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Effects of additives on the dyeing of cotton yarn with the aqueous extract of *Combretum latifolium* Bl. stems

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Abstract

Flavonoid constituents from the aqueous extract of the stems of *Combretum latifolium* sourced in Thailand, have potential use as dyestuffs for cotton dyeing. In an effort to improve current natural dyeing methods with this extract, further aspects of the process were studied. It was found that before equilibrium was reached, an increase in temperature lead to an increase in dye adsorption rate of the extract; the initial rate and extent of dye adsorption was further increased by the addition of sodium chloride to the dye bath. In addition, cotton yarn

which was pretreated with a chitosan solution (with and without a cross-linking glyoxal solution), followed by dyeing with the *C. latifolium* extract provided better depth of shade (*K/S*) and also gave better fastness to light and washing than the untreated cotton yarn. Post-mordanting cotton yarn with a bio-mordant solution from *Memecylon scutellatum* leaves also gave good light and wash fastness of the resultant dyed cotton, comparable to the dyeing results with the less environmentally friendly alum as a mordant.

Keywords: Combretum dyeing, Flavonoids, Dyeing, Chitosan, Mordants

Introduction

Combretum latifolium Blume (*Combretaceae*) is a woody climber widely distributed in tropical Asia including Thailand where it is known as Man daeng or Uat chueak [1]. In Thailand, the wood of this plant is used for medicinal purposes [2], while antibacterial activities have been reported in extracts of this plant and other *Combretum* species [3,4]. Methanolic extracts from leaf or stem material of *C. latifolium* showed strong antioxidant activities [5,6], as well as some *in vitro* anti-cancer cell activity [6], possibly due, at least in part, to the presence of flavonoid antioxidants. From our analysis based on the Shinoda and FeCl₃ tests [7], it was found that the yellow aqueous extract from the stem of this plant contained flavonoid constituents. Villagers who live in the Cheanyai District, Nakhon Si Thammarat, Thailand have used the aqueous extract from the stem of this plant for cotton dyeing together with metal salts such as alum, ferrous sulfate or calcium hydroxide as mordants. The dyed cotton yarn with alum as mordant gave good fastness to light and washing, but there are potential environmental impact issues with the use of such a mordant.

The flavonoid constituents from *C. latifolium* have potential use as natural yellow colorants for textile dyeing. Detailed aspects of the dyeing process with dye from the aqueous

extract of the stems of *C. latifolium* have not been assessed previously, and as far as we are aware, the specific identification of these water-soluble flavonoids has not been reported although they are known from other *Combretum* species [8,9]. In order to more accurately assess village dyeing conditions, a crude aqueous dye preparation was used in this work.

It is well known that natural dyes often require a metallic mordant to form water-insoluble metal ion-dye complexes and to prevent the color from either fading after exposure to light or being washed out. However, the waste water in the residual dye bath containing these heavy metal mordants such as potassium dichromate, stannous chloride, ferrous sulfate and copper sulfate, may affect the environment and public health. To avoid these problems and enhance the rate and extent of dye uptake or the fastness properties of the dyed fabric, other additives like sodium chloride, or chitosan [10] and a crosslinking agent (e.g. glyoxal) [11] can be used. While glyoxal is not a natural agent, only dilute aqueous solutions are required for crosslinking.

The overall goal of the current work was to investigate potential ways to improve present cotton dyeing practices with *Combretum* - based dyes. To this end dye uptake effects in cotton yarn of sodium chloride in the dye bath were probed, as well as the effects of chitosan, with and without a crosslinking reagent. A comparison of the chitosan-mediated dyeing results with those of alum as a mordant were also examined, together with the effect on dyeing through post-mordanting of the cotton with a more environmentally compatible bio-mordant [12] extract from the plant *Mimocylon scutellatum* Naud. (Melastomaceae). The results are now reported in this paper.

Experimental

Materials and Chemicals

The cotton yarn (double yarns, 100% combed yarn, model CM/20/2, code 0058/1282; Union Star Co., Ltd.) used was purchased from villagers living in Cheanyai District, Nakhon Si Thammarat, Thailand. To remove wax and impurities, the cotton yarn (100 g) was added to boiling water (2 L) to which had been added soap flakes (*ca* 7 g) and sodium carbonate (3 g). The mixture was then boiled for 1 h. The cotton was then removed, and washed sequentially with hot water and cold water (in order to avoid break down of the emulsion and precipitation of the impurities onto the cotton), squeezed to remove excess liquor and then air dried at room temperature [13]. While this is an old method for wax and impurity removal, it is still a viable one and is the method used by the villagers.

