p-ISSN 2615-787X e-ISSN 2615-790X
Accredited by Directorate General of Strengthening for Research and Development No: 30/E/KPT/2018

Tropical Animal Science Journal, September 2022, 45(3):368-373
DOI: https://doi.org/10.5398/tasj.2022.45.3.368
Available online at http://journal.ipb.ac.id/index.php/tasj

The Color Fastness and Quality of Eco-Printed Leather with Different Types of Mordant in Natural Dyes from Mangrove Extract (*Rhizophora mucronata*)

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(Received 07-10-2021; Revised 31-01-2022; Accepted 08-02-2022)

ABSTRACT

This study evaluated the color fastness and crust quality of eco-printed leather using various types of mordant in the natural dye of mangrove extract. This study used 20 sheepskin crusts and mangrove bark extract. This experimental study employed a Completely Randomized Design (CRD) with different mordant types as the treatments. The types of mordant used were Aluminum Sulfate ($Al_2(SO_4)_3$), Calcium Carbonate ($CaCO_3$), Citric Acid ($C_6H_8O_7$), and Ferrous Sulfate ($FeSO_4$). The results showed that the use of Ferrous Sulfate ($FeSO_4$) as a mordant gave a darker color and produced better color fastness on eco-printed leather to dry and wet rubbing with a value of 5 (very good), to washing with a value of 4-5 (good), and to sweat with a value of 5 (very good). The use of Ferrous Sulfate ($FeSO_4$) as a mordant improved the tensile strength (p= 0.078), decreased the elongation (p= 0.008), increased the elasticity (p= 0.000), and increased the cracking resistance (p= 0.013) of the eco-printed leather as well as produced more stable stitch-tear strength of the eco-printed leather. It is concluded that using Ferrous Sulfate ($FeSO_4$) as a mordant gives a brownish yellow color, increases color fastness to wet rubbing, dry rubbing, washing, sweating, improves crust qualities especially tensile strength and cracking resistance, and decreases the elongation of eco-printed leather.

Keywords: color fastness; crust quality; eco-print; leather; mordant

INTRODUCTION

Currently, eco-printed leather products with natural dyes are in great demand because they also produce natural shapes of leaves and flowers in addition to transferring color. Characteristically, natural dyes have many advantages, including various colors produced, non-carcinogenic, non-toxic, biodegradable, and safe for life (Thiyagarajan *et al*., 2015).

There have been numerous studies on the use of natural dyes for leather, including studies conducted by Teklay & Kechi (2018) related to *Osyris Quadripartita* extracts and Pervaiz *et al.* (2017) on *Tagetes erecta* L. (Marigold) waste. Singh *et al.* (2020) examined the natural dyeing of leather with *Acacia catechu* extract for the combination of chrome tanned and vegetable tanned sheep crust leather.

The potential of mangrove (*Rhizophora mucronata*) as a natural dye requires deeper studies. Mangroves are useful as a store of carbon stocks. The waste from mangrove trimming has a high economic value that can be used as a natural dye for various media (Anburaj & Jothiprakasam, 2017). Mangrove stem extract is reported to be used as leather tanning (Pancapalaga & Nitiharjo, 2019). The results of the phytochemical test of mangroves were reported by Pringgenies *et al.* (2018) that the pigments found in mangroves that give brownish, yellow, and red colors are tannins, flavonoids, and quinones, respectively.

However, the disadvantage of natural dyes is that they fade quicker and require a mordant in their application. Mordant serves to bind natural dyes to the leather. The benefits of calcium carbonate ($CaCO_3$), citric acid ($C_6H_8O_7$), and ferrous sulfate ($FeSO_4$) as mordant agents provide color variations on the leather (Sofyan *et al.*, 2020). A study related to mordant has been carried out by Kumar Jha *et al.* (2015). The study reported that the two best mordants using marigold are chromium and ferrous sulfate, with optimum mordant concentrations of 3% and 5%, respectively. Different mordants produce different colors. The use of iron sulfate as mordant investigated by Zubairu & Mshelia (2015) found that the post-mordanting technique gave the best result of the color-fast of the onion skin dye. Sofyan *et al.* (2020) describe that CaO as mordant produces a higher color intensity (K/S value of 19.174) and color-fastness to washing (40 °C), resistance to sunlight and heat are categorized into good to very good (4-5). Besides, the mordant material in the manufacture of ecoprint leather is more environmentally friendly.

