Journal of Metals, Materials and Minerals, Vol. 32, No. 4, pp. 71-78, 2022

Ecological dyeing of acrylic yarn with colorant derived from natural lac dye

Nattaya VUTHIGANOND[1,*], Pisutsaran CHITICHOTPANYA[1], Monthon NAKPATHOM[2], and Rattanaphol MONGKHOLRATTANASIT[3]

[1] *Division of Materials and Textile Technology, Faculty of Science and Technology Thammasat University, Rangsit Center, Pathumthani 12121, Thailand*
[2] *Textile Lab, Polymer Research Unit, National Metal and Materials Technology Center, National Science and Technology Development Agency, Pathumthani 12120, Thailand*
[3] *Department of Textile Chemistry Technology, Faculty of Industrial Textiles and Fashion Design, Rajamangala University of Technology Phra Nakhon, Bangkok 10300, Thailand*

***Corresponding author e-mail:** pnattaya@tu.ac.th

Received date:
17 August 2022
Revised date
13 September 2022
Accepted date:
1 October 2022

Keywords:
Dyeing;
Acrylic;
Natural dye;
Lac;
Fastness

Abstract

In recent years, the application of natural dyes in textile coloration has attracted significant study interest. In this current study, the bio-based dyeing of synthetic textile fiber, specifically acrylic yarn, with natural lac dye was investigated. The yarn was dyed using a meta-mordanting technique. The dyeing parameters, including temperature, time, pH of the dye bath, dye concentration, mordant type, and mordant concentration, were examined. Two eco-mordants, aluminium sulfate and ferrous sulfate, are compared against an eco-restricted copper sulfate in terms of color strength and fastness. The results demonstrated that the meta-mordanting technique can be used to dye acrylic fiber using natural lac dye. The optimum dyeing results were achieved using an acidic pH (2-3) and a temperature of 100°C for 40 min of dyeing time. The color strength of the dyed samples was enhanced by the application of mordants. Color uniformity was achieved by controlling the temperature gradient in the T_g region. In terms of color change, the wash fastness properties of the dyed samples were found to be poor to moderate due to the dye's susceptibility to alkaline washing. In terms of color fastness to rubbing and light, ferrous sulfate and copper sulfate produced superior results.

1. Introduction

The textile industry, particularly the textile coloration sector, has a significant impact on the ecosystem as a result of the use of a large amount of energy and water, the emission of greenhouse gases, the generation of hazardous waste, and the discharge of toxic effluent into the environment [1-3]. The effluent consists of dyes, finishing chemicals, and auxiliaries. Nowadays, an increasing number of people are becoming aware of the importance of environmentally responsible manufacturing practices in the clothing and textile industries [4,5]. The use of natural dyestuffs in the textile dyeing process is gaining popularity, and customer demand is increasing [6,7]. This is attributed to safety concerns over synthetic dyes [8].

Acrylic or polyacrylonitrile (PAN) fiber is one of the three most widely used synthetic fibers in the textile industry. Acrylic fibers are copolymers that include 85 percent or more by weight of acrylonitrile monomer. To reduce structural regularity and enhance dye diffusion, typical comonomers such as methyl acrylate, vinyl acetate, and methyl methacrylate are added. Moreover, monomers containing carboxylic, sulfonic acid, or phosphoric acid groups generate the dye site for cationic or basic dyes. Acrylic fiber shares similar features with wool, including elasticity, thermostability, photostability, softness, and bulkiness. Additionally, it is widely used in the apparel, home textiles, and decorating industries.

Lac dye is a natural reddish colorant (C.I. Natural Red 25; C.I. 75450) that is commonly used in the food, cosmetics, and textile sectors. As the alkaline salt, lac coloring is derived from the bodily fluid of the female insect *Coccus laccae (Laccifer lacca Kerr).* This insect inhabits the branches of trees native to South and Southeast Asia, particularly Thailand and India. Laccaic acids A and B [9-15], which are derived from anthraquinone, are the two primary coloring agents in lac. The minor components are laccaic acids C, D, and E [13-15]. In addition, lac dye contains an additional coloring agent called erythrolaccin [11]. Figure 1 depicts the structures of colorants found in lac dye.

Lac dye is frequently used in textile dyeing applications to dye natural fibers such as wool, silk, and cellulosic fibers in the presence of mordants to enhance dye-fiber affinity [16]. Mordants are metallic salts that act as electron donors to establish coordination bonds between dyes and fibers, hence enhancing dyeability and colorfastness. Aluminium potassium sulfate ($AlK(SO_4)_2$), potassium dichromate ($K_2Cr_2O_7$), ferrous sulfate ($FeSO_4$), and stannous chloride ($SnCl_2$) are the most typical metallic mordants [17,18]. As a result of their harmful effects on the environment and human health, however, the usage of some

DOI: 10.55713/jmmm.v32i4.1529

heavy metals is regulated, as is their presence in textile products. Aluminium potassium sulfate and ferrous sulfate are considered more sustainable and unrestrained by eco-regulation compared to other metal mordants used in natural dyeing [19,20]. Due of the hydrophobic and crystalline structure of synthetic fibers, the majority of natural dyeing research focuses on natural fibers. Some research on acrylic dyeing has employed berberine, a cationic natural dye [21], and modified acrylic fibers dyed with curcumin [22].