(+)-Catechin hydrate and chitosan from crab shells (medium molecular weight, 75-85% deacetylation, viscosity 200.000 cps, CAS 9012-76-4) were purchased from the Aldrich Chemical Company. Both the sodium chloride (NaCl) and alum ($KAl(SO_4)_2 \cdot 12H_2O$) (both from Merck, Germany) used in this work were analytical grade. An aqueous solution of glyoxal (40% v/v) was employed without purification as a crosslinking reagent. This solution was supplied by VPC group Co., Ltd, Thailand.

Cotton Pretreatment with Chitosan Solution

Chitosan is a deacetylated derivative of the natural polymer chitin, which is found in crab and shrimp shells as well as other sources [14]. Chitosan contains two main functional groups, namely hydroxyl and amino groups, as well as ether linkages and some *N*-acetyl groups. It can be used as a binder in the dyeing of cellulosic fabrics and has been reported to improve the dye uptake and adsorption of lac dye on cotton [13,15], while pretreatment of cotton fabrics with chitosan increased the exhaustion of direct dyes [16]. In our study, 0.10%, 0.30%, 0.60%, and 1.20% w/v aqueous solutions of chitosan were prepared by dissolving the required amount of chitosan in 5% v/v aqueous acetic acid solution. The cotton yarn (10 g),

prepared as noted above, was then immersed directly in 0.10%, 0.30%, 0.60%, and 1.20% w/v aqueous solutions of chitosan at room temperature for 12 h with a material to liquor ratio (MLR) of 1:50. After this time the yarn was removed, squeezed and air dried in the sun for 2 days before further use.

To obviate the loss of some chitosan from the cotton yarn, glyoxal was used as a crosslinking reagent. We needed to assess whether crosslinking would further improve dyed cotton properties over chitosan pre-treatment alone. The cotton yarn sample coated with a 0.30% w/v aqueous solution of chitosan, the optimal concentration of the chitosan solution in this work, was immersed in a 4% v/v aqueous solution of glyoxal at pH 4 and 40 °C for 60 min. Following the crosslinking reaction, the sample yarn was removed, pressed, washed several times with deionised water and then dried in an oven at 50 °C for 16 h [17].

Preparation of an Aqueous Leaf Extract of *M. scutellatum*

Dried leaves of *M. scutellatum* (250 g) from Buriram province, Thailand, were boiled with water (1 L) for 30 min. The solution was then filtered affording a yellow-green filtrate. This solution was used as a post-mordant in our work. An aqueous extract of the leaves of *M. scutellatum* is also used in the village dyeing context.

Preparation of Crude Dye Powder from the Aqueous Extract of *C. latifolium*

Fresh stems (1 kg) of *C. latifolium* (Cheanyai District, Nakhon Si Thammarat, Thailand, herbarium specimen number WU 01454, Walailak Botanic Garden: Walailak University, Miss Wassana Chongkrajak) were cut into small pieces and dried at 40 °C in the oven for 24 h. The dried material (240 g) was extracted with deionized water (2.0 L) at 80 °C in a thermostatted bath for 60 min. The aqueous solution was filtered and the filtrate (pH 4.82) then dried using a freeze dryer (PFR-100, EYELA) to give the crude dye extract as a

yellow powder (54.8 mg g⁻¹; 5.48 % of the dried stem material). The solid crude dye extract was easier to handle and more stable than the solution, which was thus prepared each time from this solid before use. The UV-Visible absorption spectrum of the aqueous crude extract showed an absorption band at λ_{max} 370 nm against deionized water as the blank. The crude yellow dye gave positive color tests for the presence of flavonoids (FeCl₃ test; Shinoda's test [7]), and flavonoid content was determined by the aluminium chloride colorimetric method noted below. The crude dye extract was used without further purification in this work. Crude dye extract solutions for the cotton dyeing experiments were then made up by dissolution of the powder in deionized water to give a concentration of 3000 mg/L. All the dye powder was soluble in water.

