

Dyeing of Shoe Upper Leather with Extracted Dye from *Acacia Nilotica* Plant Bark-An Eco-Friendly Initiative

M. M. Mahdi¹, F. T. Zohra², S. Ahmed*¹

¹ Department of Leather Engineering, Institute of Leather Engineering and Technology, University of Dhaka, P.O. Box: 1209, Dhaka, Bangladesh

² Department of Leather Products Engineering, Institute of Leather Engineering and Technology, University of Dhaka, P.O. Box: 1209, Dhaka, Bangladesh

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ABSTRACT

In recent years, the application of non-toxic and eco-friendly natural dyes for textile and leather coloration is getting interest among manufacturers and buyers due to the hazardous impact of conventional azo dyes. Natural dyes have extensive use in textiles, yet their application to leather is very limited. In this research, the prospect of using dye extracted from Acacia nilotica bark for dyeing of shoe upper leather was examined. Chrome-tanned goatskins were employed as raw material for dyeing with the extracted dye using drum dyeing method. The dyeing process was conducted with and without mordant, following pre-mordanting and post-mordanting techniques. Color analysis, various colorfastness tests, and determination of dye exhaustion rate were conducted to evaluate the dyeing quality. It was observed that the experimental dye imparted amber color to the leather that turned brown and gray upon mordanting with potash alum and ferrous sulfate, respectively. The dyed leather samples showed excellent colorfastness but moderate heat and light fastnesses. Mordanted dyed samples, especially the pre-mordanted dyed samples, possessed a better color intensity and more dye exhaustion rate. The leather samples dyed with extracted dye also increased the strength property of the leather and various color shades were produced by mordanting techniques. Prog. Color Colorants Coat. 14 (2021), 241-258© Institute for Color Science and Technology.

1. Introduction

The leather industry has considered as a traditional manufacturing industry since the beginning of human civilization and still recognized as a potential export-earning source for many countries [1-3]. During leather production, animal hides/skins undergo several chemical and mechanical operations to achieve its final features according to buyers' requirement [4, 5]. Dyeing is an important process in leather making and the quality of dyeing is considered as the first leather assessment criterion of buyers in the most cases [6]. It is the process

of imparting color into the leather fiber (collagen) with various coloring agents (dyes) to make the product appealing and artistic to the user [7]. These dyes are synthesized from non-renewable petroleum sources (tar), mainly azoic in nature, known as 'synthetic dyes' [8-10]. Despite having several benefits, these dyes also have some harmful effects on environment and human health [11]. Dye-loaded tannery effluent discharges an ample amount of suspended solids and liquids with high degree of biochemical and chemical oxygen demands that decreases dissolved oxygen (DO) in surface water and

*Corresponding author: soburahmed@du.ac.bd
soburahmed2001@yahoo.com

hamper sunlight penetration and photosynthesis rate in aquatic plants [12-14]. Moreover, some of the dyes degrade into banned amines in contact with human sweat (high pH) producing toxic substances for their xenobiotic nature and complex aromatic structure, which results in allergy, skin irritation, and carcinogenic symptoms in many cases [15-18]. Therefore, it has become essential to search for non-toxic eco-friendly dyes to reduce the negative impacts of synthetic dyes on human health and the environment.

The colorants derived from plant/animal sources produce a very uncommon, soothing, and softer shade on the dyed substrate as compared to synthetic dyes [19]. These coloring components are known as 'natural dyes' which have recently been acquiring the attention of manufacturers and buyers [20-22]. Natural dyes are renewable, non-toxic, and undeniably exhibit better biodegradability and compatibility with the environment. Moreover, they possess a number of special characteristics like deodorizing, flame retardant, resistance to ultraviolet radiation, antibacterial potency, etc. yet having a few demerits like less yield, complex dyeing techniques, and inadequate colorfastness [23, 24]. However, the adaptation of modern extraction technologies like ultrasonic, enzyme assisted extraction, etc., optimized extraction process, dyeing recipe, advanced dyeing machinery i.e. sonication, gamma radiation, etc., and associated materials have overcome those drawbacks and started to create the market potential of natural dyes [25-27]. Hence, the use of natural dyes can become a suitable substitute for the conventional synthetic dyes in leather dyeing.

In this research, the possible application of *Acacia nilotica* dye in dyeing of leather was investigated. *Acacia nilotica* is a leguminous and shrub genus plant of Mimosoideae subfamily under the *Fabaceae/Leguminosae* family [28]. It is globally well known for

its executed gum named 'Gum Arabic', which is widely applied in traditional agro-forestry systems and pharmacology [29-31]. It is an indigenous plant at Rajshahi, Pabna, Kushtia, Jashore, Bogura, and Faridpur district in Bangladesh and locally known as Babul (English)/Babla/Kata Babla/Kikar (Bengali) [32]. The plant bark is a by-product of timber farm which is enriched with tannin and coloring components [33, 34]. Some of the plant parts are used as antibacterial agents, antioxidants, medicines of various diseases of human and animal, vegetable tanning agent and bio-adsorbents to remove heavy metals and colors from industrial effluents [35-56].

The extracted dye from *Acacia nilotica* bark is brown in color (CI Natural Brown 3), and the coloring components of this dye are acacetin, quercetin, and ellagitannins [57, 58]. Among these coloring agents, acacetin and quercetin belong to the flavonol class and known as 'flavonoids' [59-61]. In addition, ellagitannin is a well-known hydrolysable tannin substance [62]. Therefore, the dye is listed as a 'flavonoid dye' or 'tannin-based dye' in the literature of natural dye's chemical classification [11, 56, 62, 63]. In recent years, the dye is extensively used in dyeing of natural substrates like paper, cotton fabric, wool, silk, and manmade fibrous materials like nylon, polyester, etc. apart from dyeing of leather; as very limited studies are found [64-72].

The main objective of this research is to extract dyes from *A. nilotica* bark to be used in dyeing of leather. In specific, we focused on (i) studying the physical characteristics of the extracted dye, (ii) studying the effect of mordant and mordanting techniques on dyeing quality and colorimetric properties of the dyed substrates, (iii) studying the interaction of dye molecule with collagen fiber.

