are interested in researching the use of mangosteen leaf extract for dyeing cotton fabric. The concentrations of mangosteen leaves were varied from 40, 80, 120, and 160 g/L for the extraction process. The CIE L* a* b* technique observed the optimal concentration after the dyeing process. The chemical composition of the mangosteen leaf extract solution was examined. The effect of different mordants on the color shade of dyed cotton was studied.

MATERIALS AND METHODS

Materials

The cotton fabric and alum were purchased from a local store. Mangosteen leaves were collected from Na-Ku-Ha village, Amphoe Muang, Phrae province, northern Thailand. Natural mordants such as Carandas-plum, tea, guava leaves, raw banana peel, and neem trees were obtained locally from Amphoe Muang, Lampang province, northern Thailand. Iron (II) sulfate ($FeSO_4$) was commercially provided by Gammaco (Thailand) Co., Ltd., Copper (II) sulfate ($CuSO_4$) was purchased from Ajax Finechem Pty Ltd., Australia.

Methods

Dye extraction from mangosteen leaves

Collected mangosteen leaves were air-dried at ambient temperature for a few months with a moisture content of 50%. Dried leaves were extracted by immersing 40, 80, 120, and 160 g of dried leaves in a liter of water, respectively. Each concentration's crude dye extraction solutions were boiled using a cooking gas burner for 30 minutes. The crude dye extraction solution samples were then separated into two portions. One portion was further used for chemical characterization, and the other was for dyeing.

Identification of mangosteen leaves extracted dye

The crude dye extraction solution was filtered with filter paper (Whatman No. 1) and used for the following determination:

UV-Visible spectral characteristic

The crude dye extraction solution samples were diluted 100-fold for characterization by UV-visible spectroscopy. The spectrophotometer (Biochrome, S50-S80) measured in the range of 200 to 800 nm to obtain spectra.

Phenolic compound content

The quantification of total phenolic content (TPC) in water extracts was conducted in triplicate employing the Folin-Ciocâlteu reagent. The formation of a blue complex resulting from electron transfer was monitored using a UV-Vis spectrophotometer (Dshing Instrument Co., Ltd., China) following the procedure outlined by Singleton and Rossi (1965). Specifically, 20 µL of the extract was combined with 100 µL of FC reagent (Merck, Damstadt, Germany) in 1,980 µL of deionized water, followed by a 5-minute incubation at room temperature. Subsequently, 300 µL of a 7% solution of Na_2NO_3 (Univar, Ajax Finechem, Australia) was introduced and vigorously mixed before a 60-minute incubation in darkness at room temperature. Ultimately, the absorbance was measured at 765 nm. The TPC values were expressed as micrograms of gallic acid equivalent per milligram of dry weight of the extract, determined through linear regression analysis utilizing gallic acid standards at concentrations ranging from 0 to 25 µg/mL.

Flavonoid content

The determination of flavonoid content (FC) was conducted in triplicate using the aluminum trichloride ($AlCl_3$) colorimetric method as described initially by Christ and Müller (1960), with adaptations to accommodate 96-well microplates, as outlined by Yodthong et al. (2020). In this procedure, a 20 µL aliquot of the ethanolic solution of each extract or various concentrations of quercetin was combined with 75 µL of deionized water, followed by the addition of 25 µL of a 5% $NaNO_2$ solution (Univar, Ajax Finechem, Australia) within a microplate. Following a 5-minute incubation at ambient temperature, 25 µL of a 10% $AlCl_3$ solution (Lobachemie, India) was introduced, and the mixture was further incubated for 6 minutes under the same conditions. Subsequently, 100 µL of a 1M NaOH solution (Merck KGaA, Germany) was added, and the reaction was allowed to continue for 60 minutes in the absence of light before measuring the absorbance at 405 nm using a microplate reader (Biobase EL-10A China). The flavonoid content was expressed as micrograms of quercetin (Sigma–Aldrich, Germany) equivalent (QE) per milligram of dry weight of the extract.

Tannin content

The content of tannin also had a direct impact on the reducing properties of the sample. To quantify the tannin content (TC), the vanillin assay was performed in triplicate, following the procedure outlined initially by Broadhurst and Jones (1978), with adaptations to minimize reagent volume and consequently reduce the generation of hazardous waste. Specifically, 250 µL of the extract was combined with 450 µL of a 1% vanillin solution (Merck, Germany) in methanol (QRëC, New Zealand). After a 5-minute incubation at room temperature without light, the mixture was vigorously mixed with 300 µL of concentrated hydrochloric acid (QRëC, New Zealand) and allowed to incubate for 30 minutes in darkness. Subsequently, the solution turned red, and this change was detectable at 500 nm using a UV-Vis spectrophotometer (Dshing Instrument Co., Ltd., China). The tannin content was expressed as micrograms of Epigallocatechin gallate (Sigma–Aldrich, Germany) equivalent (EE) per milligram of dried weight of the extract.

