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A novel bioactive fabric dyed with unripe *Citrus grandis* Osbeck extract part I: dyeing properties and antimicrobial activity on cotton knit fabrics

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Abstract

In order to explore the potential use of *Citrus grandis* Osbeck in biomedical applications for improving human's skin health, cotton knit fabric was dyed with the citrus extract under varied conditions including temperature, duration, pH, and dye concentration and evaluated for fastness and antimicrobial activity. The fabric was optimized for maximum dye uptake at a pH of 3, dyeing temperature of 60°C, dyeing duration of 60 min, and dye concentration of 800% owf. The dye fastness to rubbing, washing, perspiration, and light were reasonably good for 800% owf dyed fabrics. In terms of antimicrobial efficacy, fabric dyed with the high concentration showed excellent bacterial reduction rates against *Staphylococcus aureus* for up to five repetitions of laundering, which implies there is a need to improve antimicrobial durability. In a following work, the optimally dyed fabric will be tested for in-vivo dermal effects.

Keywords

Antimicrobial activity, bioactive fabric, *Citrus grandis* Osbeck, dye fastness, functional dyeing

There have been increasing needs to impart antimicrobial and infection-resistant properties into textiles such as inner clothing which comes into direct contact with human skin because the growth of microbes on it may negatively affect the wearers as well as the textile itself.¹ The active ingredients applied in antimicrobial finishing are currently preferred if they are safe, non-toxic, and furthermore environmentally-friendly in order to meet worldwide consumers' desire for comfort, hygiene, well-being, and sustainability. For these reasons, a number of reviews on the application of natural resources have been performed in which human-friendly characteristics of natural materials were applied as enzyme treatments, chitosan finishes, dyeing with natural colorants, and so on.²

Dyeing with natural products has been employed for a long time not only for coloring textile but also for imparting various functions on fibers. As natural dyes were progressively replaced by the chemicals, they were overlooked for several decades. However, natural dyes have attracted the most attention recently again in research fields and in industries. Application of naturally functional materials to fibers via dyeing can be a type of functional dyeing technology which has many advantages both economically and environmentally.^{3,4}

This has stimulated a number of papers to address a variety of natural raw materials which have been traditionally used as dye stuffs by newly exploring their antibacterial, anti-fungal, and deodorizing effects as well as dyeing procedures, shades of dyed fabrics, and fastness properties.^{2,5–9} In addition, some new natural dyes have been investigated in terms of their antimicrobial effects with potential use as antibiotic finishing agents.^{10–12}

Citrus grandis Osbeck (Dangyuja in Korean) is one of the citrus species that originated in Jeju Island, Korea. Its fruit has been used for a long time as a traditional herb medicine.¹³ Recently, anti-cancer and antioxidant activities of the citrus fruit extract were reported^{14,15} and they were thought to be caused by flavonoids, including naringin, hesperidin, and

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nobiletin, in the citrus fruit.¹⁶ These flavonoids were revealed as being present at higher concentrations when the fruits were premature.^{17,18} This indicates that the unripe citrus fruits are expected to exhibit medicinal efficacies more. Actually, the unripe fruit extract of *Citrus grandis* Osbeck was found to show an anti-inflammatory effect in a skin cell model.¹⁹ A few studies involving dyeing fabric with some citrus fruits have been performed,^{20,21} but little study has been reported dealing with the functional aspects like antibacterial activity. In addition, the unripe fruit has not been used yet for any functional treatment on fabrics. Therefore, it is considered as worthwhile to employ unripe fruit of *Citrus grandis* Osbeck as a novel hygienic and medicinal agent on fabric via dyeing.

In terms of evaluation methodology, most of the studies incorporating bioactive agents into textiles are still concerned with in-vitro antibacterial activities based on the degree of inhibiting the growth of a bacterium on fabric. More recently, textile materials have been regarded as significantly influential on skin disorders such as irritation, allergic responses, and even dermal illness.²² These have forced textile manufacturers to explore medicinally functional textiles for wear. Nevertheless, less study has been done focusing on in-vivo evaluation using human subjects or animal models in the process of developing new skin health-related fabrics. Therefore textiles for skin health and medicinal effects now need to be investigated with in-vivo tests in order to identify the active involvement of them in establishing an optimal environment for improving dermal health.