Determination of Flavonoid Content in *C. latifolium* Stem Extracts

The flavonoid content of crude aqueous and crude methanol extracts of *C. latifolium* stems was determined using the aluminium chloride method and with catechin as a standard [18]; catechin was the major flavonoid in another *Combretum* species, *C. hereroense* [9]. The plant extract solution (0.25 mL) was added to distilled water (1.25 mL) followed by 5% sodium nitrite solution (0.075 mL) and allowed to stand for 5 min. Then 10% aluminium chloride solution (0.15 mL) was added. After 6 min, 1.00 M sodium hydroxide (0.5 mL) was added to the mixture and the mixture then diluted with distilled water (0.275 mL). The absorbance of the resultant solution was measured immediately at 510 nm using a Shimadzu UV-2450 UV-Visible spectrophotometer; determination of the aluminium(III)-flavonoid complex in alkaline solution with sodium nitrite at 510 nm is indicated for rutin, luteolin and catechin flavonoids [19]. All experiments were performed in triplicate. The results were expressed as milligrams of catechin equivalents per gram of *C. latifolium* extract; the plant extract volume was chosen so absorbance values were on the catechin standard curve.

Instruments

A thermostatted shaker bath (SWB 5050 Shaking Water Bath, Labnet National Company, Type SBD50 Labnet), operated at 150 strokes/min was used for all experiments. A colorimetric spectrophotometer (HunterLab, Model: ColorFlex 45°/0°, illuminant D65/10° observer) was used to measure the reflectance of dyed yarn and color coordinate CIE $L^* a^* b^*$ values. A pH meter (Orion 420, USA) was employed to measure the pH values of the solutions. A Shimadzu UV-2450 UV-Visible spectrophotometer was used for determination of flavonoid content and a Unicam 310 UV-Visible spectrophotometer was employed for the UV-Visible spectra and absorbance measurements using quartz cells of 1 cm path length.

Methods

Effect of Sodium Chloride on the Dyeing of Cotton Yarn

The experiments were carried out by shaking the dye extract solution (50 mL; 3000 mg/L) in a conical flask (125 mL) with and without sodium chloride (8.60×10^{-2} M) without pH control at 50 °C in a thermostatted shaker bath operated at 150 strokes/min; the material to liquor ratio was kept at 1:50 in each experiment. After 30 min, the cotton yarn, which had been pre-warmed in the thermostatted bath for 30 min, was immersed in the dye solution. The cotton samples were then quickly withdrawn after different immersion times. Dye concentrations were determined at time zero and at subsequent times using a calibration curve based on absorbance at λ_{\max} 370 nm versus dye concentration in standard dye solutions. The amount of dye adsorbed per gram of cotton (mg/g cotton) at any time (q_t) was calculated by a mass-balance relationship equation (1) as follows:

$$q_t = (C_0 - C_t) \frac{V}{W} \quad (1)$$

Where C_0 is the initial dye concentration (mg/L) and C_t is the dye concentration after dyeing time t (mg/L), V is the volume of dye solution (L) and W is the weight of cotton yarn (g) used.

The effect of temperature on the adsorption of dye extract solution on cotton in the presence of sodium chloride (8.6×10^{-2} M) was investigated at 30 and 50 °C in a similar way. Dye concentrations were determined at time zero and at subsequent times using a calibration curve based on absorbance at λ_{\max} 370 nm versus dye concentration in standard dye solutions. This major absorption band may correspond to one or more flavonoid components.

Dyeing of Untreated and Treated Cotton yarn with the *C. latifolium* Dye Extract

The untreated cotton yarn and cotton yarn pretreated with a 0.30 % w/v aqueous solution of chitosan with and without a crosslinking solution (glyoxal) were dyed with the aqueous dye solution (3000 mg/L) from the stem of *C. latifolium*, keeping a material to liquor ratio of 1:50. Dyeing was carried out by shaking the cotton yarn with dye solution in a conical flask at 70 °C (the optimal temperature condition for dyeing) for 60 min in a thermostatted shaker bath operated at 150 strokes/min. After dyeing, the cotton yarn was washed with tap water to remove the unfixed dye and then air dried.