Saponin

A total of 5 mL of an ethanolic extract was vigorously agitated with 5 mL of distilled water within a test tube and subsequently subjected to warming in a water bath maintained at 39 ± 1 ºC for 10 minutes. The observation of a persistent honeycomb froth formation for 10 minutes served as an indicative marker for the presence of saponins in accordance with the criteria defined by Harborne (1998).

Reducing sugar

Reducing sugar was determined by the DNS method, which was adjusted by Saqib and Whitney (2011). DNS reagent was prepared by mixing 3,5-dinitro salicylic acid (DNS) solution (2.5 g in 100 mL of 1 M NaOH) with the heated dissolved sodium potassium tartrate solution (75 g in 125 mL of distilled water). The volume of the mixing solution was then adjusted to 500 mL with distilled water. DNS reagent 1 mL was added to 0.1 mL crude dye extraction solution samples and adjusted to the final volume of 4 mL with distilled water. The tube was heated in boiling water for 5 minutes. The absorbance was measured at 540 nm. The standard measurement was performed using 2 mM glucose as a reducing sugar standard.

Dyeing process

Cotton fabrics were soaked in hot crude extracted dye solution for 30 min with a ratio of fabric 20 g: 1 L of liquor. Dyed cotton fabrics were brought to squeeze until damp and mordanted with calcium hydroxide solution (5% w/v). The fabrics were eventually washed and air-dried.

Dyeing repetition

The dyeing repetition was carried out for five treatments: one-time, two-time, three-time, four-time, and five-time repeated dying.

Mordanting process

Different mordants were used, including alum, Carandas-plum (*Carissa carandas* L.) fruit, tea, guava leaf, raw banana peel, Neem tree (*Azadirachta indica*) bark, ash, calcium hydroxide solution at the concentration of 5% w/v and $FeSO_4$, and $CuSO_4$ at the concentration of 10 mg/L. After the dyeing process with the crude dye extraction solution, the damped dyed fabrics were soaked and squeezed in each mordant for 5 minutes. All dyed fabrics were then washed and air-dried.

Color measurement and fastness test

The color coordinates CIE L^* a^* b^* values of all dyed clothes were measured by HunterLab, ColorQuest XE model. The dyed clothes were tested and evaluated for colorfastness to washing.

RESULTS AND DISCUSSION

The powder of mangosteen leaves was prepared by drying fresh leaves at ambient temperature for months until the moisture content dropped to 50%. To make a fine powder, dried leaves were ground and sieved. The obtained powder has a light brown color, and the crude dye extraction solution has a dark brown color (Figure 1).

Figure 1. Sample preparation.

Property and chemical composition analysis

The extract prepared by dissolving mangosteen leaf (ML) powder in various concentrations in hot DI water showed a pH value in the range of 6.0 to 7.6. It might result from the high composition of tannic acid, as indicated in Figure 2 and Table 1. However, the results also show that the ML-extract consisted of other organic acids such as tannins, saponin, and phenolic compounds, as reported by Alsultan et al. (2017), which contributed to the decrease of the extract pH. Among these compounds, tannins are an important component of the dyeing process as they can act as a natural mordant to sustain the color of matter permanently. As reported by Sassa-deepening et al. (2019), it was found that the aqueous extract of mangosteen leaves possessed high reducing properties. To investigate the source molecules of reducing power, the total phenolic content (TPC) assay was conducted using the Folin-Ciocalteu colorimetric method. The highest TPC of plant aqueous extracts was found gradually with increasing concentration. This indicates ML-extract had a high ability to reduce Folin-Ciocalteu reagent. In addition, ML extract also exhibited the highest flavonoid content and reducing sugar, which contributes to the reducing properties. Therefore, ML extract might be an excellent tentative plant for using traditional cotton fabrics dyeing with mordants. To characterize the composition of the ML extracts, the UV-Vis spectra of the extract were recorded by spectrophotometry and shown in Figure 2.