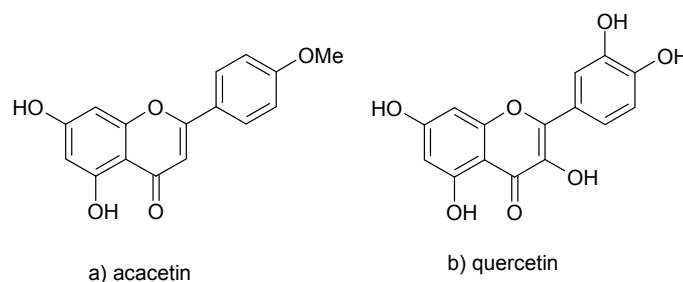


Figure 1: Coloring components of *Acacia nilotica* natural dye.

2. Experimental

2.1. Materials and chemicals

Sufficient amount of *Acacia nilotica* barks were collected in a plastic bag from a local herbal shop at Rangpur, Bangladesh. They were thoroughly washed with tap water to remove dirt and surface adhered coarse matters, and then sundried for a week. Dried barks were powdered by a grinder and sieved in particle size ranged 500-750 μm . Bark powder was stored in an airtight polybag in dark at room temperature until dye extraction.

Auxiliary leather processing chemicals for post-tanning including, wetting agents, neutralizing agents, re-tanning agents, and fat-liquors were purchased from Hazaribagh market at Dhaka. Charcoal powder, potash alum [$\text{KAl}(\text{SO}_4)_2 \cdot 12\text{H}_2\text{O}$], ferrous sulfate (FeSO_4), standard sodium carbonate (Na_2CO_3) and acetic acid (CH_3COOH) solutions were obtained from Merck, India.

2.2. Dye extraction

The dye was extracted from bark powder by aqueous extraction method following the work of Patel (2013) [73] under optimum condition [63]. The bark powder was soaked into distilled water (MLR 1:20) in a 1000 mL beaker for overnight. The next morning, the mixture was boiled at 100 °C for 2 h on a hotplate (Jeo Tech TM-14SB) while stirring with a magnetic stirrer. The mixture was cooled at room temperature and filtered with a fine fabric obtaining a dark brown color solution. The filtrate was further boiled with charcoal powder for an hour and filtered by Whatman No. 41 filter paper after cooling. The pH of the extract solution (6.75) was measured with a digital pH meter (Hanna instruments). The physical properties of the obtained dye were assessed through different tests following standard methods [74-80].

2.3. Leather dyeing

Shaved chrome tanned goat leather (thickness: 0.9-1.0 mm, weight: 0.8-0.9 kg) were dyed with the extracted dye to make the shoe upper leather. Globally around 70% of the produced leather is used for shoe making [81]. The leather samples were dyed with and without mordant following drum dyeing method. Potash alum and ferrous sulfate were applied as metallic mordant in the dyeing process of leather. In the pre-mordanting and post-mordanting processes, the concentration of the mordant was 10% (o.w.l.) in the

case of potash alum and 5% (o.w.l.) in the case of ferrous sulfate. In pre-mordanting step, the leather samples were treated with the mordant solution at 50 °C for 60 min., then washed thoroughly with tap water to the remove surplus mordant, and finally placed into the dye bath. However, in post-mordanting technique the leather samples were treated with mordant after dyeing. The dyeing process for both the unmordanted (control) and mordanted leather samples was identical, as shown in Table 1.

2.4. Colorimetric assay

The dyed leather samples were subjected to X-Rite color tester (CI-7) under illuminant D65 light at 10° observation angle to measure the color coordinates in terms of CIELab and CIELch (L^* , a^* , b^* , c^* , and h°) system in visible spectrum region (400-700 nm). The chroma (c^*) and hue angle (h°) of the dyed samples was measured by the following equations (Eq. 1 and 2) [82]:

$$\text{Chroma } (c^*) = \sqrt{a^2 + b^2} \quad (1)$$

$$\text{Hue angle } (h^\circ) = \tan^{-1}\left(\frac{b}{a}\right) \quad (2)$$

where, L^* = lightness/darkness, a^* = redness/greenness and b^* = yellowness/blueness.

2.5. Dye exhaustion

Exhaustion is expressed as the process through which dye molecules are transferred to the substrate from the dye bath [83]. The following equation was used to calculate the dye exhaustion rate (Eq. 3) [84]:

$$\% \text{ Exhaustion} = \left(\frac{C_g - C_t}{C_g}\right) \times 100 \quad (3)$$

where, C_g is the initial concentration of the dye bath, C_t is the dye concentration in spent dye-liquor. The dye concentration of both samples was measured by a UV-Visible spectrophotometer (UV160U, Shimadzu) at 340 nm. According to Rather et al. (2015) [58], the coloring components of *Acacia nilotica* dye show two bands in the emission spectrum at 340 nm excitation and the peaks are located at 450 nm and 553 nm. Therefore, the wavelength of the highest absorbance (λ_{max}) was considered 340 nm to draw the calibration curve for *A. nilotica* dye, to determine the dye concentration of both dye baths and the spent dye-liquor.