The dyed cotton yarn sample from the untreated cotton yarn was impregnated with an aqueous leaf extract of *M. scutellatum* at room temperature for 30 min using the post-mordanting method. The yarn was then removed, washed with tap water to remove the unfixed dye, and finally air dried.

The dyed cotton yarn sample from the untreated cotton yarn was immersed directly in an aqueous solution of alum (1.00% w/v; deionized water) at room temperature for 30 min. It was then removed, washed with tap water to remove the unfixed dye, and then air dried.

Color Measurement and Fastness Testing

Each dyed cotton sample was wound around the skein holder twice in taut layers and an average of three readings was taken for each measurement [11]. The reflectance of dyed yarn and color coordinate CIE $L^*a^*b^*$ values were measured on a ColorFlex 45°/0° spectrophotometer using illuminant D65 and 10° standard observer. The tristimulus values, X , Y , and Z under illuminant were converted to CIE $L^*a^*b^*$ coordinates. The color indices obtained from the CIE $L^*a^*b^*$ measurements delineate the hue color. Depth of shade of dyed cotton expressed as K/S values was measured by the light reflectance technique using the Kubelka-Munk equation [20]:

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

where R is the observed reflectance, K is the absorption coefficient, and S is the light scattering coefficient. A higher K/S value points to better dye receptivity of the substrates.

Fastness properties of the dyed yarn were assessed according to standard methods; the specific tests were for color fastness to washing, ISO 105-C01: 1989(E), and color fastness to light, ISO 105-B02: 1994(E).

Results and Discussion

Effect of Dyeing Temperature

The effect of dyeing temperature on the depth of shade (K/S) of dyed cotton yarn is shown in Fig. 1. The result showed that the K/S values increased with the dyeing temperature up to 70 °C and remained near constant in the dyeing temperature range 70-80 °C. The result is consistent with increased fiber swelling with higher temperature, leading in turn to a higher dye uptake and an enhancement of dye diffusion. A suitable dyeing temperature for cotton

yarn dyed with the aqueous stem extract of *C. latifolium* was thus fixed at 70 °C for all subsequent experiments apart from those assessing the effect of sodium chloride.

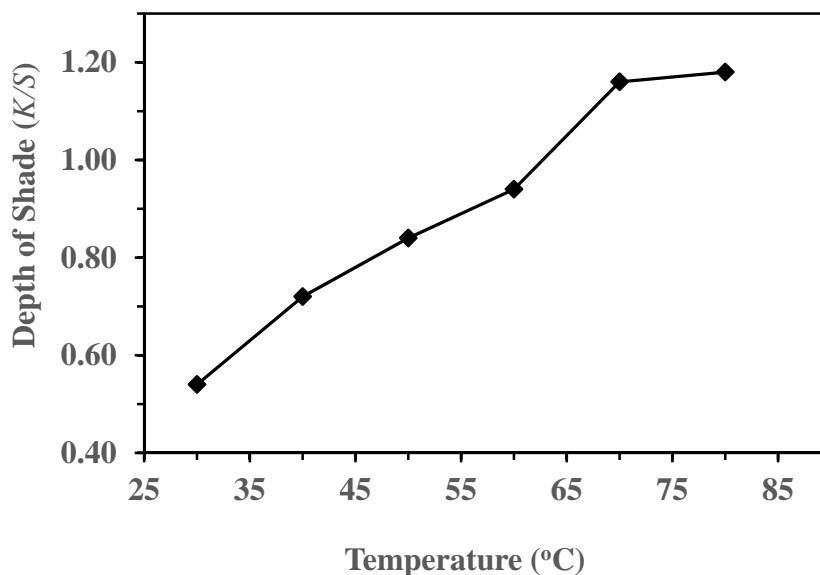


Figure 1. Effect of dyeing temperature on the depth of shade (*K/S*) of dyed cotton yarn.