Table 1. Property and chemical composition of extracted dyes from mangosteen leaves

Extracted dyes (g/L)	Phenolic compounds (GAE/mg DW)	Flavonoid (QE/mg DW)	Tannin (ECGC /mg DW)	Reducing sugar (g/L)	pH	Saponin
40	0.32	0.70	17.57	0.91	7.6	–
80	0.69	1.64	20.39	4.19	6.8	+
120	0.85	2.48	22.29	5.47	6.4	++
160	1.10	2.97	24.79	7.15	6.0	++

– = N.D. + = mild positive. ++ = strong positive.

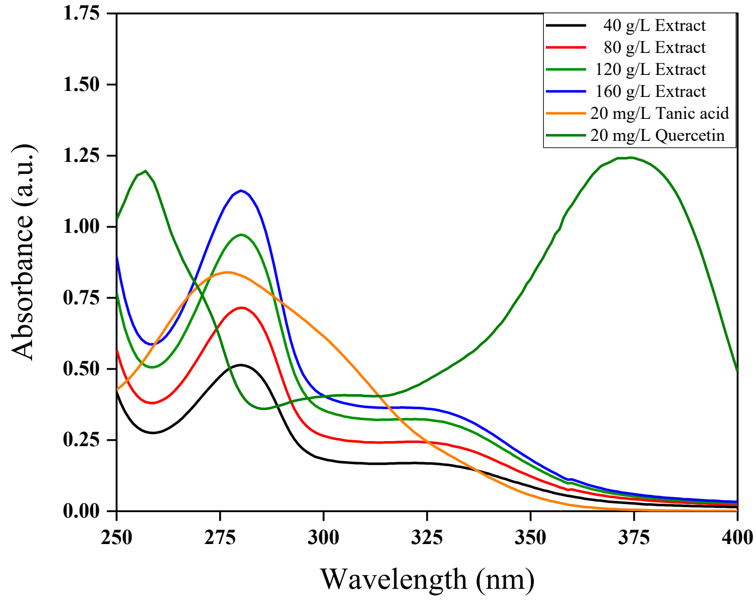


Figure 2. UV-Vis spectra of extract dyes from mangosteen leaves at different concentrations.

The characteristic color of ML extract could be the consequence of the presence of the major compounds. Tannin and quercetin are transparent in the visible region (Grasel et al., 2016). Therefore, Figure 2 shows the absorption spectra of those compounds generated during the analysis of the aqueous extract at various concentrations of ML extract (40–160 g/L). It was found that the strong absorption at 280 nm was obtained from aqueous solutions. The result is supported by the founding of Grigsby et al. (2013), who documented that the major peaks of tannic acid and tannin derivatives were observed in the UVB (285–315 nm). Unfortunately, UV-visible spectra for quercetin (spectrum) exhibited two absorption bands at 256 and 374 nm were not detected. From the UV data ML-extracted, it can be stated that there was a tannin-related compound present in the dye extract.

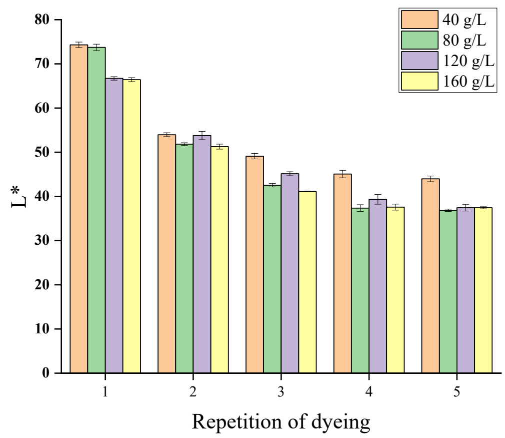


Figure 3. The lightness value (L*) of ML-extract.

The color of the dyed samples is given in CIE Lab coordinates (L^*, a^*, b^*): L^* corresponding to the brightness (100 = white, 0 = black), a^* to the red-green coordinate (+ve = red, –ve = green or blue) and b^* to the yellow-blue coordinate (+ve = yellow, –ve = green). From Figure 3, it was found that the ML-extract at a concentration of 80 g/L shows the effective cost with efficiency for dyeing due to the saturated perceptual darkness on cotton fabric at the third repetition. However, the a^* value was also considered, and the result is shown in Figure 4.