Table 1: Post-tanning and dyeing processes for shoe upper leather manufacturing.

| Process | Chemicals | Amount % (o.w.l)* | Duration (minutes) | Remarks |
|----------------|------------------------|-------------------|--------------------|---------------------------------------------------------------------------------|
| Acid wash | Water | 200 | 30 | pH: 3.0-3.2; Drain & rinse |
| | Wetting agent | 0.3 | | |
| | Oxalic acid | 0.5 | | |
| Re-chroming | Water | 150 | 15 | pH: 2.8-2.9 |
| | Formic acid | 0.3 | 60 | |
| | Basic chromium sulfate | 6 | | |
| | Chrome syntan | 3 | | |
| | Sodium formate | 1 | | |
| | Chrome stable fat | 0.8 | | |
| Basification | Sodium bicarbonate | 1 | 90 | pH: 3.8-4.0; Drain, wash & leave overnight |
| Neutralization | Water | 150 | 30 | pH: 4.5-4.6 |
| | Neutralizing syntan | 2 | | |
| | Sodium formate | 1 | | |
| | Sodium bicarbonate | 0.5 | 30 | pH: 4.8-5.0 Drain & wash |
| Re-tanning | Water at 45°C | 150 | 20 | Drain & wash |
| | Acrylic resin syntan | 3 | 30 | |
| | Melamine resin syntan | 2 | | |
| | Phenolic syntan | 3 | | |
| | Replacement syntan | 3 | | |
| | Mimosa | 4 | | |
| | Quebracho | 2 | | |
| | Formic acid | 0.5 | 30 | |
| Dyeing | Water at 50°C | 100 | 20 | Check penetration by cross-section test; Drain & wash |
| | Dye leveler | 2 | 50 | |
| | Dye | 6 | | |
| | Formic acid | 1.5 | 30 | |
| Fat-liquoring | Water at 55°C | 100 | 60 | Check the bath, Darin and Wash; leave overnight; continue to drying and pile up |
| | Synthetic fat | 4 | | |
| | Semi-synthetic fat | 2 | | |
| | Sulfated oil | 3 | | |
| | Raw oil | 0.5 | | |
| | Formic acid | 1.5 | 30 | |

*o.w.l. =on weight of shaved leather

2.6. Quality assessment

2.6.1. Sampling and conditioning

The test specimens were cut from the sampling location with a stainless sharp knife to assess their dyeing quality.

Sampling location was determined by following IUP 2 norm [85]. After cutting, the test specimens were conditioned at (20±1) °C, (65±3) % relative humidity for 48h and further proceeded to carry out colorfastness tests

using calibrated apparatuses according to standard operating procedure (SOP) [86].

2.6.2. Colorfastness tests

Different colorfastness tests (viz. light fastness, heat fastness, perspiration fastness, wash fastness and color rub fastness (both wet and dry)) were performed to examine the quality of the dyed samples following standard test methods [87-91]. Three experts put their ratings in terms of grayscale after the assessment of color change in leather samples [92].

2.6.3. Evaluation of the leather strength

The strength of the dyed leather sample was evaluated by conducting tensile strength following IUP-6 norm [93] and stitch tearing strength according to DIN-53331 method [94]. SATRA tensile strength tester and a thickness gauge (with a dead load of 1kg) were used to carry out these tests.

3. Results and Discussion

3.1. Dye extraction mechanism

The extraction of dyestuff from *Acacia nilotica* plant bark was conducted in aqueous media where the dye

was formed in three steps. In the first step, dye molecules dispersed into the solvent media (water) by loosening their cell structure after soaking with water for a long time (usually overnight) and then boiled to get the dye solution [11]. Removal of non-dye plant remnants, suspension, and plant tissues was done by the addition of charcoal powder and continuous filtration. The pH of the dye extract (6.75) was similar to the findings of Rather et al. (2015) in case of wool dyeing with *A. nilotica* dye by fluorescent technique. Generally, the presence of polyphenolic groups in the coloring component is the major cause of the mild acidic nature of this dye [58]. According to Gerhard John (1996) [95], the favorable pH range for dyeing of leather is 4.0-6.5. The dyes are salts of organic colored materials that are ionized in the dyeing bath during leather dyeing [96]. Therefore, the crude dye solution was mixed with sodium carbonate solution (0.001 N) at a volume ratio of 1:1. The dispersed coloring components react with the cations (Na^+) in the alkali media to form salt (Figure 2). Finally, the pH of the dye solution was standardized by using (0.01 N) acetic acid to neutralize the excess alkali and the whole mixture was heated until obtaining the dye powder.

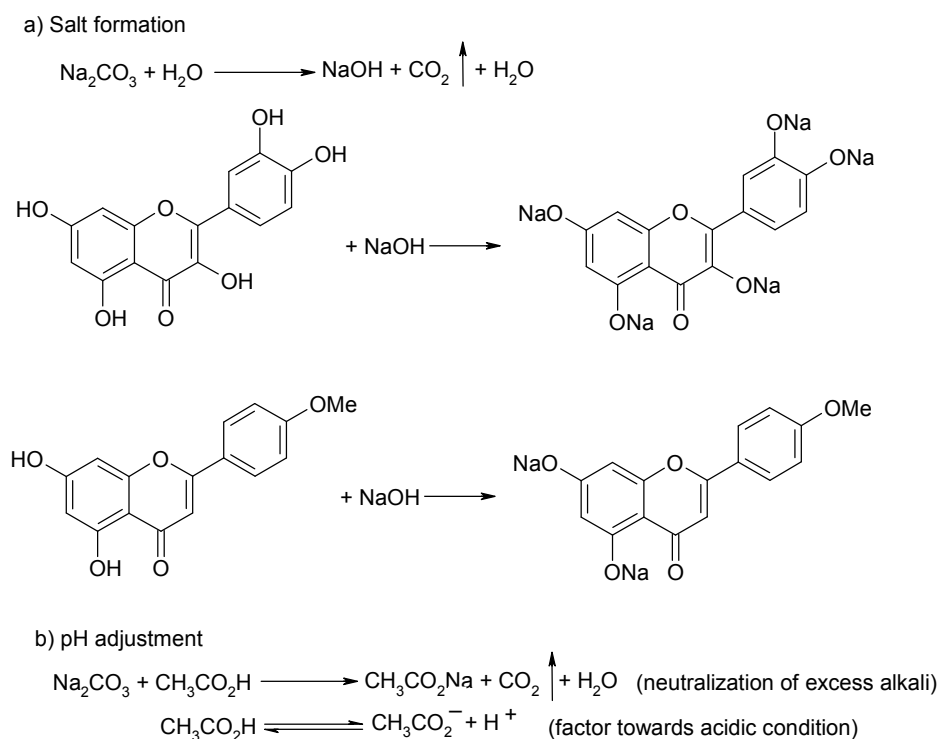


Figure 2: Formation of dye salt.