Effect of Sodium Chloride on the Adsorption of Dye Extract onto Cotton Yarn without pH Control

The natural cellulose fibers carry electron rich hydroxyl groups, and some carboxylate groups (from primary alcohol oxidation and dissociation in water) and the flavonoid dyes from the stem of *C. latifolium* are likely to contain phenolic hydroxyl groups in the molecule providing a negative charge on ionization. Therefore, flavonoid molecules and cellulose would be expected to repel one another. This means that electrical repulsion must be overcome by other forces of attraction between the flavonoid dye and cellulose fibers in the dyeing process. For this reason, electrolytes such as sodium chloride have been added into the dye bath to promote the adsorption of anionic dye on cellulose fibers [13,21]. Therefore, the effect of sodium chloride on the adsorption of the dye onto cotton yarn without pH control was investigated and the results are shown in Fig. 2. It was found that adding the sodium

chloride to the dye bath increased the amount of dye adsorbed per gram of cotton (q_t) compared to the sodium chloride-free dye bath. Also the time required to reach equilibrium in either case was the same (60 min) (Fig. 2), although the initial rate of adsorption was faster with sodium chloride present. The amount of dye adsorbed per gram of cotton (mg/g cotton) at equilibrium time (q_e) was found to be approximately 18 and 10 mg/g cotton in the presence and absence of sodium chloride respectively. These results can be explained by the Donnan model [22] in which dyeing of the cellulose fibers occurs by the transfer of dye from the external to the internal solution. With the addition of sodium chloride (as an electrolyte) to the dye solution, the sodium ions could distribute between the external solution and internal solution and through electrostatic interactions ameliorate the negative potential on the cellulose surface. This in turn would allow the dye molecules to be adsorbed onto the cotton.

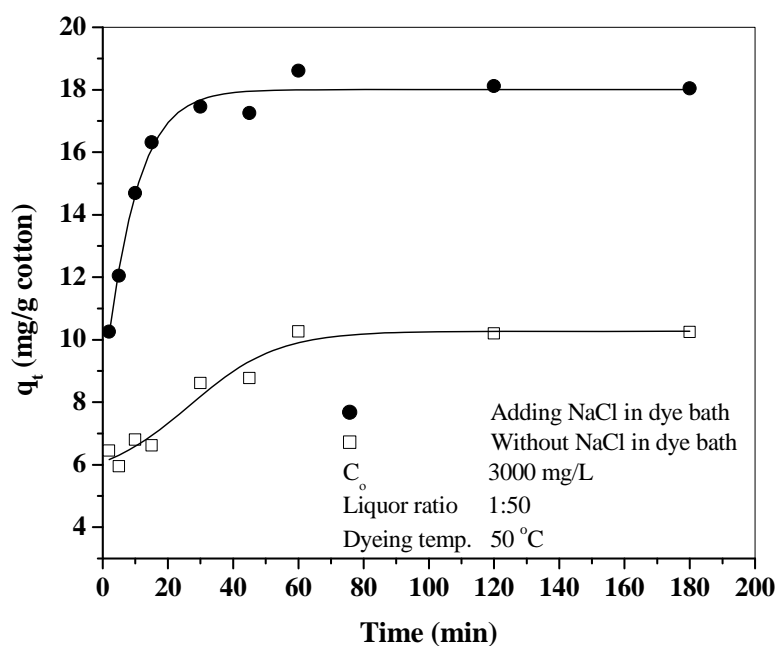


Figure 2. The effect of sodium chloride (NaCl) on the adsorption of dye extract from *C. latifolium* on cotton without pH control.

Flavonoid Content of the *C. latifolium* Stem Extracts

Flavonoid content of the crude aqueous and crude methanol extracts of *C. latifolium* stems was determined using a colorimetric method. Using the standard plot of catechin concentration as a reference ($y = 0.003x + 0.0681$, $R^2 = 0.9951$), the flavonoid content in the crude aqueous and crude methanol extracts of *C. latifolium* stems was significant and found to be 45.87 ± 2.53 and 25.29 ± 1.90 mg catechin equivalent per gram of crude extract, respectively.