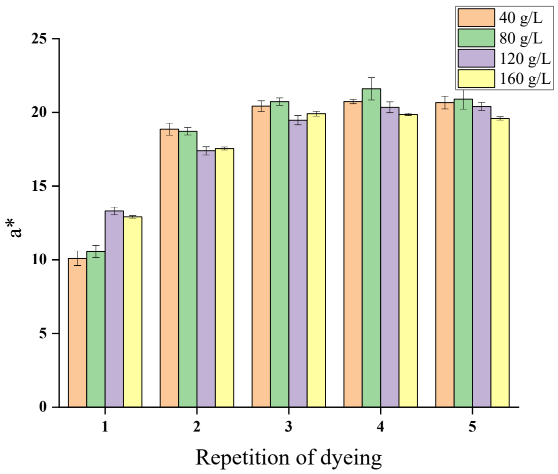


Figure 4. The green-magenta opponent color value (a*) of ML-extract.

The a^* value, represented as a red and green scale, runs from 0 to 60. From the data, it can be seen that cotton fabric dyed three times repetition at a concentration of 80 g/L without mordant revealed the highest a^*value of 20.73 ± 0.25.

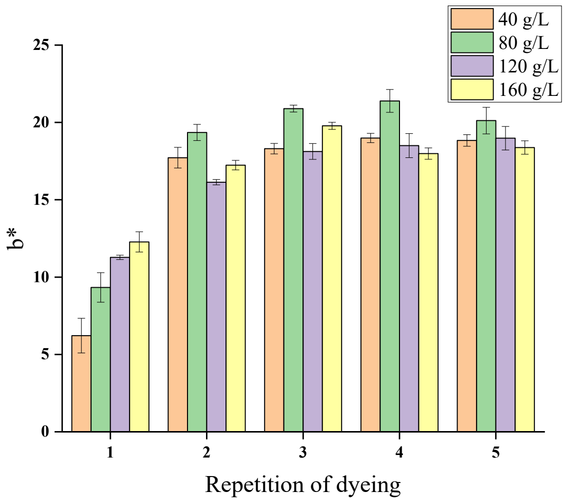


Figure 5. The blue-yellow opponents' value (b*) of ML-extract.

The b^* notation represents the chromatic color in the direction of blue and yellow. A positive value of b^* ($+b^*$) from 0 to 60 and a negative value of b^* ($-b^*$) from 0 to -60. The average value of b^* in this study was 20.89±0.22 (Figure 5). Therefore, it is clear that cotton fabric dyed three times of repetition at a concentration of 80 g/L is a suitable condition for further study on the mordanting step.

Dyeing cotton with extract dyes from mangosteen leaves

In Table 2, the cotton dyeing process with various concentrations of ML-extract at 40, 80, 120, and 160 g/L showed that the lightness value (L*) decreased with higher concentrations. This represents the effect of the dye's concentration on the shades of dyed fabrics. It was obviously seen that the higher the dye uptake, the darker yellow-brown shade of the fabric color appeared. This evident was confirmed by an increase of the blue-yellow coordinate (b^*) in the first round of dyeing repetition, particularly.

Dyeing repetition has been reported to darken the color of the fabric and reinforce the fabric's dyed color (Suciatmih, 2020). In this study, it caused a deep brown color on the fabric sample (Table 2). The decrease of the L^* value of 1–5 dyeing repetitions was observed and illustrated that the frequency of dyeing applied affected the intensity of the brown color. This is due to the high negative charge in the tannin compound, which is contained dominantly in mangosteen leaves, as well as in its peels, which reacted favorably with the Ca^{2+} atom in a mordant compound. The $Ca(OH)_2$ solution, as a mordant compound in this case, plays an important role in the link between fabric fiber and dyes (Kusumawati et al., 2017).

Color fastness

Table 3 revealed the result of color fastness of dyed cotton fabrics with mangosteen leaves to washing. It provided promising lightfastness results with the good to excellent range of 4–5. The fabric samples, after washing repetition 5 times, showed non-significant color variations when subjected to wash fastness according to the lightness value (L^*).