In this context, we attempt to produce a novel functional fabric for skin health treated with *Citrus grandis* Osbeck by a dye technology and to evaluate its medicinal efficacy by in-vivo dermatological observations of humans who wear it as well as by in-vitro quantification of its antimicrobial activity. More specifically, the objectives of this study in Part 1 are to investigate the use of *Citrus grandis* Osbeck extract as a dye on cotton knit fabrics and fastness properties of the dyed fabric, and to evaluate antimicrobial activity of the dyed fabric quantitatively according to laundering, in order to optimize the dyeing conditions for a potential skin health-care fabric. In-vivo dermal functions of the dyed fabric will be discussed in a following part of this paper.

Experimental

Materials

A number of fresh unripe *Citrus grandis* Osbeck grown in Jeju Island, Korea were picked in August. They were crushed and sequentially extracted by being soaked in

80% aqueous ethanol solution for 4 hrs at 60°C. After filtration, the liquor extract was concentrated for 3 hrs at 40°C under reduced pressure with a vacuum rotatory evaporator and after adding 20 L of distilled water to the remaining residue, it was concentrated once more to remove any residual alcohol. The combined extract was evaporated and frozen to final solid content of dye powder. Cotton knit fabric (jersey, 0.73 mm, 157 g/m²) generally used for inner wear was desized and scored prior to dyeing. The pH conditions of the dyeing bath were adjusted by using citric acid for acidic conditions and by sodium hydroxide for alkaline, respectively.

Characterization of dyes

FT-IR transmission spectra were recorded, using KBr plates, by a spectrophotometer (IFS-66/S, Bruker Optik, Germany) in order to identify the structure of the citrus extract. In transmittance mode, 32 scans at 4 cm⁻¹ resolution over the range of 700–4000 cm⁻¹ were collected. A background scan of the clean KBr plate was also obtained.

Dyeing procedures

The cotton fabric was dyed at a liquor ratio of 1:50 under a variety of conditions including pH (2–9), dye concentrations (100–900%, owf), dyeing temperatures (40–90°C), and durations (20–90 min) in order to determine the optimum conditions for maximum dye uptake. A fixed bath temperature for each dyeing process and a constant stirring velocity were maintained by a shaking water bath (SWB-10, Jeio Tech, Korea). After dyeing, the fabric was rinsed with cold water and line-dried at ambient temperature.

Dye uptake and color measurement

Dye absorbed on the fiber and color properties were measured using a spectrophotometer (GretaMacbeth Color Eye 7000A, USA) under illuminant D65, with a 10° standard observer. The K/S value which has been employed as the dye uptake or color strength was calculated for each dyed specimen as well as for an undyed one at a maximum absorption wavelength of 360 nm from the reflectance values using the Kubelka-Munk equation as follows;

$$K/S = (1 - R_{\lambda \max})^2 / 2R_{\lambda \max} \quad (1)$$

where K is the coefficient of absorption; S is the coefficient of scattering; $R_{\lambda \max}$ is the reflectance value of the fabric at peak wavelength.

As for color properties of the dyed fabric, CIE L^* , a^* , b^* , and C^* were obtained. L^* describes lightness; a^*

measures redness or greenness; b^* measures yellowness or blueness; and C^* means color saturation or strength in the CIE system. The color difference, ΔE was calculated according to equation 2 in order to investigate the apparent colors of dyed fabrics compared with those of undyed fabric.

$$\Delta E = (\Delta L^{*2} + \Delta a^{*2} + \Delta b^{*2})^{1/2} \quad (2)$$

Fastness evaluation

Fastness was investigated on the dyed cotton knit for rubbing, washing, perspiration, and light. Dry and wet rub fastness was tested by KS K 0650; washing fastness was examined by KS K ISO 105 C06:2006; perspiration fastness was evaluated with KS K ISO 105 E04:2005; and light fastness was measured by KS K 0700. Fading and staining of adjacent fabrics in color were determined using gray scales.

Determination of antimicrobial activity of dyed fabrics

The antimicrobial activity of the cotton knit fabrics dyed with *Citrus grandis* Osbeck extract was evaluated by a quantitative measurement (KS K 0693:2006). The test samples were prepared by dyeing with 800%, 600%, and 300% owf of the extract, respectively at the optimum conditions (60°C, 60 min) in order to determine dye concentrations suitable for antimicrobial purposes. The bacteria used were *Staphylococcus aureus* (*S. aureus*, ATCC 6538) and *Klebsiella pneumoniae* (*K. pneumoniae*, ATCC 4352). The visible bacteria colonies on the agar plate were counted and the percentage reduction of the bacterium was calculated as follows;

$$R(\%) = (B - A) / B \times 100 \quad (3)$$

Where R is the percentage reduction of the bacterium; B is the number of bacterial colonies from an untreated fabric, and A is the number of bacterial colonies from the dyed fabrics. The effects of laundering on the antimicrobial activities of the dyed fabric were also investigated for one, five, and ten pre-washes, respectively, performed by KS K ISO 105 C06:2006, which was the same as the test standard used for wash fastness.