The current study aims to evaluate the dyeing characteristics of acrylic yarn using a bio-based dye derived from lac, utilizing the exhaustion dyeing process with non-toxic mordants, particularly aluminium potassium sulfate and ferrous sulfate. Furthermore, the dyed acrylic samples were compared to the commonly used effective mordant, copper sulfate ($CuSO_4$). The dyeing conditions, including pH, temperature, time, mordant type, and mordant concentration, were systematically studied.

2. Experimental

2.1 Materials and chemicals

Commercially available acrylic knitting yarn was purchased from Venus Thread Co., Ltd. (Thailand). The lac was sourced from Nan, a northern province in Thailand. Metallic salt mordants (aluminium potassium sulfate dodecahydrate ($AlK(SO_4)_2.12H_2O$), ferrous(II) sulfate heptahydrate ($FeSO_4.7H_2O$), and copper (II) sulfate pentahydrate ($CuSO_4.5H_2O$)) were obtained from Ajax Finechem Pty Ltd. (Australia). A non-ionic soaping agent was supplied by Boonthawee Chemephan Co., Ltd. (Thailand). All the other chemicals used in this study were of laboratory reagent grade.

2.2 Extraction of lac dyes

Lac was dried and finely ground with a blender. The aqueous extraction technique was used for extraction. Lac (30 g) was suspended in water (1 L). The extraction was performed in an infrared dyeing machine at 100°C for 1 h. After that, the liquor was then filtered.

2.3 Dyeing with lac dye

Acrylic yarn samples weighing 2 g were immersed in a 1:30 liquor ratio dye bath at room temperature. The meta-mordanting procedure was selected and a series of dyeing on acrylic yarn was performed using an infrared dyeing machine. Figure 2 depicts the dyeing profile utilized in this investigation, and Table 1 shows the dyeing parameters evaluated. After dyeing, the dyed samples were washed in a bath containing 2 $g{\cdot}L^{-1}$ non-ionic detergent at 60°C for 20 min to remove excess and unfixed dyes at a liquor ratio of 1:30. The samples were then cold rinsed thoroughly and dried at the ambient temperature.

2.4 Color measurements

A spectrophotometer was used to evaluate the color attributes CIE (L^*, a^*, b^*) and color intensity (K/S) of dyed acrylic samples (GretagMacbeth LLC, Switzerland). All measurements were conducted with an illuminant D65, a 10° standard observer, and specular and UV filters. The dyed samples were examined three times, with the average values recorded. The Kubelka-Munk equation was used to determine the K/S values.

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

Where K is the absorption coefficient, R is the reflectance of the samples at the maximum absorption wavelength, and S is the scattering coefficient.

2.5 The relative unlevelness index (*RUI*)

The color uniformity of dyed samples was assessed by measuring reflectance values at 10 nm intervals between 400 nm and 700 nm. Using a spectrophotometer, each sample was measured (GretagMacbeth LLC, Switzerland) with five randomly selected measuring regions of the dyed sample. The relative unlevelness index (*RUI*) was calculated using the following formula:

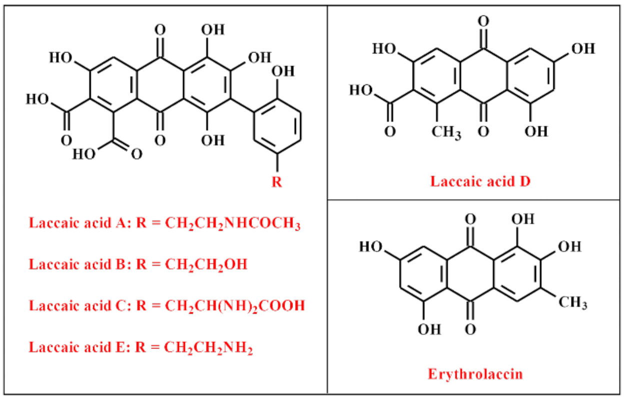


Figure 1. The chemical composition of several colorants in lac natural dye.

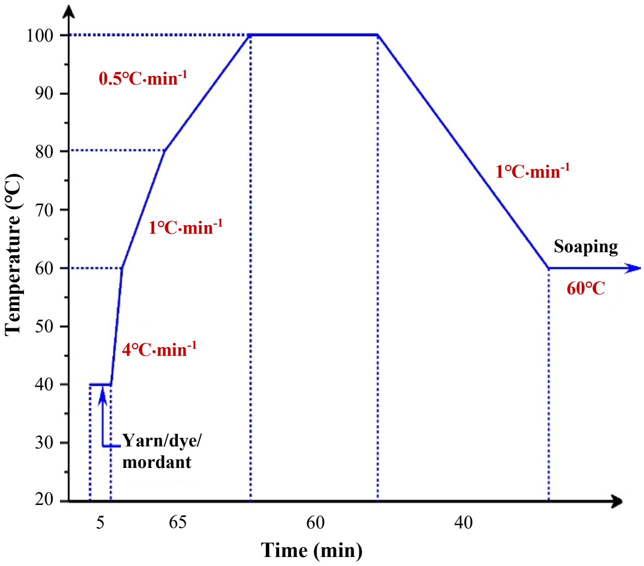


Figure 2. Lac dyeing profile for acrylic yarn.

Table 1. The dyeing parameters for acrylic dyeing with lac dye.