3.2. Yield and characterization

The crude solid dye was treated at 20°C and 65% relative humidity for 24h [97]. Finally, the weight of crude dye was measured in a digital mass balance to calculate the percentage of yield with respect to the weight of material used for dye extraction using the following equation (Eq. 4) [71]:

$$\% \text{ Yield} = \frac{W_2}{W_1} \times 100 \quad (4)$$

where, W_1 is the weight of the bark powder, W_2 is the weight of the crude dye. The yield was 42.7%, which is similar to the experimental result of Konar during dye extraction from plant source under optimum condition [98].

Since the extracted dye is tannin-based, the total tannin content in the crude dye was calculated with the method followed by Jain et al. [99]. The equation is as followed (Eq. 5):

$$\% \text{ w/w total tannin content} = \frac{\text{weight of n butanol fraction (g)}}{\text{weight of sample (g)}} \times 100 \quad (5)$$

The total amount of tannin content in the obtained dye was 10.33%. The researchers found the similar quantity of tannin content in *Acacia nilotica* bark extract [100-102]. Subsequent to standardization, the pH of the dye became acidic (Table 2) which is quite suitable for leather dyeing. The dye possesses excellent colorfastness and stability to various solvents.

From the chemical structure of the dye components (Figure 1), it was found that both acacetin and quercetin contain methoxy and hydroxyl groups respectively, in the side chain along with quinonoid group as their fundamental aromatic compound.

According to Witt's color production' theory [103], the quinonoid ring acts as the main coloring component named as 'Chromophore' and hydroxyl and methoxy groups act as 'Auxochrome' that help to impart color to the substrate, so the compound show color due to the presence of p-quinonoid ring in flavone structure, according to Armstrong's 'Quinoid' theory [103].

3.3. Application of dye

Several trials have been carried out to determine the coloring ability of the extracted dye on wet blue goat leathers with and without mordant. The color intensity on the grain and flesh sides of the dyed leather samples (Figure 3) was found uniform, indicating the even dispersion and distribution of the dye in the leather fiber from dye bath [104]. The penetration of dye was visually observed by taking the cross-section of the dyed leathers. Hence, *A. nilotica* natural dye can be applied for leather dyeing.

3.4. Color measurement

The color variables of both unmordanted and mordanted dyed leather samples are shown in Table 3. The colorimetric values (L^* , a^* , b^* , c^* , and h°) of the grain side of each of the leather samples were observed in this experiment. The colorimetric properties of the dyed samples depicted that the dye imparted dull color to the substrates and offered a hypsochromic effect, i.e. lower color depth. Alum mordanted samples showed brighter color than iron mordanted ones. From the color coordinates, it was observed that the lightness (L^*) of the dyed leathers decreased after mordanting due to the formation of dye-metal coronate complex, that reduced the L^* values of the mordanted dyed leathers by shade modification [105].

Table 2: Physical properties of the crude dye.

| Parameters | Remarks |
|-----------------------------------------|-------------------------------------------|
| pH | 6.75 (Experimental) 5.0 (Standardized) |
| Moisture content | 16% |
| Solubility | Aqueous media |
| Stability to acid | 5 |
| Stability of hardness of water | 5 |
| Colorfastness to solvent (formaldehyde) | 4 |
| Colorfastness to acid/alkali | 4/5 |
| Colorfastness to water spotting | 4/5 |

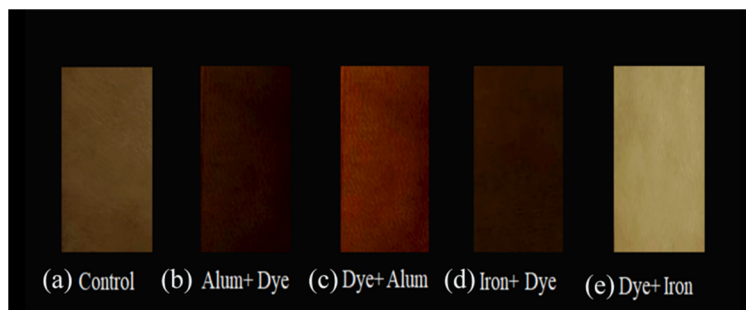


Figure 3: Leather samples dyed with *A. nilotica* natural dye (a) control; (b) pre-mordanted with alum (Alum+Dye); (c) post-mordanted with alum (Dye+Alum); (d) pre-mordanted with ferrous sulfate (Ferrous sulfate+Dye); and (e) post-mordanted with ferrous sulfate (Dye+Ferrous sulfate).

Table 3: Color coordinates¹ of the dyed leather samples.

| Sample | Dyeing Technique | L* | a* | b* | c* | h° | Color name |
|--------|-----------------------------------|-------|------|-------|-------|-------|-----------------|
| 1 | Control (Unmordant) | 58.42 | 6.56 | 15.49 | 16.82 | 67.05 | Amber |
| 2 | Pre-mordant with Alum | 40.62 | 7.96 | 15.39 | 17.33 | 62.66 | Brown |
| 3 | Post-mordant with Alum | 43.40 | 7.26 | 14.42 | 16.14 | 63.26 | Pale Brown |
| 4 | Pre-mordant with Ferrous sulfate | 49.99 | 3.61 | 8.42 | 9.16 | 66.81 | Chocolate brown |
| 5 | Post-mordant with Ferrous sulfate | 39.57 | 6.73 | 12.30 | 14.03 | 61.31 | Gray |

3.5. Dye exhaustion

Quantification of the dye uptake by test specimens is presented as the dye exhaustion rate in a graphical form (Figure 4). The dye uptake was $57.83 \pm 2\%$ in the control sample which revealed that the dye has an affinity to bind to leather fiber. However, the maximum dye exhaustion was found in the case of pre-mordanted samples especially for pre-mordanted

sample with ferrous sulfate, followed by alum and control samples. For this reason, it can be assumed that the affinity of *A. nilotica* dye with leather fiber is increased upon pre-mordant treatment. The differences in exhaustion rates were due to the differences in interactions between the fiber, the dye, and the mordant [58].