Results and discussion

Chemical characterization of the citrus extract

Figure 1 shows FT-IR transmission spectra of prepared unripe *Citrus grandis* Osbeck extract. A strong absorption band centered at 1070 cm^{-1} indicated the existence

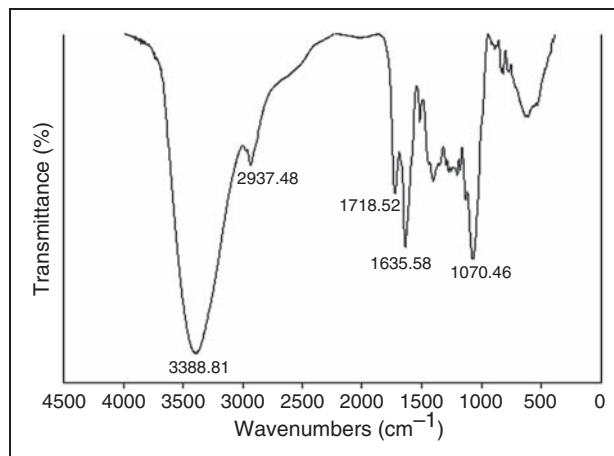


Figure 1. FT-IR spectra of unripe *Citrus grandis* Osbeck extract.

of an ether (OCH_3) of nobiletin, a type of flavonoid. Therefore the extract used in this study was characterized as containing some flavonoids, which are reported as providing some medicinal effects.¹⁶ Other functional flavonoids such as naringin and hesperidin seemed not to exist in the extract of unripe *Citrus grandis* Osbeck because they have been reported as showing peaks at 1645 cm^{-1} and 1605 cm^{-1} , respectively, in the FT-IR spectrum. Another strong intensity peak corresponding to the $\text{C}=\text{O}$, a carbonyl group which is a common functionality in flavonoids, was observed at 1635 cm^{-1} . The deepest peak at 3388 cm^{-1} may be caused by an OH group in the carboxylic acid and another one at 2937 cm^{-1} suggests the presence of C–H, all of which were known to be frequently contained in organic compounds. In addition, a band centered at 1718 cm^{-1} could indicate the existence of a $\text{C}=\text{O}$ stretching vibration of carboxylic acids in citric acids.

Effect of dyeing conditions on dye uptake

In order to identify the optimum dyeing condition for cotton knit with extract of unripe *Citrus grandis* Osbeck, various conditions of pH environments, dyeing temperature, time, and concentrations of dye bath were investigated along with dye uptake, K/S values are as given in Figures 2–5. Figure 2 shows the effects of pH values in the dye bath on dye uptake, K/S. Generally, among various pH conditions manipulated by citric acid or sodium hydroxide, an acidic condition is preferred for increased dye uptake, K/S. In particular, pH 3 seemed to provide the highest K/S for all of dye concentrations from 100% to 800% owf, which means that at that pH condition, cotton knits absorbed dyes the most. Thereafter as pH increased to alkaline, K/S values tended to decrease. Thus, high pH should be avoided in the dyeing of cotton with the citrus extract.

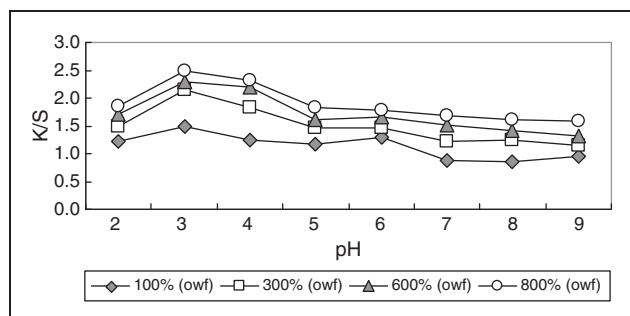


Figure 2. Effect of pH on dye uptake of cotton knits (60°C, 60 min).