Dyeing parameters	**Values**
pH	2, 3, 4, 5, 7, 9, 10
Temperature (°C)	60, 70, 80, 90, 100
Time (min)	30, 40, 50, 60
Mordant types	None, $AlK(SO_4)_2$, $FeSO_4$, $CuSO_4$
Mordant concentrations ($g{\cdot}L^{-1}$)	1, 5, 10, 15, 20
Dye concentrations (% owf)	5, 10, 15, 20

$$RUI = \sum_{\lambda=400}^{700} \frac{S_\lambda}{R_\lambda} V_\lambda$$

Where R_λ is the mean reflectance values for each wavelength, S_λ is the standard deviation of reflectance values, and V_λ is the photopic relative luminous efficiency function [23]

The resulting value of *RUI* were used to demonstrate the degree of color levelness. Excellent and good levelness are indicated by *RUI* < 0.20 and 0.20-0.49, respectively. Poor and unacceptable levelness is defined as *RUI* values of 0.50 – 1.00 and > 1.00, respectively.

2.6 Color fastness testing

The color fastness to washing, light, and rubbing of the dyed samples was assessed according to ISO 105-C06 A1S:1994, ISO 105-B02:1994, and ISO 105-X12:2001, respectively.

2.7 Cross-sectional examination

The cross-section of the dyed sample was inspected and photographed at 100x magnification with a Nikon Upright Microscope Eclipse Ci-L plus.

3. Results and discussion

3.1 Optimization of the dyeing conditions

3.1.1 The effect of temperature

In terms of temperature, duration, and pH, the dyeing characteristics of acrylic yarn with lac natural dye in the presence of alum as a mordant are examined. To assess the influence of dyeing parameters, the color strength (*K/S*) and *a** (ranging from negative (green) to positive (red); the higher the *a** value, the redder the color) are measured. Acrylic fiber samples were dyed using 20% owf. dye, pH 3 (the extract's inherent pH), 10 $g{\cdot}L^{-1}$ of alum, and a dyeing time of 30 min. Figure 3(a) depicts the effect of dyeing temperatures on Al-mordant acrylic yarn. It is evident that both color intensity and *a** increase as the dyeing temperature rises. At 70°C, the dye uptake increased significantly. As a thermoplastic fiber, the glass transition temperature (T_g) is a crucial factor in controlling the dyeing behavior of acrylic fiber. Acrylic fibers typically have T_g range of 70°C to 85°C [24]. Below the T_g, polymer chain mobility is inadequate, and it is difficult for dye molecules to permeate into the polymer matrix. As soon as the temperature surpasses the T_g, the dyeing mechanism is started and dye absorption into the fiber begins to increase dramatically. Moreover, when temperatures increase, the mobility and diffusion of dye molecules within the fiber increases [25]. Consequently, the dissolution of dye aggregation in the dye bath was increased. Thus, the color intensity of the dyed yarn was increased. The dyeing temperature also affects the *a** values; as the temperature rises, the dyed samples become redder (Figure 3(b)). The samples dyed at 100°C with lac dye had the greatest color saturation and reddest hue.

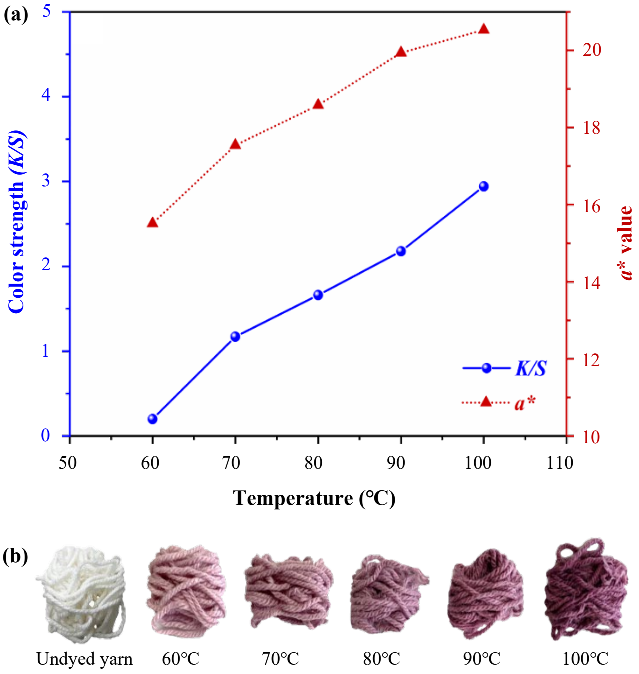


Figure 3. (a) *K/S* and *a** values of Al-mordant acrylic yarn as a function of dyeing temperature, and (b) apparent color of dyed samples at various dye bath temperatures (20% owf. lac, pH = 3, 10 $g{\cdot}L^{-1}$ $AlK(SO_4)_2$, 30 min).