The use of aluminum sulfate ($Al_2(SO_4)_3$), calcium carbonate ($CaCO_3$), citric acid ($C_6H_8O_7$), and ferrous sulfate ($FeSO_4$) as mordant agents in fabrics has been reported by Sofyan (2020). Still, it has not been widely studied in other media, such as leather, using the eco-print method. Therefore, the objective of the research is to evaluate skin coloring using the eco-print method using natural mangrove dyes from various types of mordant.

MATERIALS AND METHODS

This study used 20 sheepskin crusts (6.8 feet each) from leather artisans in Yogyakarta. In addition, this study involved $Al_2(SO_4)_3$, $CaCO_3$, $C_6H_8O_7$, $FeSO_4$, mangrove leaves, and Natural Dyes from mangrove bark.

The Process of Eco-Printing on Leather

The eco-print leather method used a modified method (Pancapalaga *et al.,* 2021). The crust leathers for eco-printing were soaked in water for 6 hours until they turned bluish. Then the crust leathers were soaked in a mordant solution with a ratio of 1:1 between water and mordant for another 6 hours. The mordant solutions were made by weighing 70 g of each ingredient ($Al_2(SO_4)_3$, $CaCO_3$, $C_6H_8O_7$, and $FeSO_4$) and dissolved in 1 L of water. The mordant solutions were stirred until completely dissolved. After finishing the soaking with mordant, the mangrove leaves were set on the grain layer above the crust or the surface of the eco-printed leather with the tattoo. Basic leather dyeing used a cover cloth. The cover cloth was previously soaked with Natural Dyes from mangrove bark extract *(R. mucronata*). The cloth that had been dipped in natural dyes was used as a medium for covering the crust leathers which had been pasted with mangrove leaves. They were covered with a plastic cover. The leather, cover cloth, and plastic cover were rolled up and tied. They were steamed over low heat to a temperature of 78 °C. After steaming, it was cooled down so the leaves could be easily removed from the leather. After the leaves were removed, the leathers were dried in the room to avoid damage from sunlight. Finally, the eco-printed leather was ready to be tested for quality.

The Extraction of Natural Dyes from Mangrove Bark *(R. mucronata)*

The method of extracting natural dyes from mangrove bark used the method of Subandriyo & Setianingsih (2016). First, mangrove trees with more than 10 cm diameter were peeled. The mangrove bark obtained was air-dried without direct sunlight for one month. Mangrove bark was cut into smaller pieces and ground using a disk mill FFC 15 with a milling speed of 8800 rpm until it became powder. The extraction of Natural Dyes was obtained by Soaking 100 grams of mangrove bark powder with 1 liter of water at 70 °C for 1 hour. The results of this extract are used to color the cover of eco-print leather by soaking the cover in a ZWA solution of mangrove bark for one hour.

Color Value Test of Eco-printed Leather

The color quality analysis of eco-printed leather used Hunterlab Color flex EZ. The reflectance spectrum was based on the percentage of reflectance at every 10 mm wavelength with an interval of 400-700 nm.

Fouier-Transform Infrared Spectrometer (FTIR) Analysis

FTIR test was administered to determine the functional groups of various types of mordant. The FTIR test was carried out using an FTIR spectrophotometer with a TGS detector and attenuated total reflectance (ATR, Pro-One) at an angle of the incident by 45 deg and a scanning speed of 2 mm/second.

Color Fastness Test

Dry and wet rubbing tests were carried out according to SNI-08-0288-1998 (BSN, 1998). Testing color fastness to dry rubbing was carried out using a crock meter. The color fastness test to washing was carried out using the SNI 08-0288-1998 test method (BSN, 1998) and to sweat using the BS 1006:1990 UK LF.