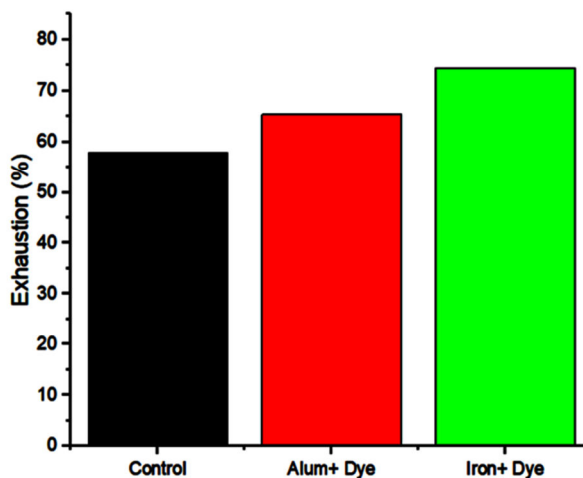


Figure 4: Dye exhaustion of *A. nilotica* dye.

3.6. Colorfastness properties

The results of several color fastness tests are represented in Table 4. The color fastness of both control and experimental mordanted samples were excellent, proving their good and comparable dyeing quality. However, the fastness to light and heat was found moderate. The presence of tannin content (ellagitannin and gallic acid) in the dye tends to fade or darken the color after exposure to high temperature [106]. The addition of 'Tocopherol' in the finishing stage could be a remedy for this problem [107]. Since this research aimed to find out the use of *A. nilotica* natural dye for leather dyeing, the attempt to improve those fastness properties is continuing in the laboratory to get satisfactory results.

3.7. Leather strength

Figure 5 represented the effect of dye molecules associated with various metallic mordants on the strength property of the dyed leather samples. The strength properties of each leather sample were found satisfactory as compared to standard values. The strength properties of the pre-mordanted dyed sample with ferrous sulfate were higher than other leather samples. The strength values for both test are in the descending order of pre-mordanted with FeSO_4 > post-mordanted with FeSO_4 > pre-mordanted with Alum > post-mordanted with Alum > control sample. Generally, the changes in strength properties of these leather samples depend on several factors such as the characteristics of the applied re-tanning materials, mordanting methods, etc. [108].

Table 4: Colorfastness test results of the dyed leather samples.

| Sample | Dyeing Technique | Light fastness | Heat fastness | Wash Fastness | | Color rub fastness | | Perspiration fastness | |
|--------|-----------------------------------|----------------|---------------|---------------|------|--------------------|-----|-----------------------|------|
| | | | | L. | C.F. | Dry | Wet | L. | S.F. |
| 1 | Control (Unmordant) | 3/4 | 3/4 | 4/5 | 5 | 5 | 5 | 4/5 | 5 |
| 2 | Pre-mordant with Alum | 3/4 | 3/4 | 4/5 | 5 | 5 | 5 | 4/5 | 5 |
| 3 | Post-mordant with Alum | 3/4 | 3/4 | 4/5 | 5 | 5 | 5 | 4/5 | 5 |
| 4 | Pre-mordant with Ferrous sulfate | 3/4 | 3/4 | 4/5 | 5 | 5 | 5 | 4/5 | 5 |
| 5 | Post-mordant with Ferrous sulfate | 3/4 | 3/4 | 4/5 | 5 | 5 | 5 | 4/5 | 5 |

L. = Leather sample, C.F. = Cotton fabric and S.F. = Standard fabric.

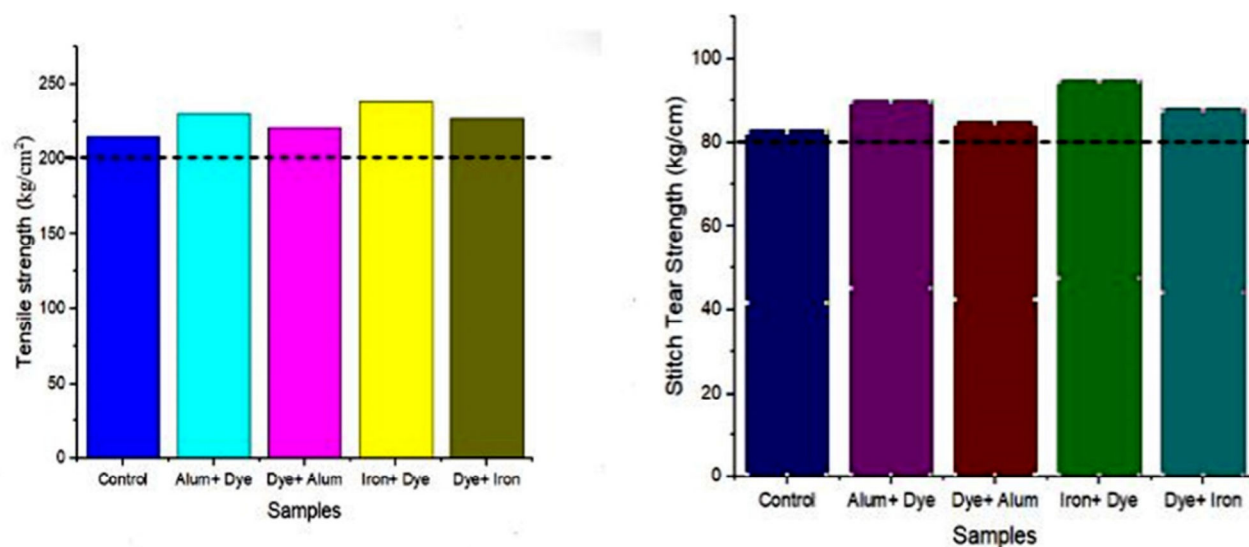


Figure 5: Leather strength of the dyed leather samples.