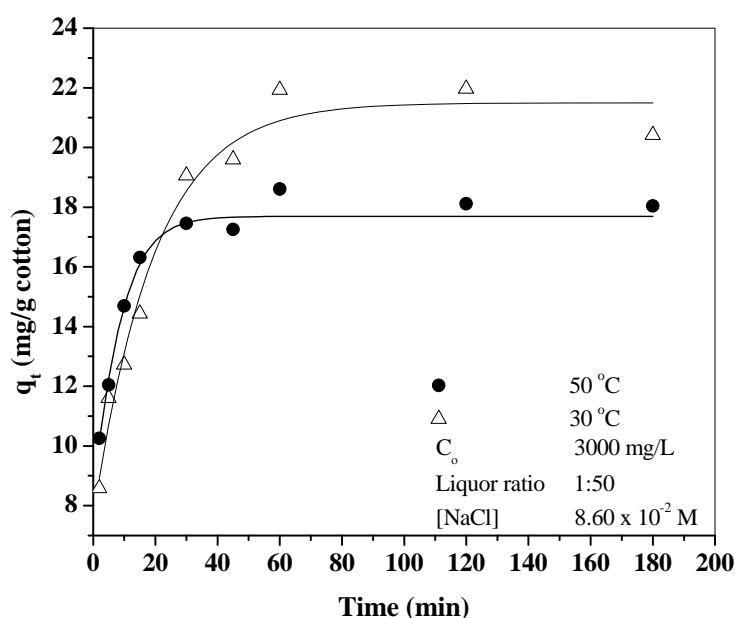


Figure 3. The effect of contact time and temperature on the adsorption of dye extract from *C. latifolium*, in the presence of sodium chloride (NaCl), on cotton without pH control.

Effect of Temperature on the Adsorption of Dye Extract on Cotton in the Presence of Sodium Chloride

The effect of temperature on adsorption of the *C. latifolium* dye extract on cotton at an initial dye concentration of 3000 mg/L at 30 and 50 °C in the presence of sodium chloride is shown in Fig. 3. Before equilibrium was reached, an increase in the temperature led initially

to an increase in the amount of dye adsorbed per gram of cotton. However, the rate of dye uptake then decreased as the amount of dye on the fiber increased, consistent with the necessity for dye diffusion from the fiber surface binding sites to the interior sites to allow for further dye adsorption from the dye bath [21]. After equilibrium was attained, the adsorption of dye decreased with increased temperature pointing to an exothermically-controlled process. Thus, while a higher temperature increased the rate of dyeing, it decreased the amount of dye ultimately adsorbed on the cotton [22]. A similar observation was also reported in our previous studies on the adsorption of lac dye on cotton and silk [23,24].

Table 1. *K/S* and CIE $L^*a^*b^*$ values of dyed cotton yarn samples at different concentrations of an aqueous solution of chitosan, and with glyoxal cross-linking. All cotton samples were dyed with the dye extract solution from *C. latifolium*.

Concentration of chitosan solution (% w/v)	Cross-linking with a 4% v/v glyoxal solution	<i>K/S</i> ($\lambda = 370$ nm)	CIE		
			L^*	a^*	b^*
0 (control)	×	1.16	80.24	-0.01	17.94
0.10	×	2.90	73.53	1.42	22.80
0.30	×	3.67	70.15	1.84	23.33
0.30	√	7.43	46.97	21.67	27.76

L^* : lightness (0=black, 100=white), a^* : red-green coordinates (positive values=red, negative values=green), b^* : yellow-blue coordinates (positive values=yellow, negative values=blue).

Table 2. *K/S* and CIE $L^*a^*b^*$ values of dyed cotton yarn samples with or without sodium chloride present. Untreated cotton yarn refers to no additive pre-treatments before dyeing, and post-mordanting treatment with *M. scutellatum* extract or alum was on untreated dyed cotton yarn. All cotton samples were dyed with the dye extract solution from *C. latifolium*.

Cotton yarn	<i>K/S</i> ($\lambda = 370$ nm)	CIE		
		L^*	a^*	b^*
Untreated cotton yarn (control)	1.16	80.24	-0.01	17.94
Untreated cotton yarn dyeing in the presence of sodium chloride	1.96	75.21	1.36	20.76
Post-mordanting with aqueous leaf extract of <i>M. scutellatum</i>	3.62	71.00	-0.46	32.85
Post-mordanting with alum	3.77	68.05	-0.14	25.66

L^* : lightness (0=black, 100=white), a^* : red-green coordinates (positive values=red, negative values=green), b^* : yellow-blue coordinates (positive values=yellow, negative values=blue).

Table 3. The color obtained and fastness properties of cotton dyed with the aqueous stem extract solution of *C. latifolium* with various additive treatments.