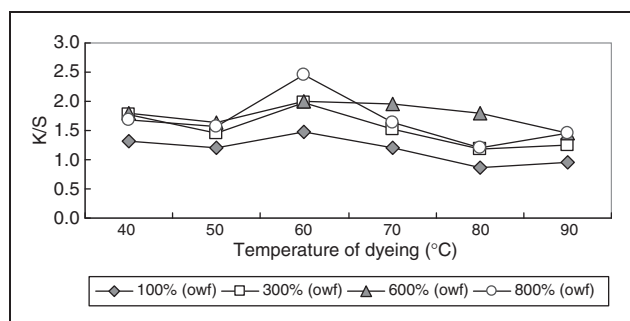


Figure 3. Effect of dyeing temperature on dye uptake (60 min).

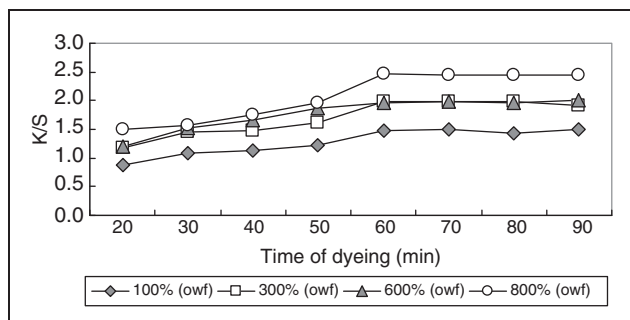


Figure 4. Effect of dyeing time on dye uptake of cotton knits (60°C).

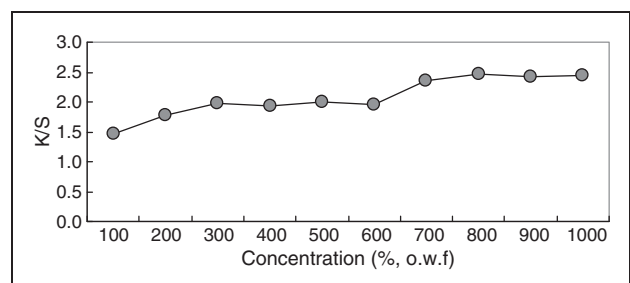


Figure 5. Effect of dye concentration on dye uptake (60°C, 60 min).

A pure aqueous solution of the extract without any pH modification showed pH values from 3.2 to approximately 3.5. Therefore we carried out further experiments without any pH regulation in order to minimize additives for dyeing because this study ultimately aimed to investigate human-friendly dermal functions of the citrus-dyed fabrics.

The temperature of the dye bath was varied to determine the optimum dyeing temperature for several levels of dye concentration. Figure 3 illustrates dye uptake on cotton fabrics at temperatures that varied from 40 to 90°C in the bath. A dyeing temperature of 60°C seemed to maximize the K/S values for every dye concentration. It should be noted that much higher temperatures than 60°C for the dye bath resulted in a decrease of dye uptake. For example, for the dye concentration of 800%, the K/S value increased abruptly up to 2.5 at approximately 60°C and thereafter it decreased rapidly down to 1.2 at around 80°C. In general, higher dye temperatures such as 80°C and 90°C are likely to cause even less affinity of dyes with cotton fibers because the K/S which raised up at 60°C tended to decrease above 60°C at all of concentrations. From this result, it was concluded that the optimum dyeing temperature of unripe *Citrus grandis* Osbeck is 60°C for cotton.

The influence of dyeing duration from 20 to 90 min on dye uptake was also examined and presented in Figure 4. Up to 60 min of dyeing time, the K/S values increased gradually for all dye concentrations, thereafter they reached equilibrium. The concentration of 800% owf gave much higher dye uptake value when it was applied for 60 min duration. Although 300% and 600% owf dye bath did not show any obvious differences to each other at any of the dyeing durations, both of them also showed the saturation of dye uptake at 60 min duration. It could be concluded that the optimum dyeing time for the extract of unripe *Citrus grandis* Osbeck on cotton is 60 min.

Finally, K/S values were investigated in terms of dye concentrations under the condition of 60°C and 60 min as shown in Figure 5. The K/S increased gradually with an increasing concentration of unripe *Citrus grandis* Osbeck up to approximately 800% owf extract in the bath. We determined 800% owf to be the optimum concentration of the citrus extract for dyeing cotton knits because the increase of K/S plateaued above the concentration of 800%.