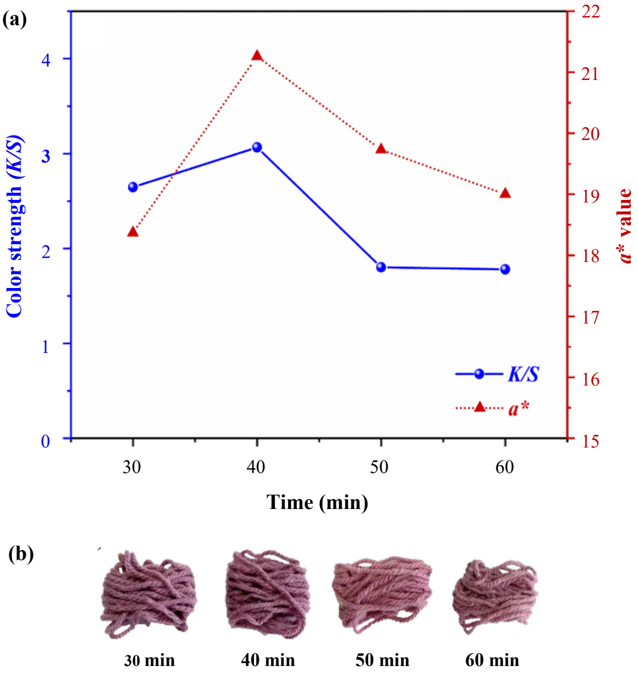


Figure 4. (a) *K/S* and *a** values of Al-mordant acrylic yarn as a function of dyeing time, and (b) apparent color of dyed samples at various dyeing times (20% owf. of lac, 10 $g{\cdot}L^{-1}$ $AlK(SO_4)_2$, pH = 3, 100°C).

3.1.2 The effect of dyeing time

Figure 4 shows the impact of the dyeing period on color strength (*K/S*) and *a**, as well as the perceived color of acrylic yarn dyed with lac dye. Both color strength and *a** were found to increase as the time increased. The maximum color strength and *a** were achieved at 40 min and then slightly declined as time progressed. The reduction in dye uptake of the acrylic fiber might be attributed to dye desorption of more dye molecules from the substrate as a consequence of the extended dyeing time [26].

3.1.3 The effect of pH

Different pH of the dye bath in the presence of alum (10 $g{\cdot}L^{-1}$) were adjusted in the ranges of 2 to 10 by using 1% NaOH or 1% HCl. The dyed samples were then evaluated in terms of the color strength (*K/S*) and *a** values. As shown in Figure 5(a), pH is an essential parameter affecting the color strength of Al-mordant acrylic yarn. In acidic pH, *K/S* and *a** increased significantly, with the highest *K/S* achieved in the pH range of 2-3. This indicates the great stability of metal chelation at low pH. After adjusting the pH of the dye bath to a neutral and basic pH, the hue of the dye bath gradually changed from red to purple. However, after the dyeing cycle was completed, the dyed samples exhibited the yellow color. This suggests that increasing the pH of the dye solution results in the dissociation of the color product from unstable metal-fiber complexes. The color of the Al-mordanted acrylic samples is displayed in Figure 5(b). At pH 2-5, the acrylic dyed with lac had a red color. The color loss is observed at a higher pH (pH ≥ 7), resulting in a yellow hue.

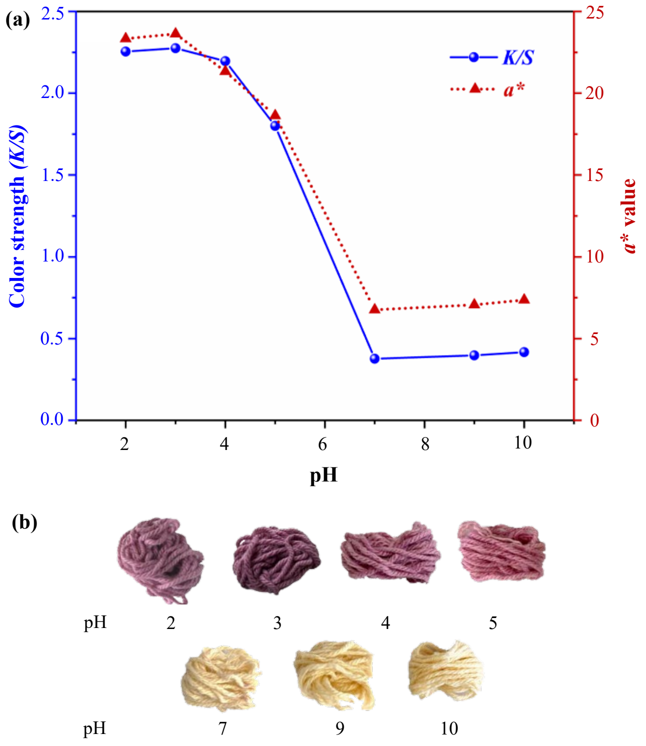


Figure 5. (a) *K/S* and *a** values of Al-mordant acrylic yarn as a function of dye bath pH, and (b) apparent color of dyed samples at various dye bath pH (20% owf. lac, 10 $g{\cdot}L^{-1}$ $AlK(SO_4)_2$, 100°C, 40 min).