Crust Quality Testing of Eco-Printed Leather

Tensile strength testing was carried out according to SNI 06-1795-1990 with a Strength tester (BSN, 1990b). The tear strength test was carried out based on SNI 06-1794-1990 (BSN, 1990a). The elasticity test was carried out according to SNI ISO 17235: 2015 with an ST 300 Softness tester (BSN, 2015b). The cracking resistance test was carried out according to SNI 3379-2015 with Satra STD Lastometer equipped with the scale of force needed to make the leather crack (BSN, 2015a).

Data Analysis

The research method used was a completely randomized design (CRD). The treatments were P0= Without Mordant, P1= $Al_2(SO_4)_3$, P2= $CaCO_3$, P3= $C_6H_8O_7$, and P4= $FeSO_4$ with 5 replications. Data related to FTIR, color, and color fastness were analyzed descriptively. The color fastness assessment was conducted by comparing the color difference with standard grayscale. The rankings are as follows: value 5 (very good, no change in leather color or discoloration to other materials), value 4 (good, slight color change or staining), value 3 (fair, there was a change or color staining), a value of 2 (moderate, there is a marked change or color staining), and a value of 1 (less, there is a very striking color change and staining). Meanwhile, data on the physical quality of eco-print leather were analyzed using analysis of variance. To find out the difference between treatments, it was continued with Duncan's test.

RESULTS

Eco-Printed Leathers' Color Value

Based on the L*.a*.b* color value test results on eco-printed leather, with the Hunterlab Color flexEZ s/n CFEZ0 530 tool, L*a* and b* color values were obtained. L* values are represented as lighter or darker (lightness–darkness) in the range of 0-100 (black to white). The value of a* is represented as redness for positive and greenness for negative. The value of b* is yellowness for positive and blueness for negative.

Table 1 shows that dyeing with mordant using $FeSO_4$ produced a darker color than the other mordants. The value of L* in $FeSO_4$, which was 34.40, was the lowest value compared to the other mordants. In contrast, the brightest staining was mordant using $Al_2(SO_4)_3$ since it had a value of 76.16.

Table 1 depicts that all staining results, whether using mordant or not, produced a positive b value (+b*), which indicates that the results of the eco-print leather staining were brownish-yellow. This can be seen in the highest positive b* value, 46.23 obtained from staining with mordant using $C_6H_8O_7$. The lowest was 8.63 obtained from staining with mordant using $FeSO_4$.

Table 1. The color value of eco-printed leather with different types of mordant

No.	Treatments	Color value		
		L *	a*	b*
1	Without mordant	58.25±0.36	11.66±0.74	39.49±0.14
2	$Al_2(SO_4)_3$	76.16±0.81	2.80±0.26	34.42±0.17
3	$CaCO_3$	62.64±0.52	2.79±0.15	29.69±0.18
4	$C_6H_8O_7$	60.48±1.55	15.54±0.18	46.23±1.09
5	$FeSO_4$	34.40±1.71	2.08±0.11	8.63± 0.09

Note: L*= lighter or darker (lightness–darkness) in the range of 0-100 (black to white); a*= redness for positive and greenness for negative; b*= yellowness for positive and blueness for negative.

Reflectance Value of Eco-Printed Leather

Table 2 shows that the reflectance value can be seen with a 400-700 nm wavelength. The highest value was at a wavelength of 700 nm, and the highest reflectance value was obtained in mordant using $Al_2(SO_4)_3$ by 78.47%. It shows that eco-print produced on mordant using $Al_2(SO_4)_3$ is lighter. On the contrary, at the same wavelength, the lowest value was obtained on mordant $FeSO_4$ by 11.92%, which means the color produced is darker and not much-reflected light.

The Analysis of FTIR of Eco-Printed Leather

Table 3 presents the results from the FTIR spectrophotometer to determine the functional groups contained in the eco-printed leather of various mordants. Identifying samples using an FTIR spectrophotometer obtained several identical wave number spectra between the leathers that were given mordant and without mordant.