Color properties of the dyed fabrics

Colorimetric properties of cotton knit dyed with unripe *Citrus grandis* Osbeck extract were investigated by the CIE color system as shown in Table 1. All of dyed fabrics seemed to be colored lighter rather than

Table 1. Color properties of cotton knits dye with unripe *Citrus grandis* Osbeck extract according to dye concentration

Dye Concentration (owf)	CIE Color Properties				
	L*	a*	b*	C*	ΔE
undyed	93.34	0.35	−0.18	0.39	–
100%	85.63	−4.77	15.89	16.59	4.72
300%	82.48	−5.76	19.13	19.98	5.16
600%	80.05	−6.4	19.40	20.43	5.30
800%	78.98	−6.64	20.30	21.36	5.42

darker because their L* values were much higher. As for C*, color saturation, the values were lower, which means they colored less strong. In general, unripe *Citrus grandis* Osbeck extract was thought to give light and weak shade rather than dark or strong to cotton knits considering C* together with L*. As for shade or hue, the dyed fabrics showed slightly negative a* but highly positive b*, which means that they gave shades of greenish yellow. As dye concentration increased, color lightness, L* got lower whereas color saturation, C* did increase, which means that the dyed cotton knits which underwent higher dye concentrations were colored stronger whereas less light. Nevertheless, even by maximum dye uptake under the optimum dye condition (800% owf, 60°C, 60 min), cotton knits showed soft or pale tone predicted by CIE color values.

Dye fastness

Dye fastness rates for rubbing, washing, perspiration, and light are summarized in Table 2. Rubbing fastness rates of 600% and 800% dyed fabrics assessed in terms of dry and wet rubbing were very good (4–5) whereas those of fabrics that underwent 300% and 100% were good (3–4). Fastness to washing was also very good for 600% and 800% fabrics in terms of staining whereas it was good (3–4) or fair (2–3) for the two other concentrations, that is, 100% and 300%. However color change rates in washing were good only for 800% fabric whereas the other fabrics showed lower rates for it. The ratings for staining of adjacent fabrics in fastness to perspiration were generally very good for 600% and 800% fabrics while good or fair for the other two concentrations. On the contrary, shade changes in perspiration were not very good (3–4). In terms of light fastness, 100% and 300% dyed fabric had fair grade (2–3) for their shade changes with light whereas both of 600% and 800% fabric showed good grades (3–4). However all of the dyed fabrics failed to obtain very good grades for light fastness, similar to shade changes by perspiration, which means this

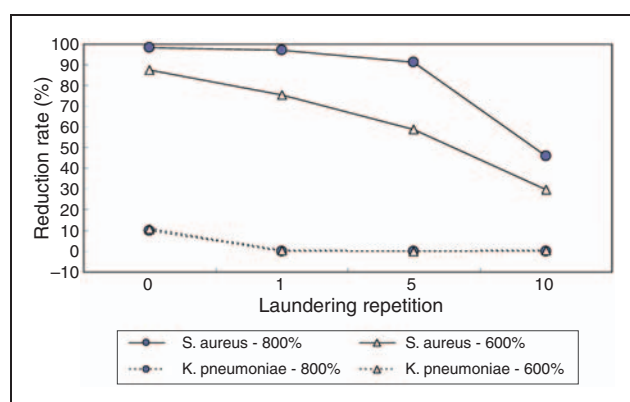
needs to be improved. In general, 800% dyed cotton knit was fairly acceptable for all dye fastness parameters considering that natural materials usually display very low substantivity towards fibers. Therefore it can be considered that *Citrus grandis* Osbeck extract provides acceptable fastness to rubbing, washing, and perspiration when it is dyed on cotton under higher dye concentrations. More specifically, the dyed fabric with 800% of extract was good for most fastness parameters whereas the fabric with 600% showed only generally good fastness. However, the improvement of fastness to shade changes by washing, perspiration, and light dyeing of *Citrus grandis* Osbeck extract on cotton could be recommended for a future study.