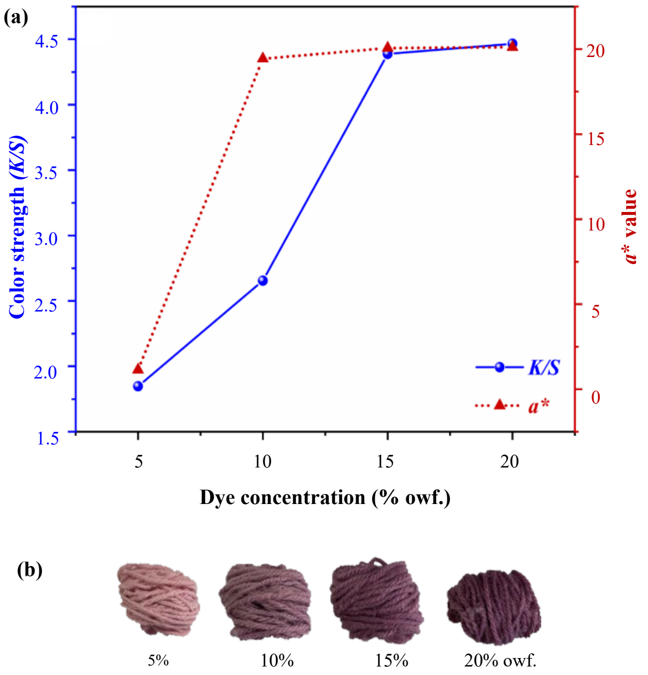


Figure 6. (a) *K/S* and *a** values of Al-mordant acrylic yarn as a function of dye concentration, and (b) apparent color of dyed samples at various dye concentrations (10 $g{\cdot}L^{-1}$ $AlK(SO_4)_2$, 100°C, 40 min, pH = 3).

3.1.4 The effect of dye concentration

The dyeing properties of Al-mordant acrylic yarn were analyzed in terms of the influence of dye concentration on color intensity (*K/S*) and *a**. Figure 6(a) demonstrates that the *K/S* increased sharply when the lac dye concentration increased from 5% to 10% owf. and then remained constant. Increasing the initial dye concentration enhanced the *K/S* of the dyed sample. This is because the concentration gradient of dye molecules by adsorption increases as the concentration of the dye bath increases [25]. However, the shade depth hit a plateau and there was no significant increase in *K/S* after further increases in dye concentration. During polymerization, the amount of acidic groups (COO^-, SO_3^-, and SO_4^-) incorporated into the polymer was controlled [24]. Therefore, the concentration of the dye-fiber coordination complex will be proportional to the acidic groups present in the polymer structures. Figure 6(b) shows the hue produced by Al-mordanted acrylic samples with varying dye concentrations.

3.1.5 The effect of metal mordant type and mordant concentrations

Various concentrations of alum, ferrous sulfate, and copper sulfate, ranging from 1 $g{\cdot}L^{-1}$ to 20 $g{\cdot}L^{-1}$, were utilized to examine the lac-dyeing performance of acrylic yarn. As demonstrated in Figure 7(a), all three mordants significantly enhance dye-fiber fixation, resulting in greater *K/S* values. The metal ions serve as electron donors to create insoluble coordination bonds between the color and textile strands. Copper sulfate was the most effective metal mordant on *K/S*, followed by ferrous sulfate and then alum. In addition, The *K/S* increased as metal concentrations increased and reached a maximum *K/S* of 7 for the mordanted sample containing 20 $g{\cdot}L^{-1}$ of copper sulfate. However, because copper sulfate is considered a hazardous chemical, ferrous sulfate and alum are excellent alternatives for environmentally friendly natural dyeing.

As illustrated in Figure 7(b), lac have no affinity for the acrylic fiber in the absence of a mordant. Metal ions are necessary for the yarn to acquire its deep color. It is generally known that metal mordants have a significant effect on both the color and the dye-fiber fixation. The hue of the alum-dyed samples kept the original hue of the lac natural dye with more vibrant red tones. Using ferrous sulfate as a mordant, samples exhibited darker black shades. The addition of copper sulfate generated hues that were dark reddish-black. Figure 8 depicts the proposed acrylic-metal-lac coordination complex. The color properties of mordanted samples are displayed in Table 2 using *CIE* color coordinates *(L*, a*, b*)*. *L** denotes lightness (the lower the *L**, the darker the shade), while *a** and *b** are chromaticity coordinates (+*a** represents redness, -*a** represents greenness, +*b** represents yellowness, and -*b** represents blueness). The results also demonstrated that the colorimetric parameters were greatly influenced by the type of mordant.

3.2 Dye penetration ability and color uniformity

The color images of the cross-section of acrylic yarn after dyeing with various mordants are shown in Figure 9. The saturated dyeing was clearly observed with a good distribution of dyes within the fibers. Table 3 demonstrates the relative unlevelness indices (*RUI*) and visual levelness assessment of dyed samples. *RUI* values obtained range between 0.33 and 0.38, showing good levelness. This result indicates that dyeing acrylic fiber with a meta-mordanting process can be effectively performed. Mordants and lac dye molecules were in non-precipitate form throughout the dyeing process. It is worth noting that another key factor affecting the level of dyeing of thermoplastic fibers is controlling the temperature gradient during dyeing, in which the dyeing rate above the T_g region is carefully controlled at a low rate (0.5°C$\cdot$min^{-1}) [24].

3.3 Fastness properties

The mordanted acrylic dyed specimens were tested for color fastness under optimum conditions. As shown in Table 4, the three primary tests are color fastness to washing, color fastness to rubbing, and color fastness to light. In the case of color fastness to washing and rubbing, both gray scales for color change and staining rating range from 1 to 5, with 5 indicating the least amount of color change from the original specimen, and 1 being the most. The wash fastness rating of mordanted samples in terms of color change was rated 2-3 (poor-moderate) regardless of the mordant concentrations. This low wash fastness was a result of the lac dye's sensitivity to the alkaline pH of the soaping solution, which caused some color loss during testing. Therefore, it is suggested to use neutral pH detergent or soap to prevent color fading during home laundering. The level of staining on cotton and wool was good (grade 4).