Mordants	Color obtained	Light fastness ¹	Wash fastness ²	
			Color change	Staining
Untreated cotton yarn (control)	Pale yellow	2	2	2-3
Untreated cotton yarn dyeing in the presence of sodium chloride	Yellow	2	2	2-3
Pretreated with a 0.30% w/v aqueous solution of chitosan	Brown	4-5	4-5	4
Pretreated with a 0.30% w/v aqueous solution of chitosan and then cross-linking with a 4% v/v glyoxal solution	Yellow	4-5	4-5	4
Post-mordanting with the aqueous leaf extract of <i>M. scutellatum</i>	Yellow	4-5	4-5	4
Post-mordanting with Alum	Dark yellow	3	4-5	4

¹The rating scale of light fastness is from 1 (poor) to 8 (excellent).

²Wash fastness was evaluated according to ISO105-C01: 1989(E).

The rating scale of wash fastness (both color change and color staining to adjacent fiber) is from 1 (poor) to 5 (excellent).

Color Hue and Fastness Properties of the Dyed cotton

Table 1 lists K/S and CIE $L^*a^*b^*$ values of dyed cotton yarn samples. It was found that the K/S values increased with increasing concentration of the chitosan solution indicating better dye receptivity of the substrate. However, cotton yarn pretreated with a 0.60 and 1.20 % w/v aqueous solution of chitosan gave an uneven coating which resulted in a variable distribution of adsorbed dye on the cotton yarn and the K/S and CIE $L^*a^*b^*$ values in these two cases were not determined. All samples had a yellow shade with different CIE $L^*a^*b^*$ values. Cotton yarn pretreated with a more dilute aqueous solution of chitosan (0.30% (w/v) provided better depth of shade and an even dye coating. A similar even dye coating and

smooth surface was obtained when glyoxal was used to chemically link the chitosan to the cotton yarn.

The effect of post-mordanting, as well as the effect of sodium chloride in the dye bath, on depth of shade and color hue is shown in Table 2. The aqueous extract of the leaves of *M. scutellatum*, which has been shown to promote the adsorption of flavonoid dye from the leaves of *E. odoratum* on cotton yarn [11] and the adsorption of laccic acid on silk [25], was also found to give a higher depth of shade (*K/S*) than the untreated cotton yarn (control); similarly sodium chloride in the dye bath had a positive effect on the *K/S* value but significantly less than the post-biomordant effect. The mordant effect of this plant extract is thought to be mediated via the significant concentration of tannins present [11]. A better depth of shade resulted, however, in the dyed cotton yarn pretreated with chitosan solution with or without cross-linking (Table 1). In addition, post-mordanting with alum after dyeing also gave a higher depth of shade (*K/S*) than in the untreated cotton yarn (control), and comparable to that observed with the bio-mordant treatment. Flavonoid complex formation with aluminium (III) ions from the alum is expected to be a significant factor in the former case; consistent with this, a bathochromic shift of 40 nm was observed in the UV-Visible absorption band at λ_{max} 370 nm on treatment of the extracted dye solution with Al(III).

The color obtained and fastness properties of dyed cotton with the dye extract solution from *C. latifolium* are summarized in Table 3. It can be seen that pretreatment of cotton yarn with chitosan before dyeing afforded material with good light and wash fastness, as did the pretreated cotton yarn sample with the combination of glyoxal and chitosan, as compared to the other samples. This may be due to the glyoxal binding chitosan to the cellulose fibres via acetal formation [11]. Post-mordanted dyed cotton yarn with the aqueous leaf extract solution of *M. scutellatum* or with alum gave very good to good light and wash fastness properties, while the control sample (dyed untreated cotton yarn) and untreated cotton yarn dyed in the

presence of sodium chloride, showed very low fastness to light and washing because of the low affinity between the dye molecules and cotton yarn.