Identical sample wave numbers include the wavenumber 3302.50 cm^{-1} indicating an OH asymmetry range, 2923.66 cm^{-1} indicating the CH sp3 range, 1548.56 cm^{-1} indicating aromatic ring range, and 1630.52 cm^{-1} indicating a secondary alcohol C-O group.

Color Fastness Value of Eco-Printed Leather

Table 4 shows that the dry rubbing fastness test on staining without mordant has a value of 3 (medium). After being given mordant $CaCO_3$ and $C_6H_8O_7$ to dry rubbing fastness, the value increased to 3-4 (sufficient), and dry rubbing fastness increased even more with mordant using $Al_2(SO_4)_3$ increased to 4-5 (good). The best value for using mordant $FeSO_4$ was 5 (very good).

The value to wet rubbing fastness of various mordant ranges from 3 (sufficient) to 5 (very good). The highest value was the use of mordant using $FeSO_4$, and subsequently, below was $Al_2(SO_4)_3$, $C_6H_8O_7$, and $CaCO_3$.

Table 2. The percentage of reflectance value at a wavelength of every 100 mm with an interval of 400-700 mm on different types of mordant

Wavelength (nm)	Reflectance value (%)				
	Without mordant	$Al_2(SO_4)_3$	$CaCO_3$	$C_6H_8O_7$	$FeSO_4$
400	4.41±0.06	11.23±0.09	6.96±0.08	4.50±0.06	4.35±0.08
500	15.45±0.13	40.49±0.11	28.94±0.15	14.11±0.17	7.32±0.25
600	36.47±0.11	59.58±1.41	33.57±1.05	42.39±0.22	9.17±0.21
700	56.71±0.15	78.47±1.18	55.32±1.15	56.27±0.65	11.92±0.19

Table 3. Qualitative data analysis of FTIR spectrum on different types of mordant

Functional groups	Wavenumber cm^{-1}				
	Without mordant	$Al_2(SO_4)_3$	$CaCO_3$	$C_6H_8O_7$	$FeSO_4$
O-H (Hydroxyl)	3302.50±0.09	3302.50±0.21	3288.04±0.02	3302.50±0.12	3302.50±0.14
C-H (Aromatic)	1548.56±0.19	1549.52±0.13	1546.63±0.15	1549.52±0.81	1548.56±0.23
C-O (Carbonyl)	1630.52±0.08	1630.52±0.05	1630.52±0.10	1630.52±0.46	1631.48±0.45
Csp3-H	2920.66±0.82	2920.66±0.06	2920.66±0.12	2920.66±0.23	2920.66±0.52

Table 4. Color fastness on different types of mordant

Variables	Color fastness value					SNI 06-0996-1989
	Without mordant	$Al_2(SO_4)_3$	$CaCO_3$	$C_6H_8O_7$	$FeSO_4$	
Dry rubbing	3.2±0.46	4.2±0.64	3.4±0.54	3.2±0.48	5.0±0.44	3.0
Wet rubbing	3.0±0.21	4.0±0.24	3.4±0.28	3.4±0.36	5.0±0.22	3.0
Washing	3.4±0.48	4.0±0.50	3.4±0.26	3.2±0.44	4.5±0.26	3.0
Sweating	3.0±0.22	4.5±0.18	3.4±0.23	3.4±0.34	5.0±0.42	3.0