The colorfastness of the dyed acrylic yarns against rubbing was assessed. In a dry condition, both Fe- and Cu-mordanted samples were rated as good to excellent with a rating of 4-5. Due to the weak fiber alum coordination complex, Al-mordanted samples were given a significantly lower grade. In every case, the wet rubbing fastness qualities were shown to be inferior to the dry condition. This is due to water molecules facilitating in the transfer of unfixed dyes to the rubbing cloth adjacent to them.

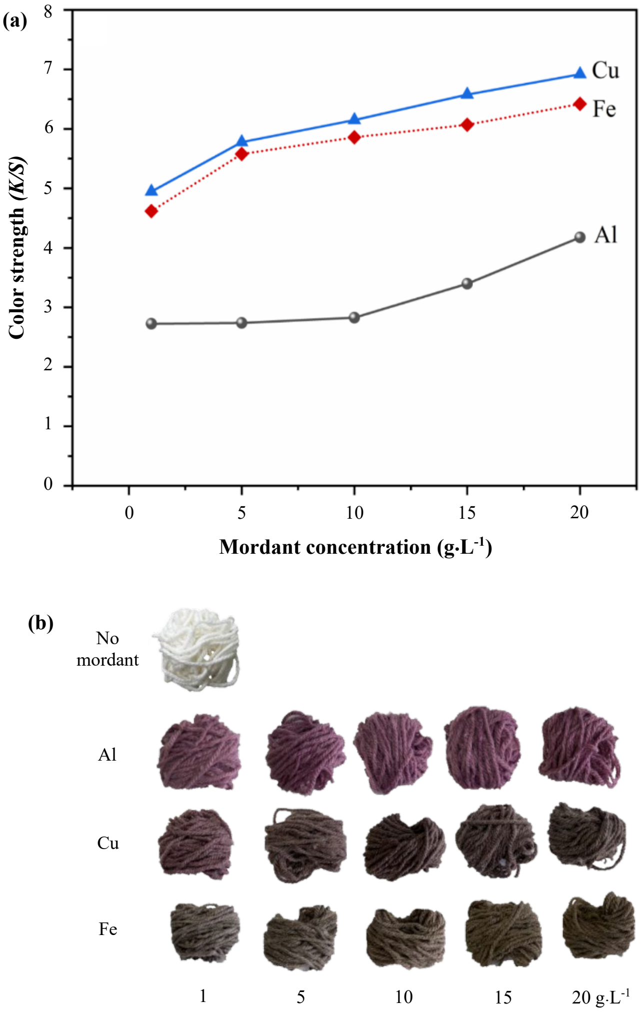


Figure 7. (a) *K/S* and *a** values of mordanted acrylic yarn as a function of metal mordant types and mordant concentrations, and (b) apparent color of dyed samples (20% owf. lac, 100°C, 40 min, pH = 3).

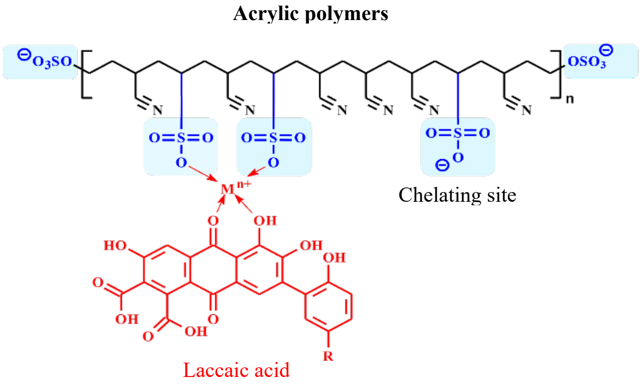
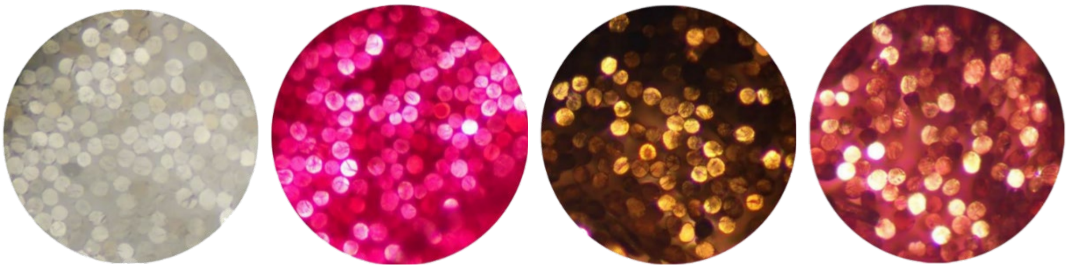


Figure 8. The suggested coordination complex of acrylic, metal, and lac

Color fastness to light was assessed in comparison with the color change of the standard blue wool fabrics, whose ratings are in the range of 1 (very poor) to 8 (outstanding). In comparison to other mordanted samples, the light fastness properties of Al-mordanted acrylic samples were less satisfactory (grade 3; moderate). This is most likely owing to alum's weak chelating complex with natural lac dye molecules [27]. The dyed samples of ferrous sulfate and copper sulfate were more resistant to light fading (grade 4; fairly good). These results demonstrated that iron and copper have the ability to generate stronger bridges between colorants and textile fiber, hence inhibiting photochemical degradation of colorants [27]. Thus, mordants contribute in the promotion of the transfer of loaded energy from the bonded dye molecule, as well as in increasing the light fastness properties of dyed materials. However, various factors influence light fastness, including the chemical structure of natural colorants, the physical state of the colorant within the fiber matrix, the dye concentration, substrate type, other bioactive components in the extracts, and the source and intensity of illuminant.