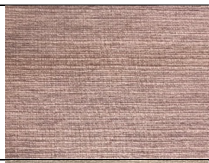
Table 2. Dyeing cotton with extract dyes from mangosteen leaves at different concentration

Extracted dyes (g/L)	Dyeing repetition				
	1	2	3	4	5
40	 L* 74.30 a* 13.44 b* 6.22	 L* 53.97 a* 18.86 b* 17.72	 L* 49.10 a* 20.42 b* 18.30	 L* 45.06 a* 20.74 b* 18.99	 L* 43.97 a* 20.66 b* 18.84
80	 L* 73.73 a* 10.57 b* 9.33	 L* 51.84 a* 18.72 b* 19.35	 L* 42.54 a* 20.73 b* 20.89	 L* 37.36 a* 21.60 b* 21.39	 L* 36.86 a* 20.90 b* 20.12
120	 L* 66.71 a* 13.31 b* 11.28	 L* 53.78 a* 17.39 b* 16.14	 L* 45.13 a* 19.47 b* 18.12	 L* 39.33 a* 20.35 b* 18.50	 L* 37.55 a* 20.41 b* 18.98
160	 L* 66.43 a* 12.91 b* 12.28	 L* 51.26 a* 17.55 b* 17.24	 L* 41.11 a* 19.91 b* 19.78	 L* 37.57 a* 19.87 b* 17.99	 L* 37.44 a* 19.59 b* 18.38

Table 3. Color fading of cotton-dyed washing

Items	**Repetition of washing**				
	1	**2**	**3**	**4**	**5**
Dyed clothes					
L*a*b*	L* 45.86 a* 17.19 b* 16.31	L* 43.81 a* 17.81 b* 18.40	L* 42.35 a* 18.23 b* 18.67	L* 45.75 a* 17.45 b* 16.43	L* 45.66 a* 17.47 b* 16.64
Grey scale	5	5	5	4-5	4-5

Table 4. Color strength of ML-extract dyed cotton fabrics with mordants

Mordants	pH	Cotton fabrics	Color		
			L^*	a^*	b^*
1. None	6.9		71.01	10.98	8.52
2. Alum	3.3		80.23	2.59	9.94
3. Carandas-plum (*Carissa carandas* L.) fruit	3.4		80.83	4.32	7.19
4. Tea	6.3		79.15	1.79	14.99
5. Guava leaf	6.6		78.64	3.30	12.37
6. Raw banana peel	7.4		77.13	6.91	3.05
7. Neem tree (*Azadirachta indica*) bark	7.6		72.85	9.45	7.46
8. Ash	9.5		73.25	8.25	5.44
9. $Ca(OH)_2$	11.8		67.21	11.81	10.96
10. $FeSO_4$	5.8		61.73	5.56	-1.86
11. $CuSO_4$	5.6		66.44	8.65	10.27

The analysis results show that there were differences between the types of mordanting agents (Table 4). The lowest pH value was found in alum solution, while the highest pH value was detected in $Ca(OH)_2$. It seems that a higher pH can develop darker color shades of ML-extract. The dye solution of ML-extract using $FeSO_4$ as a mordanting agent has the lowest brightness level of 61.730.12 while using Carandas-plum as a mordanting agent has a brightness level of 80.83 ± 0.23. The variation of chemicals ferrous sulfate as a mordanting agent seems to be more affected by color darkening compared to those bio-mordants. As reported by Mongkholrattanasit et al. (2015), copper sulfate and ferrous sulfate, mordants are well known for their ability to form coordination complexes to readily chelate with the dye. The darker color shades by mordanting with tannic acid indicated strong interactions of this mordant with dye molecules to form an insoluble complex that firmly attached to cotton fabric (Ali, et al. 2010), while metal ions constituted a firm bonding with dye molecules as well as cotton fabric ultimately approaching towards maximum color strength as discussion by Bouatay et al. (2016). The fabric dyed using ML-extract under $Ca(OH)_2$ as mordant has the highest* value of 11.810.26, while fabric dyed under tea as mordanting agent provided a* value of 1.79 ± 0.06, indicating $Ca(OH)_2$ was proper mordant in this study. The average value of b^* was in the range of −1.864 to 14.99. ML-extract dye solution with tea has the highest b^* value, while $FeSO_4$ possesses the lowest value of b^*. The results obtained from the measurement of b^* notation revealed the variations of green and yellowish green in the sample. The greater value of b^* indicated the more yellow sample color. Based on the data in Table 4, it can be indicated that ML-extract natural dyes generated many shades under mordant use.

CONCLUSIONS

In summary, mangosteen leaves extracted with hot water are suitable for use in the dyeing process. The 80 g/L of ML extract provided the dark-brown color of the cotton fabric caused by contained phytochemical compounds. Dyed cotton fabric with mordants showed many shades, including brown-grey, pale brown, yellow-brown, and yellow colors. Therefore, ML-extract has a high potential in traditional cotton fabric dyeing. This presents a promising low-cost natural dye for green dyeing fabric in the near future.

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