Table 5. Crust quality on different types of mordant

Variables	Value of crust quality					Sig.
	Without mordant	$Al_2(SO_4)_3$	$CaCO_3$	$C_6H_8O_7$	$FeSO_4$	
Tensile strength (N/cm^2)	1162.98±1.23^a	1160.58±1.25^a	1620.33±1.42 ^{ab}	1346.54±1.26^{ab}	2155.08±1.23^c	p= 0.078
Elasticity (%)	93.11±0.24^c	66.78±0.23^{ab}	77.29± 0.35^{bc}	51.06±0.32^{ab}	45.62±0.43^a	p= 0.008
Tear resistance (N/cm)	195.80±1.23^a	153.53±1.38^a	159.15±1.35^a	202.24±1.32^a	273.35±1.42^a	p= 0.375
Elongation (mm)	4.7±0.54 ^{b}	4.6±0.65 ^{b}	2.4±0.22 ^{a}	4.6±0.41 ^{b}	4.9±0.43 ^{b}	p= 0.000
Cracking resistance (mm)	9.4000±0.54^b	9.6225±0.52^b	6.7650±0.48^a	9.5900±0.52^b	10.0450±0.55^b	p= 0.013

Notes: Means in the same column with different superscripts differ significantly ($p<0.05$).

The lowest was without mordant with a value of 3 (sufficient). The color fastness value to washing of various mordants showed different results. The best was $FeSO_4$ d with a value of 4-5 (good) as well as the best sweat resistance in $FeSO_4$ with a value of 5 (very good). Using mordant $Al_2(SO_4)_3$, $C_6H_8O_7$, $CaCO_3$, and FeSO4 with natural dyes of mangrove extract on eco-printed leather increased the value of color fastness that complied with SNI 06-0996-1989 (BSN, 1989).

The Value of Crust Quality of Eco-Printed leather

Table 5 describes the results of the crust quality of eco-printed leather with the application of various types of mordant. The highest tensile strength of eco-printed leather was obtained by mordant using $FeSO_4$. In contrast, the lowest elongation of eco-printed leather was obtained at $FeSO_4$ that eco-printed leather with mordant using $FeSO_4$ caused the leather not to stretch easily. However, the stitch strength e was almost the same among various mordant with P-value= 0.260. Eco-print leather with mordant using $CaCO_3$ results in stiffer, easier cracking leather than the other mordants.

DISCUSSION

Color Value of Eco-Printed Leather

The complete details of the test results regarding color differences of eco-printed leather with natural mangrove dyeing using various types of mordant are shown in Table 1. The treatments of using various types of mordant on eco-printed leather that has been stained will affect the color brightness. However, in this study, the value of L* (brightness) of all types of mordant ($Al_2(SO_4)_3$, $C_6H_8O_7$, $CaCO_3$, and $FeSO_4$) showed positive results that the resulting color led to light or bright color.

The most visible color change of the four mordants was the use of $FeSO_4$ with a slightly darker color. This was due to the reaction between tannins and Fe^{2+} producing complex salts with black color during dyeing with the mordant. The more concentration of mordant used, the more the dye absorbed by the leather fiber so that the color obtained is sharper. Adsorption that occurs with the addition of mordant is classified as chemical adsorption, which has relatively stronger bonding properties than physical adsorption (Rekaby *et al.*, 2009).

Similar results show that mordant using FeSO4 gives a dark yellow color (Kanchana *et al.*, 2013), a light brownish color (Belemkar & Ramachandran, 2015). On the other hand, mordant using acetic acid shows a bright yellow color.

Reflectance Value of Eco-printed Leather

Reflectance is the light from light energy that is reflected from a surface against the light that hits it or falls on a plane. High reflectance shows higher reflected light. The distribution comparison of the reflectance values of the eco-print leather shows the same pattern at the visible light with a wavelength of 400-700 nm, which is brownish yellow. The result shows the highest reflectance value of mordant using $Al_2(SO_4)_3$. This means that using $Al_2(SO_4)_3$ as a mordant in the natural dyeing of mangroves makes the color of eco-printed leather lighter. This is because Al^{3+} can bind the tannins contained in the natural dyeing of mangrove extract with an acidic pH.

According to Jordeva *et al.* (2020), mordant using $Al_2(SO_4)_3$ will form a coordination complex that has an affinity between leather fibers and natural dye pigment. Aluminum salts ($Al_2(SO_4)_3$) attached to the surface of the leather will bind the organic dye/pigment in the natural dye to bind to the fiber and create a link between the dye molecules and the fiber by forming a coordination complex. It is also said that the reflectance of each object is generally unique (special or distinctive that characterizes the object) so that it has a different value with its respective characteristics.