(a) No mordant **(b)** AlK(So$_4$)$_2$ **(c)** FeSO$_4$ **(d)** CuSO$_4$

Figure 9. Cross-sectional microscopic images of acrylic fibers at 100x magnification (a) no mordant, (b) AlK(SO$_4$)$_2$, (c) FeSO$_4$, and (d) CuSO$_4$ (20% owf. of dye, 20 g·L^{-1} of mordant, pH = 3, 100ºC, 40 min).

Table 2. *CIE* color coordinates of dyed acrylic yarn by varying metal mordant types and mordant concentrations.

Mordants	Concentrations (g·L^{-1})	*L**	*a**	*b**
Without mordant	0	76.53	9.68	-13.36
AlK(SO$_4$)$_2$	1	47.34	16.8	-16.82
	5	47.33	19	-19.52
	10	48.84	19.7	-19.21
	15	44.52	18.4	-18.19
	20	41.27	17.5	-17.41
CuSO$_4$	1	41.6	11.3	-9.24
	5	37.25	6.66	-5.96
	10	37.01	6.79	-5.65
	15	36.54	6.82	-4.52
	20	34.81	4.99	-3.03
FeSO$_4$	1	44.09	6.64	-11.34
	5	41.97	6.34	-9.09
	10	43.56	6.32	-6.23
	15	43.62	5.94	-4.7
	20	44.56	5.81	-3.92

Table 3. Relative unlevelness indices (*RUI*) and visual levelness evaluation.

Mordants	*RUI*	Visual appearance of levelness
AlK(SO$_4$)$_2$	0.34	Good
FeSO$_4$	0.33	Good
CuSO$_4$	0.38	Good

Table 4. Color fastness of acrylic yarn dyed with lac**.**

Samples	Conc. (g·L^{-1})	Wash fastness			Crocking		
		Color change	Staining on cotton	Staining on wool	Dry	Wet	Light
AlK(SO$_4$)$_2$	1	2-3	4	4	4	3-4	3
	20	2-3	4	4	4	3-4	3
FeSO$_4$	1	2-3	4	4	4-5	4	4
	20	2-3	4	4	4-5	4	4
CuSO$_4$	1	2-3	4	4	4-5	4	4
	20	2-3	4	4	4-5	4	4

4. Conclusion

In this study, the dyeing of hydrophobic acrylic yarn with a bio-based natural lac dye was carried out in response to the need for enhanced sustainability in the textile sector. The meta-mordanting method was chosen to dye acrylic yarn. The best dyeing results were achieved using an acidic pH (2-3) and a temperature of 100°C for 40 min of dyeing time. When dyeing synthetic fiber, controlling the temperature in the T_g zone is critical for color uniformity. Two environmentally safe mordants, alum and ferrous sulfate, have been proven to be effective. The type of mordant had a significant impact on the color of the dyed acrylic yarn. The mordanted samples displayed deep and rich color, with good dye penetration and uniformity, as well as good colorfastness.

Acknowledgements

The authors gratefully acknowledge the financial support provided by the Faculty of Science and Technology, Thammasat University, Contract No. SciGR3/2564. We also thank Assoc. Prof. Sukrit Tantrawong (Division of Chemistry, Faculty of Science and Technology, Thammasat University) for his support in cross-sectional analysis. In addition, the authors would like to thank Nattaya Inphahol for her assistance in data collection.

References

[1] P. Pandit, B. Annaldewar, A. Nautiyal, S. Maiti, and K. Singha, "Sustainability in fashion and textile," in *Recycling from Waste in Fashion and Textiles: A Sustainable and Circular Economic Approach*, ed. Wiley-Scrivener, 2020, pp. 177-198.

[2] T. Bechtold, R. Mussak, A. Mahmud-Ali, E. Ganglberger, and S. Geissler, "Extraction of natural dyes for textile dyeing from coloured plant wastes released from the food and beverage industry," *Journal of the Science of Food and Agriculture,* vol. 86, pp. 233-242, 2006.

[3] M. R. Bhuiyan, M. M. Rahman, A. Shaid, and M. A. Khan, "Application of gamma irradiated textile wastewater for the pretreatment of cotton fabric," *Environment and Ecology Research,* vol. 2, pp. 149-152, 2014.

[4] J. Liu, T. Guo, Y. Luo, X. Chai, J. Wu, W. Zhao, P. Jiao, F. Luo, and Q. Lin, "Enhancement of *Monascus* pigment productivity via a simultaneous fermentation process and separation system using immobilized-cell fermentation," *Bioresource Technology,* vol. 272, pp. 552-560, 2019.

[5] C. Gulani, S. Bhattacharya, and A. Das, "Assessment of process parameters influencing the enhanced production of prodigiosin from *Serratia marcescens* and evaluation of its antimicrobial, antioxidant and dyeing potentials," *Malaysian Journal of Microbiology,* vol. 8, pp. 116-122, 2012.

[6] W. Haddar, M. Ben Ticha, N. Meksi, and A. Guesmi, "Application of anthocyanins as natural dye extracted from *Brassica oleracea L. var. capitata f. rubra*: dyeing studies of wool and silk fibres," *Natural Product Research,* vol. 32, pp. 141-148, 2018.