Conclusions

The flavonoid dye from the aqueous extract of the stems of *C. latifolium* has promise for use as a natural dyestuff for cotton dyeing in the presence of other additives. While the addition of sodium chloride increased the amount of dye uptake at equilibrium compared to that in the absence of sodium chloride, the light and wash fastness properties of the resulting dyed cotton were poor. Pretreatment of cotton with chitosan solution (with and without a cross-linking glyoxal solution) increased the binding sites for the adsorption of the flavonoid dyes resulting in better dye adsorption on the cotton yarn. In particular, pre-treatment with a chitosan solution followed by cross linking with glyoxal and then exposure to the dye extract afforded an evenly dyed, bright yellow cotton yarn with very good color hue and fastness properties. Post-mordanting with the tannin-containing aqueous extract of leaves of *Memecylon scutellatum* also afforded dyed cotton yarn with good fastness properties and was somewhat better in terms of light fastness in comparison with alum post-mordanting.

Aspects of this work, particularly the bio-mordant results, should be directly applicable in the village context with potential future economic and environmental benefits and a decrease in the use of synthetic yellow dye and alum for the dyeing of cotton. Future work will include the characterization of the flavonoid components in the dye extract and the dyeing properties of the individual major colored components.

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References

- [1] Department of Pharmaceutical Botany, Mahidol University Foundation, *Encyclopedia of Herbs* (Bangkok: Amarin Printing & Publishing Public, 2000) (2543 Thai year) Vol. 4, 69.
- [2] V Brun and T Schumacher, *Traditional Herbal Medicine in Northern Thailand* (Bangkok: White Lotus, 1994) 257.
- [3] D L Shrishia, K A Raveesha and Nagabhushan, *J. Med. Plants Res.*, **5** (2011) 4087.
- [4] A A Elegami, E J El-Nima, M S El Tohami and A K Muddathir, *Phytother. Res.*, **16** (2002) 555.
- [5] P T A Dao, N X Hai, N T Nhan, T L Quan and N T T Mai, *Nat. Prod. Sci.*, **18** (2012) 1.
- [6] W Nopsiri, S Chansakaow, S Putiyanan, S Natakankitkul and D Santiarworn, *CMU J. Nat. Sci.*, **13** (2014) 195.
- [7] H Usman, F I Abdulrahman and A Usman, *Afr. J. Tradit. Complement. Altern. Med.*, **6** (2009) 289
- [8] J N Eloff, D R Katerere and L J McGaw, *J. Ethnopharmacol.*, **119** (2008) 686.
- [9] J D Carr and C B Rogers, *S. Afr. J. Bot.*, **53** (1987) 173.
- [10] P Kampeerapappun, T Phattararittigul, S Jittrong and D Kullachod, *Chiang Mai J. Sci.*, **38** (2010) 95.
- [11] M Chairat, U Darumas, J B Bremner and P Bangrak, *Color. Technol.*, **127** (2011) 346.
- [12] M Moeyes, *Natural Dyeing in Thailand*, (Bangkok: White Lotus, 1993) 67.
- [13] S Rattanaphani, M Chairat, J B Bremner and V Rattanaphani, *Dyes Pigment.*, **72** (2007) 88.

- [14] N V Majeti and K Ravi, *React. Funct. Polym.*, **46** (2000) 1.
- [15] S Saxena, V Iyer, A I Shaikh and V A Shenai, *Colourage*, **44** (1997) 23.
- [16] J A Rippon, *J. S. D. C.*, **100** (1984) 298.
- [17] Q Yang, F Dou, B Liang and Q Shen, *Carbohydr. Polym.*, **59** (2005) 205.
- [18] K Wolfe, X Wu and R H Liu, *J. Agr. Food Chem.*, **51** (2003) 609.
- [19] A Pekal and K Pyrzynska, *Food Anal. Methods*, **7** (2014) 1776.
- [20] D B Judd and G Wyszecski, *Color in Business, Science and Industry*, 3rd Edn (New York: John Wiley and Sons, 1975) 423.
- [21] S Janhom, P Griffiths, R Watanesk and S Watanesk, *Dyes Pigm.*, **63** (2004) 231.
- [22] W S Perkins, *Textile Coloration and Finishing* (North Carolina: Carolina Academic Press Durham, 1996) 126.
- [23] M Chairat, S Rattanaphani, J B Bremner and V Rattanaphani, *Dyes Pigm.*, **64** (2005) 231.
- [24] M Chairat, S Rattanaphani, J B Bremner and V Rattanaphani, *Dyes Pigm.*, **76** (2008) 435.
- [25] P Kongkachuichay, A Shitangkoon and N Chinwongamorn, *Dyes Pigm.*, **53** (2002) 179.