FTIR Analysis of Eco-Printed Leather

The results of the FTIR analysis of eco-printed leather obtained the similarity of functional groups with the peak of the wavenumber spectrum among treatments. This was due to the functional groups of various types of mordant bound to the tannins contained in the mangrove dye. This is indicated by the results of the FTIR analysis that these specific peaks are of tannin compounds. Thus, it strengthens the assumption that various types of mordant used are bound to tannin compounds.

According to Verenkar & Krishnan (2017), tannin compounds are at a wavenumber of 3424.96 cm^{-1} for the OH asymmetry range, 2923.56 cm^{-1} for the CH sp^3 range, and 1635.34 cm^{-1} for the aromatic ring range. Furthermore, it is said that functional groups such as hydroxyl or carbonyl, nitroso contained in natural dyes cause these dyes to bind aluminum ions in the mordant (Indrianingsih & Darsih, 2013).

Color Fastness of Eco-Printed Leather

Table 4 shows that the best mordant used in natural dyeing of mangroves to produce color fastness to wet rubbing, dry rubbing, washing, and sweating on eco-print leather are $FeSO_4$ and $Al_2(SO_4)_3$. This is because of the ions Fe^{2+} and Al^{3+} bind to pigments. The tannins contained in the mangrove dyes form complex salts that cause the dyes to stay in the fiber. Thus, the color fastness increases that it does not fade easily.

The string resistance of eco-printed leather to washing after being given mordant using $FeSO_4$ was due to the molecular differences between $FeSO_4$ and Mangrove bark extract as natural dyes. In contrast, the resistance to washing is largely determined by the molecular weight or molecular size. Larger molecules have better washing resistance. This is reinforced by Uddin & Hossain (2010) that acid dyes have good washing and light resistance. The resistance properties are strongly influenced by the molecular weight and configuration.

On the other hand, the leather is more receptive to mordant. This is due to its amphoteric nature, and the leather can absorb acids and bases evenly and effectively when the leather is given a mordant using aluminum salt. It hydrolyzes the salt into its acidic and basic components. The basic component is adsorbed on the –COOH group, and the acidic component is removed during washing. Like wool, silk is also amphoteric and can absorb acids and bases (Prabhu & Bhute, 2012).

Furthermore, it is said that the leather's ability to absorb dye is probably due to the –COOH group in the leather fiber, which is capable of forming covalent bonds. Meanwhile, in the mordant process, the hydrogen element in the hydroxyl group of the natural dye (electron donor) can be replaced with a metal element (acceptor). The bond is a carbonate bond (semi-polar) through one or more ion pairs of electrons given by the donor compound to the acceptor with an empty path (Saravana *et al.,* 2014).

The more concentration of mordant used, the greater the dye absorbed by the leather fiber so that the color obtained is sharper due to the effect of adding mordant. Adsorption that occurs with the addition of mordant is classified as chemical adsorption, which has relatively stronger bonding properties than physical adsorption.

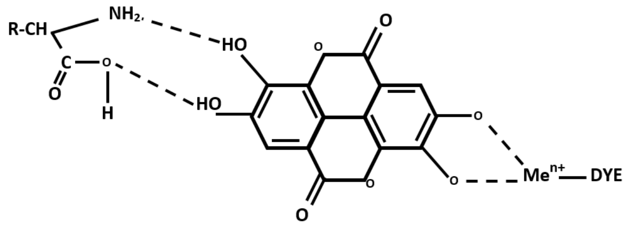

Figure 1. Mechanisms of leather proteins, tannins, mordant, and natural dyes (Prabhu & Bhute, 2012).

Crust Quality of Eco-Printed Leather

The use of mordant of $FeSO_4$ in the natural dyeing of mangroves can increase the tensile strength, elasticity, and cracking resistance as well as reduce the elongation of the eco-printed leather. This is because the mordanting process releases aluminum Fe^{2+} and forms covalent bonds with leather proteins, tannin pigments in natural mangrove dyes, with covalent bonds making the leather stronger. The covalent bonds of leather proteins, mordant, and dyes can be seen in Figure 1.