[7] A. Haji, "Improved natural dyeing of cotton by plasma treatment and chitosan coating. Optimization by response surface methodology," *Cellulose Chemistry and Technology,* vol. 51, pp. 975-982, 2017.

[8] T. A. Khattab, M. S. Abdelrahman, and M. Rehan, "Textile dyeing industry: environmental impacts and remediation," *Environmental Science and Pollution Research,* vol. 27, pp. 3803-3818, 2020.

[9] N. S. Bhide, E. D. Pandhare, A. V. Rama Rao, I. N. Shaikh, and R. Srinivasan, "Lac pigments: Part IV. Constitution of laccaic acid B," *Indian Journal of Chemistry,* vol. 7, pp. 987-995, 1969.

[10] E. D. Pandhare, A. V. Rama Rao, and I. N. Shaikh, "Lac pigments: Part III - isolation of laccaic acids A and B and the constitution of laccaic acid A," *Indian Journal of Chemistry*, vol. 7, pp. 977-986, 1969.

[11] E. D. Pandhare, A. V. Rama Rao, R. Srinivasan, and K. Venkataraman, "Lac pigments," *Tetrahedron,* vol. 22, pp. 229-239, 1966.

[12] E. D. Pandhare, A. V. R. Rao, I. N. Shaikh, and K. Venkataraman, "The constitution of laccaic acid B," *Tetrahedron Letters,* vol. 8, pp. 2437-2440, 1967.

[13] H. Oka, Y. Ito, S. Yamada, T. Kagami, J. Hayakawa, K. Harada, K Harada, E. Atsumi, M Suzuki, M. Suzuki, H. Odani, S. Akahori, K. Maeda, T. Tanaka, and M Mizuno, "Identification of lac dye components by electrospray high performance liquid chromatography-tandem mass spectrometry," *Journal of the Mass Spectrometry Society of Japan,* vol. 46, pp. 63-68, 1998.

[14] H. Oka, Y. Ito, S. Yamada, T. Kagami, J. Hayakawa, K. Harada, E. Atsumi, M. Suzuki, M. Suzuki, H. Pdani, S. Akahori, K. Maeda, H. Nakazawa, and Y. Ito, "Separation of lac dye components by high-speed counter-current chromatography," *Journal of Chromatography A,* vol. 813, pp. 71-77, 1998.

[15] C. J. Cooksey, "The red insect dyes: carminic, kermesic and laccaic acids and their derivatives," *Biotechnic and Histochemistry,* vol. 94, pp. 100-107, 2019.

[16] Y. Zhang, Q. Zhou, W. Xia, L. J. Rather, and Q. Li, "Sono-chemical mordanting as a green and effective approach in enhancing cotton bio natural dye affinity through soy surface modification," *Journal of Cleaner Production,* vol. 336, 2022.

[17] M. Shahid, I. Shahid Ul, and F. Mohammad, "Recent advancements in natural dye applications: A review," *Journal of Cleaner Production,* vol. 53, pp. 310-331, 2013.

[18] I. Shahid Ul, M. Shahid, and F. Mohammad, "Perspectives for natural product based agents derived from industrial plants in textile applications - A review," *Journal of Cleaner Production,* vol. 57, pp. 2-18, 2013.

[19] Ö. E. İşmal, "Greener natural dyeing pathway using a by-product of olive oil; prina and biomordants," *Fibers and Polymers,* vol. 18, pp. 773-785, 2017.

[20] P. S. Vankar, "Structure-mordant interaction, replacement by biomordants and enzymes," in *Natural Dyes for Textiles: Sources, Chemistry and Applications*, ed, 2017, pp. 89-102.

[21] Y. Liu, J. Cao, W. Niu, M. Liu, and X. Guo, "Studies on the dyeing and functional properties of modified berberine for a variety of fabrics," *AATCC Journal of Research,* vol. 6, pp. 8-14, 2019.

[22] R. M. El-Shishtawy, G. M. Shokry, N. S. E. Ahmed, and M. M. Kamel, "Dyeing of modified acrylic fibers with curcumin and madder natural dyes," *Fibers and Polymers,* vol. 10, pp. 617-624, 2009.

[23] C. W. Kan, "Colour fading effect of indigo-dyed cotton denim fabric by CO_2 laser," *Fibers and Polymers,* vol. 15, pp. 426-429, 2014.

[24] C. Hawkyard, *Synthetic Fibre Dyeing*: Society of Dyers and Colourists, Bradford, United Kingdom, 2004, 293 p.

[25] M. N. Bukhari, L. J. Rather, M. Shabbir, S. Ul-Islam, U. Singh, M. A. Khan, and F. Mohammad, "Dyeing of wool with anthraquinone based natural colorants from *Cassia fistula* fruit," *Journal of Natural Fibers,* vol. 16, pp. 855-865, 2019.

[26] M. M. Kamel, R. M. El-Shishtawy, B. M. Youssef, and H. Mashaly, "Ultrasonic assisted dyeing. IV. Dyeing of cationised cotton with lac natural dye," *Dyes and Pigments,* vol. 73, pp. 279-284, 2007.

[27] Y. Ding, and H. S. Freeman, "Mordant dye application on cotton: Optimisation and combination with natural dyes," *Coloration Technology,* vol. 133, pp. 369-375, 2017.