Antimicrobial activity

The antimicrobial efficacy of cotton knit fabrics dyed with unripe *Citrus grandis* Osbeck extract was evaluated using a quantitative analysis by bacterial reduction rate. Figure 6 presents the bacterial reduction rates according to laundering repetition of the dyed fabrics by two different dye concentrations, that is, 600% and 800%, which showed acceptable dyeing fastness as mentioned above. Against *S. aureus*, the dyed fabric with 800% showed an excellent bacterial reduction rate of 98.3% in bacterial growth on it before initial laundering, which means that the dyed fabric possesses microbial effects regarded as sufficiently acceptable for sanitary uses. It can be seen that very effective antimicrobial activity for the fabric was displayed up to five repetitions of laundering. These results imply that employing unripe *Citrus grandis* Osbeck extract as a functional dye is a useful approach to achieve the desired antimicrobial properties, as well. However after ten repetitions of laundering, the bacterium reduction rate dropped to 45.7%. It is thought that the low laundering durability of antimicrobial efficacy may be caused by a lack of affinity of the citrus to cotton fibers which could not overcome repeated laundering. Although the higher concentration dye-treated fabric was evaluated as good for the fastness to washing as mentioned above, it could not maintain significant antimicrobial activity against *S. aureus* after ten laundering cycles. On the other hand, the fabric dyed with the citrus extract of 600% owf showed 87.3% of bacterial reduction rate against *S. aureus* before laundering which was much lower than that of its counterpart for 800%. As laundering was repeated, its bacterial reduction rates reduced to 29.5% after ten cycles of laundering. The antimicrobial activity against *K. pneumoniae*, however, was not acceptable, so even before laundering very low reduction rates (10.2% for 800% and 11.0% for 600% dyed fabric) were observed. Moreover, after laundering, antimicrobial activities

Table 2. Dye fastness to rubbing, washing, perspiration, and light of cotton knit dyed with unripe *Citrus grandis* Osbeck extract

				Dye Concentration (% owf)			
				100%	300%	600%	800%
Fastness							
Rub	Dry			3–4	3–4	4–5	4–5
	Wet			3–4	3–4	4	4–5
Wash	Shade Change			1–2	2	2	4
	Staining	with Cotton		2–3	3–4	4	4–5
		with Wool		3	3–4	4–5	4–5
Perspiration	Shade Change	Alkaline		2–3	2–3	3–4	3–4
		Acid		2	2–3	2–3	3–4
	Staining	Alkaline	with Cotton	2–3	2–3	4	4–5
			with Wool	3	3–4	4–5	4–5
		Acid	with Cotton	2	3–4	4	4–5
			with Wool	3	3–4	4–5	4–5
Light			2–3	2–3	3–4	3–4	

**Figure 6.** Antimicrobial activity of cotton knit fabric dyed with unripe *Citrus grandis* Osbeck extract according to dye concentration and laundering repetition.

against *K. pneumoniae* were non-existent for both of them. These results indicate that unripe *Citrus grandis* Osbeck extract has no ability to destroy *K. pneumoniae* when it is dyed on cotton fibers. Generally speaking, the laundering durability of antibacterial properties for the dyed fabrics was fairly good. The dyed fabric with 800% dye concentration showed high antimicrobial efficacy against *S. aureus* even up to five repetitions of laundering. Therefore, in a future study, an effort needs to be made to improve the laundering durability for the antimicrobial activity of cotton fabric dyed with unripe *Citrus grandis* Osbeck extract.

Conclusions

This paper aimed to produce antimicrobial fabrics by employing unripe *Citrus grandis* Osbeck extract for a

potential use in bioactive applications for human skin health. Cotton knit fabric was dyed with the citrus extract at a range of dyeing temperatures, durations, pHs, and dye concentrations in order to optimize its dyeing properties. Cotton knit showed maximum dye uptake when it was treated in the dye bath with 800% owf dye concentration at 60°C and 60 min, by estimating K/S value. As for pH regulation, it was concluded that any pH modification of the dye bath was not required because cotton seemed to absorb the citrus extract the most at pH 3 which is near to the value for the pure aqueous solution of the extract. The fastness properties were reasonably good against rubbing, washing, and perspiration for the 800% owf dyed fabrics and furthermore recommended to increase its fastness to shade change by washing, acid perspiration, and light to reach very good grades. The dyed cotton fabric with 800% was highly effective against *S. aureus*, reducing the number of colonies by 98.3% and its efficacy remained after five laundering cycles.

These results lead us to the conclusion that dyeing with unripe *Citrus grandis* Osbeck extract on cotton will be useful in providing antimicrobial efficacy to textiles such as inner clothing, medical drapery, and sheets with hygienic uses. The influence of additives to improve dye fastness and microbial durability such as metallic mordants or chitosan is a further area of interest because the present study focused on the application of a non-toxic, eco-friendly, and multi-functional natural resource to sanitary textiles as a medium to improve skin-health. Also in a future study, adding inorganic electrolyte to the dye bath should be employed to improve dyeability of cotton. In a following paper, experimental in-vivo human tests will be reported in order to examine if the dyed fabric is

non-toxic, not irritant, and furthermore efficient on dermal problems such as atopic dermatitis.

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