Figure 1 shows that the mangrove dye contains anion groups that will have ionic bonding with the mordant of Fe^{2+} group and cationic amino acid groups from leather proteins. The main mechanism of dyeing with mangrove and mordant of FeSO4 is the formation of salt bonds between amino groups in leather proteins. With the addition of hydrogen ions from mangroves, free ammonium ions are formed, which are positively charged so that the leather increases the amount of cationic charge (+). In the end, the leather can bind the anions of the acid and mordant dyes (Prabhu & Bhute, 2012).

CONCLUSION

The use of $FeSO_4$ mordant for natural coloring of mangrove extract on leather with the eco-print method produces the best color quality compared to the other types of mordant, such as aluminum sulfate ($Al2(SO_4)_3$), lime ($CaCO_3$), and citric acid ($C_6H_8O_7$), especially on fastness color. This is due to wet rubbing, dry rubbing, washing, sweating, and improving physical qualities, especially on tensile strength, ductility, and crack resistance, and reducing the elongation of eco-print leather.

CONFLICT OF INTEREST

No potential conflict of interest stated by the author.

ACKNOWLEDGMENT

Acknowledgments are addressed to the Ministry of Education and Culture, Research and Technology that has funded the Higher Education Applied Research

(Program Penelitian Terapan Unggulan Perguruan Tinggi/PTUPT) with contract number 313/E4.1/AK.04. PT/2021.

REFERENCES

Anburaj, R. & V. Jothiprakasam. 2017. Bioassay of tannin rich fraction and identification of compounds using UV-Vis, FTIR and RP-HPLC. Res. J. Pharm. Biol. Chem. Sci. 8: 432-445.

Belemkar, S. & M. Ramachandran. 2015. Recent trends in Indian textile industry-exploring novel natural dye products and resources. International Journal on Textile Engineering and Processes 1: 33–41.

BSN. 1989. Cara Uji Ketahanan Gosok Cat Tutup untuk Kulit jadi dengan Alat Crok Meter (SNI 06-0996-1989). Badan Standarisasi Nasional.

BSN. 1990a. Cara Uji Kekuatan Sobek dan Kekuatan Sobek Lapisan Kulit (SNI 06-1794-1990). Badan Standarisasi Nasional.

BSN. 1990b. Cara Uji Kekuatan Tarik dan Kemuluran Kulit (SNI 06-1795-1990). Badan Standarisasi Nasional.

BSN. 1998. Kulit Sapi Samak Kombinasi (Nabati-Krom/sintesis) yang Dicat Tutup (SNI 06-4900-1998). Badan Standarisasi Nasional.

BSN. 2015a. BSN. 2015. Kulit - Penentuan Penggelembungan dan Kekuatan Permukaan (Metode Bola Meletup) (ISO 3379:2015). Badan Standarisasi Nasional.

BSN. 2015b. Kulit - Metode Uji Fisis dan Mekanis-Penentuan Kelemasan (ISO 17235:2015). Badan Standarisasi Nasional.

Indrianingsih, A. W. & C. Darsih. 2013. Natural dyes from plants extract and its applications in indonesian textile small medium scale enterprise. Jurnal Eksergi 11:16–22. https://doi.org/10.31315/e.v11i1.327

Jordeva, S., M. Kertakova, S. Zhezhova, S. G. Longurova, & K. Mojsov. 2020. Dyeing of textiles with natural dyes. Tekstilna Industrija 4:12–21. https://doi.org/10.5937/tekstind2004012J

Kanchana, R., A. Fernandes, B. Bhat, S. Budkule, S. Dessai, & R. Mohan. 2013. Dyeing of textiles with natural dyes - an eco-friendly approach. Int. J. Chemtech. Res. 5:2102–2109.