3.8. Dyeing chemistry

Fourier transform infrared (FTIR) spectrum of the dyed leather was analyzed by FT-IR spectrophotometer (IR Prestige 21) to investigate the interaction of extracted dye with collagen fiber as described in the literature. The IR spectra of the leather sample showed the characteristics of the peptide bond (-NHCHO-), which is the main unit of the polypeptide chain. For chrome tanned leather (wet blue), the oscillation of peptide bond gives two main IR spectral bands in the spectrum known as Amide I for stretching ν (>C=O) vibrations at \sim 1600-1700 cm^{-1} and Amide II for stretching ν (-C-N) vibrations and bending ν (N-H) at \sim 1400-1500 cm^{-1} which has a very sensitive configuration [109-111]. A complex band Amide III, often appears at \sim 1200-1350 cm^{-1} due to the nature of side chains and hydrogen bonding. Moreover, the Amide A appears at about 3300 cm^{-1} because of -NH stretching vibration and the backbone conformation, which is very sensitive to the strength of hydrogen bonds [109, 112].

The IR spectra of the dyed leather showed distinguished absorption peaks. A broad peak appeared near 4000-3500 cm^{-1} due to -OH bond and -NH stretching in the collagen fiber. Strong peaks at 1612, 1496, and 1369 cm^{-1} represented amide I, amide II, and amide III, respectively. The peaks ranged in 3309 cm^{-1} for amide A and other functional groups such as C-C, C-H, C=C, etc. in the figure print region depicted the nature of the backbone structure of the collagen fiber (Figure 6). The common interaction of dye molecules with fiber affected the changes in band intensity and wavenumbers of the dyed leather as compared to the

structure of wet blue leather [113].

During mordanting, the interaction of metal ions (mordant) with -OH and >C=O groups of the collagen fiber, led the absorption peaks to shift at range 2979 cm^{-1} and 1774 cm^{-1} , respectively. Strong peaks at 1645, 1413, and 1332 cm^{-1} , represented amide I, amide II, and amide III, respectively, in the case of alum mordanted leather.

In collagen fiber, the monomers are linked with various side-chains by anionic (carboxyl) and cationic (amino) groups. During dyeing process, the extracted dye salts were dissociated in the dye bath and penetrated into the leather fiber at pH near to its isoelectric point (IEP) [114]. After penetration, the dye molecules fixed with the polarized peptides by forming salt-linkages. As the H atom had weak bonding to N atom in the polypeptide chain by either hydrogen bond or van der Waals' force, the H atom presented in the cationic part (amino) was easily replaced with anionic part of the dye at an acidic pH value controlled by the addition of formic acid [6].

However, in the case of pre-mordanting, the metal ions form complex compound with water molecules and further dissociated into ligands. These ligands react with the anionic part of the collagen structure, act as salt link, and further interact with dye ions through replacement of water molecules [115]. Moreover, dye molecules reacted with the cationic amino groups of the collagen fiber by the same dyeing mechanism discussed for control sample [116]. Therefore, pre-mordanted leather samples uptake more dye onto the leather fiber than unmordanted leather [117].

Table 5: Characteristics of amide bands of dyed leather fiber in FT-IR spectra.

| Sample | Amide band | Position (cm^{-1}) | Intensity (%T) |
|-------------------------|----------------|-------------------------------|----------------|
| Chrome tanned leather | Amide band I | 1669 | 2.3485 |
| | Amide band II | 1446 | 1.1240 |
| | Amide band III | 1394 | 1.3771 |
| Dyed leather | Amide band I | 1612 | 3.1239 |
| | Amide band II | 1496 | 3.1263 |
| | Amide band III | 1369 | 3.0658 |
| Pre-mordanted with alum | Amide band I | 1645 | 3.008 |
| | Amide band II | 1413 | 2.7361 |
| | Amide band III | 1332 | 1.6967 |