Kumar Jha, C., R. Kumar, S. V. Kumar, & V. D. Rajeswari. 2015. Extraction of natural dye from marigold flower (*Tagetes erecta* L) and dyeing of fabric and yarns : A focus on colorimetric analysis and fastness properties extraction of natural dye from marigold flower (*Tagetes erecta* L.) and dyeing of fabric and yarns. Pharm. Lett. 7:185–195.

Pancapalaga, W., Suyatno, & A. Malik. 2021. Crust leather quality with eco-printing dyeing method. Indian J. Sci. Technol. 14:71–75. https://doi.org/10.17485/IJST/v14i1.1982

Pancapalaga, W. & S. Nitiharjo. 2019. The quality of rabbit tanned by mangrove (*Rhizopora mucronata*). Leather and Footwear Journal 19:189–194. https://doi.org/10.24264/lfj.19.4.3

Pervaiz, S., T. A. Mughal, F. Z. Khan, S. Hayat, A. Aslam, & S. F. Shah. 2017. Environmental friendly leather dyeing using *Tagetes erecta* L. (Marigold) waste flowers. Int. J. Biosci. 10:382–390. https://doi.org/10.12692/ijb/10.1.382-390

Prabhu, K. H. & A. S. Bhute. 2012. Plant based natural dyes and mordnats : A Review. Journal of Natural Product and Plant Resourse. 2:649–664.

Pringgenies, D., E. Yudiati, R. A. T. Nuraini, E. S. Susilo, & E. Rahayuningsih. 2018. Optimal concentration of mangrove (*Rhizophora mucronata*) leaf and propagule based natural dye. Malaysian Journal of Fundamental and Applied Sciences 168–173. https://doi.org/10.11113/mjfas.v14n1-2.1011

Rekaby, M., A. A. Salem, & S. H. Nassar. 2009. Eco-friendly printing of natural fabrics using natural dyes from alkanet and rhubarb. Journal of the Textile Institute. 100:486–495. https://doi.org/10.1080/00405000801962177

Saravana, P., G. Chandramohan, J. Mariajancyrani, & Kiruthikajothi. 2014. Eco-friendly dyeing of cotton fabric with a natural dye extracted from flowers of *Lantana camara* linn. Int. J. Bioassays 3:1653–1656.

Singh, P., D. Sharma, & S. Chatterjee. 2020. Application of *Acacia catechu* natural dye on banana fiber. Ergon. Int. J. 4:000229. https://doi.org/10.23880/EOIJ-16000229

Sofyan, S., F. Failisnur, S. Y. Silfia, & A. Ardinal. 2020. Natural liquid dyestuff from wastewater of gambier processing (*Uncaria gambir* Roxb.) as textile dye. Int. J. Adv. Sci. Eng.. Inf. Technol. 10:2512–2517. https://doi.org/10.18517/ijaseit.10.6.12891

Subandriyo & N. I. Setianingsih. 2016. Extraction process for reducing tannin of Mangrove fruit as a raw material for food flour. Aquatic Procedia 7:231–235. https://doi.org/10.1016/j.aqpro.2016.07.032

Teklay, N., & A. Kechi. 2018. Optimization of leather dyeing using natural dye extract. Int. J. Eng. Sci Res. 6:136–141.

Thiyagarajan, S., K. Balakrishnan, & S. Tamilarasi. 2015. A study of extraction and dyeing behavior of natural dye obtained from cotton a study. IOSR Journal of Applied Chemistry 8:85–89.

Uddin, K. & S. Hossain. 2010. A comparative study on silk dyeing with acid dye and reactive dye. Int. J. Eng. Technol. 10:1–6.

Verenkar, N. & S. Krishnan. 2017. Dyeing of cotton and silk with eco-friendly dyes extracted from bark of mangrove species Rhizophora mucronata and *Ceriops tagal*. Int. J. Chemtech. Res. 10:102–110.

Zubairu, A. & Y. M. Mshelia. 2015. Effects of selected mordants on the application of natural dye from onion skin (*Allium cepa*). Science and Technology 5:26–32.