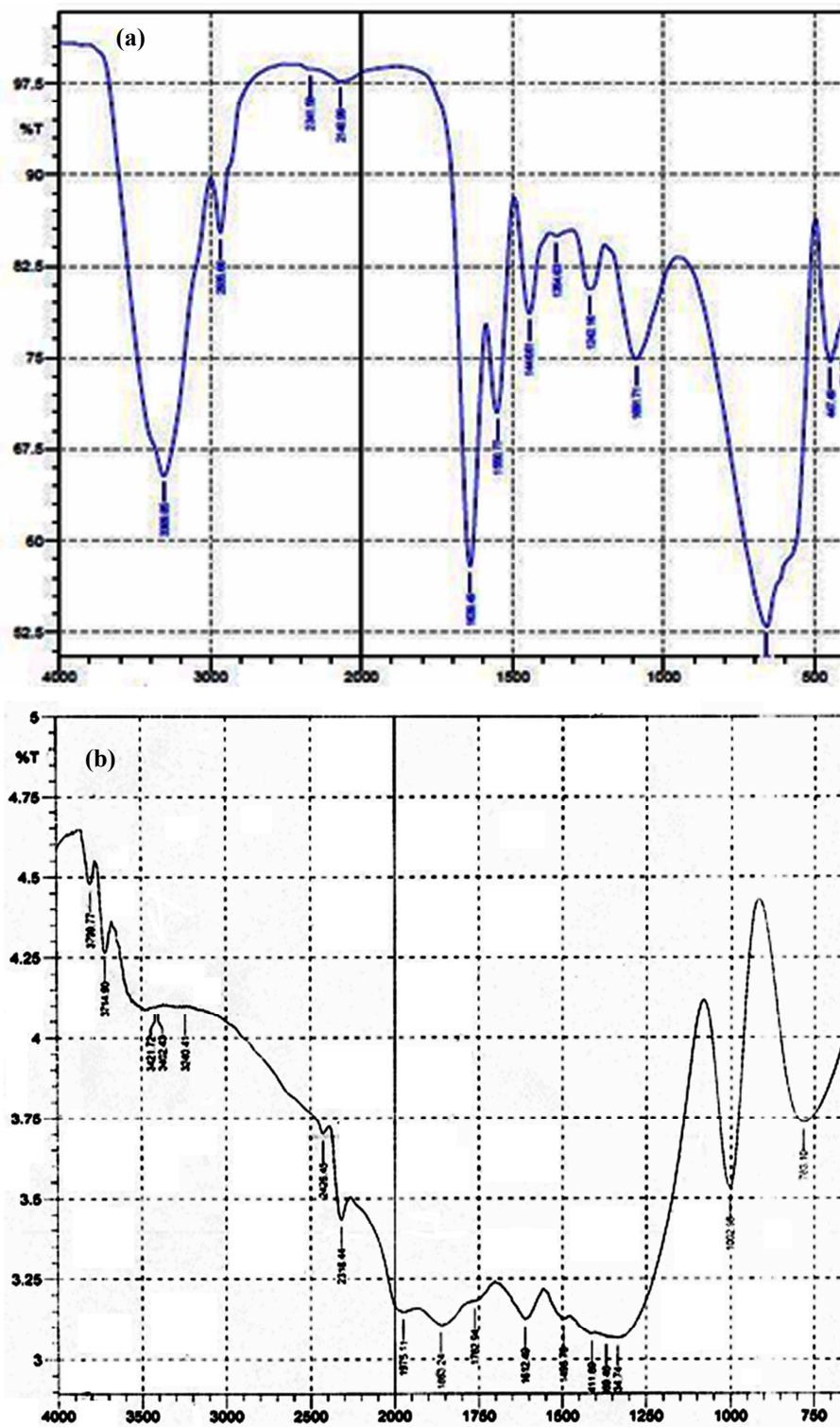


Figure 6: FT-IR spectra of (a) chrome tanned leather, (b) dyed crust leather (control) and (c) alum mordanted dyed crust leather.

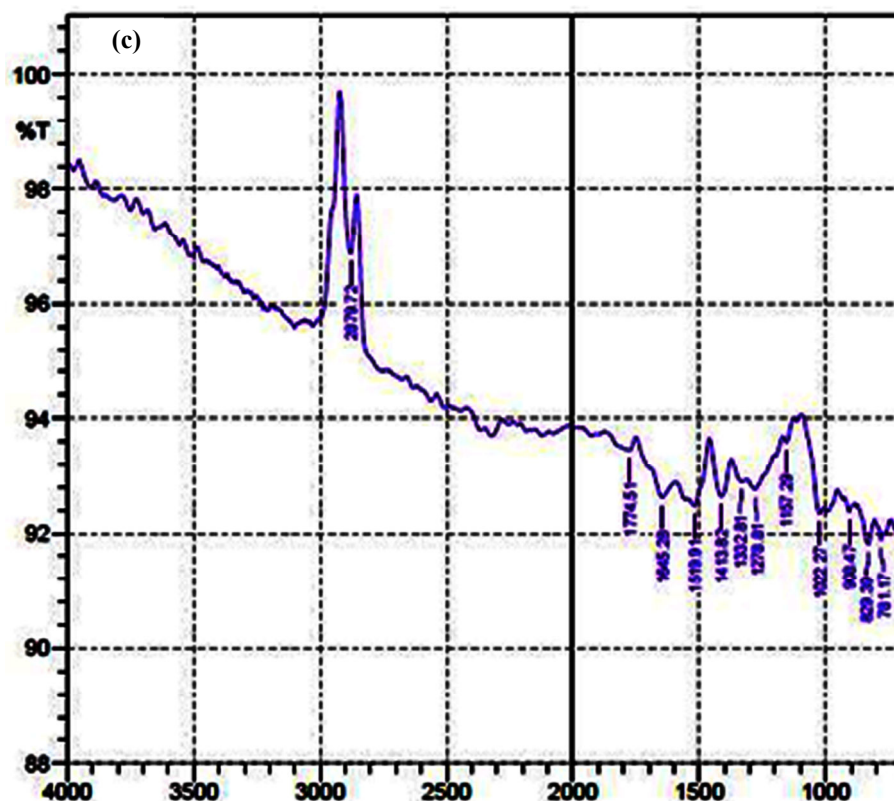


Figure 6: Continue.

3.9. Dyeing efficiency

Since natural dyes have drawn the attention of both manufacturers and buyers, scientists are now working on finding new sources of natural colors and their application. In this research, *Acacia nilotica* dye was used for leather coloration and the dyed leathers demonstrated satisfactory colorfastness. The compatibility of *A. nilotica* natural dye is evaluated in Table 6 as compared to other natural brown dyes. Among recent researches, the dye extracted from *Laccifer lacca* had an excellent binding capacity with leather fiber but tended to weaken the substrate [83, 118]. Sivakumar et al. (2009) [119] and Velmurugan et al. (2009) [125], reported that the dyed leathers with beetroot extract and fungal dye, respectively, possessed noticeable colorfastness properties, but had poor

resistance to perspiration and wet rub fastness. Moreover, the dyeing methods of those natural dyes required sophisticated machinery that might increase the production costs. Similarly, some dyes obtained from microorganisms and flower/fruit extracts showed inadequate fastness properties to light and machine-washing [119-126]. There is a lack of information on mechanical properties, dye absorption capacity, and economic benefits in most of the studies. As compared to previous literature, *A. nilotica* dye provides better fiber strength and dyeing properties. The exhaustion capacity of the dye can be improved by mordanting. Therefore, advanced studies are required to evaluate its antibacterial potency, reproducibility, and industrial application.

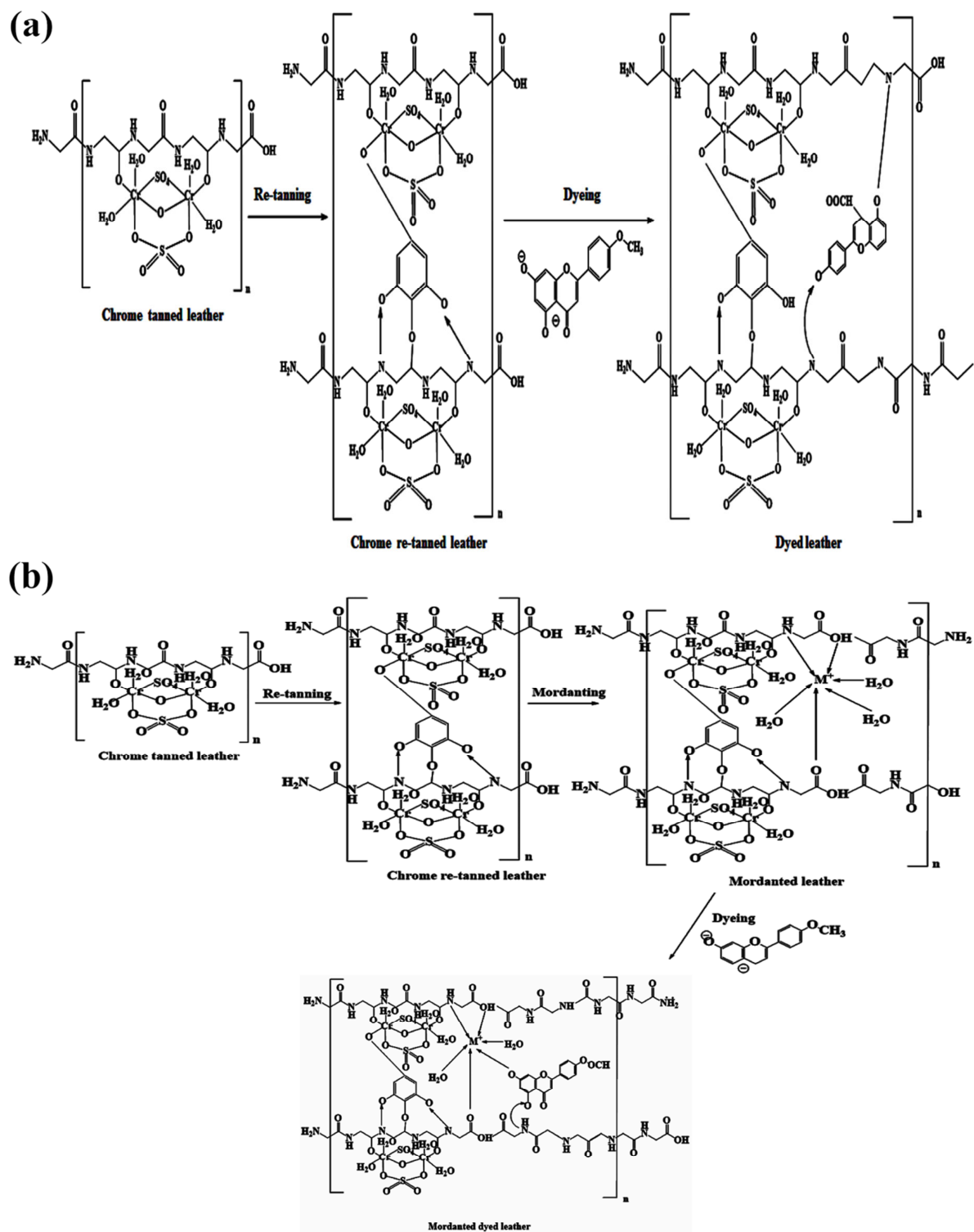


Figure 7: Possible schematic diagram of (a) dye-fiber and (b) mordant-fiber-dye interaction.

Table 6: Comparison of leather dyeing quality with other natural dyes.

| Sources | Parameters | | | | | | | | | References |
|------------------------------|----------------|---------------|----------------|---------------|--------------------|-----|-----------------------|----------------------------------------|------------------------------|---------------|
| | Exhaustion (%) | Heat fastness | Light fastness | Wash fastness | Color rub fastness | | Perspiration fastness | Tensile strength (kg/cm ²) | Stitch tear strength (kg/cm) | |
| | | | | | Dry | Wet | | | | |
| <i>Laccifer lacca</i> | 86.42 | - | 4/5 | 4/5 | 4/5 | 4/5 | - | 84.71 | - | [83, 118] |
| <i>Betal vulgaris</i> | 62 | - | - | - | 3 | 2/3 | 2/3 | - | - | [119] |
| <i>Ficus benghalensis</i> L. | - | - | 4 | - | 4 | 4 | - | - | - | [120] |
| <i>Osyris lanceolata</i> | - | - | 8(BS)≅5(GS) | - | 5 | 4 | - | - | - | [121] |
| <i>Curcuma longa</i> | - | - | 4 | - | 4 | 3 | - | - | - | [122] |
| <i>Cassia singueana</i> | - | - | 6(BS)≅4/5(GS) | - | 5 | 4/5 | 5 | - | - | [123] |
| <i>Isaria spp.</i> | 58±1.4 | | 3/4 | 4/5 | 4/5 | 2/3 | 2/3 | 3 | 3 | [124] |
| <i>Emericella nidulans</i> | 55.5 | - | - | 3/4 | 4 | 2/3 | - | - | - | [125] |
| <i>Rubia tinctorum</i> | 79.13 | | 2/3 | | 4 | 2/3 | 4/5 | - | - | [126] |
| <i>Acacia nilotica</i> | 57.83±2 | 3/4 | 3/4 | 4/5 | 5 | 5 | 4/5 | 215 | 85 | This research |

BS= Blue scale, GS= Gray scale

4. Conclusion

The extracted dye from *Acacia nilotica* plant bark was used for leather dyeing. The dyed leather samples, especially pre-mordanted dyed samples, showed satisfactory fastness properties and color strength. According to spectral analysis, the obtained dye easily interacted with collagen fiber. Moreover, the strength property of the dyed leather was increased and various color shades were produced by mordanting techniques. Therefore, the obtained natural dye from *Acacia nilotica* bark can be applied as a potential substitute for conventional synthetic dyes to bring about the 'cleaner production' concept in the leather industry. However, future studies are needed to introduce more efficient

dyeing methods to reduce dyeing cost for industrial application.

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