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Color enhancement of chrome tanned goat leathers using aqueous tea extracts as dyeing agent

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Abstract

The leather industry is undergoing a transformative shift towards sustainable practices due to increasing environmental concerns associated with synthetic dyes. This research explores the potential of natural dyes derived from tea extracts as an eco-friendly alternative for leather dyeing. In this study, the aqueous extracts of dried commercial tea leave (*Camellia Sinensis*) (10%, 12%, and 15% w/v) with and without mordant were used for dyeing of chrome tanned goat leathers and its color intensity parameters were examined using Milton Roy Colormate HDS spectrophotometer. In addition, the quality of dyed leather was evaluated by dye exhaustion test, fastness test, tensile strength test, scanning electronic microscopy (SEM) imaging, and thermogravimetric analysis. The optimized application of dye extract was 12% (without mordant) showed better color intensities (L* = 56.33 ± 0.57, DL* = -13.74 ± 0.48), highest dye exhaustion (84.68 ± 0.06%), tensile strength (18.75 ± 0.25), and residual mass in TGA, better color fastness (wet-4/5, dry-4/5, perspiration-3/4, and light fastness-4/5 in Grey Scale rating), and best fiber orientation in SEM image. In addition, another optimized was (with mordant) 15% dye extract with 1% Copper (II) Sulphate produced the highest color intensity (L* = 51.21± 1.06, DL* = -18.86±0.72). These findings offer the potential of tea extracts as a viable solution for natural leather dyeing, offering both environmental and aesthetic benefits.

Keywords: Natural dyes; Color enhancement; Chrome tanned leather; Tea extracts; Leather dyeing.

Introduction

Dyes can generally be divided into two types: natural and synthetic. Since ancient times, characteristic dyes have been used in shading nourishments, calfskin, wool, silk, cotton and more. In recent times, the number of studies on characteristic colors has also begun to expand since interest in items with general colors has expanded (Pan *et al.* 2003). Interest in natural dyes and their applications is increasing due to environmental safety and growing awareness (Bydoon, 2016; Kumar *et al.* 2009; Mirjalili *et al.* 2011; Li *et al.* 2022; Viana *et al.* 2015). Application of the natural dyes in industry has been declined due to the affordability and wide availability of synthetic dyes (Tsujimura and Takasu, 1955; Tsujimura, 1934). There is particular importance in textile, leather and in decentralization sector for special products for synthetic and natural dyes (Deo and Desai, 1999).

However, synthetic dyes have adverse effects on the environment and health related issues such as allergy, toxicity, carcinogen, cancer causing and other harmful reactions. In case of natural dyes, they are not hazardous to health and environment friendly (Mahanta and Tiwari, 2005). Eco-friendly, non-toxic natural dyes as a potentially viable "green chemistry" alternative have re-emerged as alternatives/partners to a number of synthetic dyes amid growing environmental and health concerns (Wang *et al.* 2022). Since the natural dyes are easily biodegradable, this industry is an eco-friendly process,

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besides these dyes have excellent medicinal and sustainable properties (Dweck, 2002; Gulrajani, 1999). Thus, natural dyes can play an important role in overcoming environmental degradation caused by synthetic dyes (Tsatsaroni and Eleftheriadis, 1994; Taylor, 1986). The leaves of many plants are used as a source of natural dyes, one of which is tea (*Camellia sinensis*).

Tea is an evergreen shrub that grows mainly in tropical and subtropical regions of China, Taiwan, India and Japan. This plant extract contains various compounds with potent antibacterial, anticancer, antioxidant and antifungal properties and has been widely used as medicine since ancient times to treat various diseases (Failisnur *et al.* 2019; Amutha and Annapoorani, 2019). The use of tea as a natural dye in natural fibers has been reported in the literature (Liu *et al.* 2009). Tea contains most of the poly phenols known as catechins, where epicatechin and their derivatives are the most ascending form. The highest concentration is gallic acid ester epigallocatechingallate (EGCG) making up over 61% of the epicatechin derivatives (Adeel *et al.* 2022; Keka *et al.* 2012). There are various classes of compounds such as carbohydrates, caffeine, amino acids, chlorophyll, carotenoids, nucleotides, lipids, saponins, minerals, organic acids, polyphenols, unsaponifiable compounds etc found in tea (Hilali *et al.* 2022). Theaflavins and Thearubigins are the chief coloring components in tea. Both compounds have hydroxyl groups in their structures in a position which is favorable for formation complexes with required metal (Sun *et al.* 2022).

The objectives of this study were to use natural dyes such as tea extracts for reducing the toxicity occurrence due to use of synthetic dyes in leather processing. Hence, the outcome of this study will help the leather industry of Bangladesh to initiate the utilization of natural dyes from tea processing because of availability.

In this study, various concentration (10%, 12%, and 15%) of aqueous tea extracts had been prepared from dried tea samples available in open market. Then those aqueous extracts of tea were applied on chrome tanned goat leathers for enhancing color of chrome tanned goat leathers. Those chrome tanned goat leathers were prepared in our laboratory for equal distribution of tanning with chromium at same environment. After that, aqueous extracts of tea had been used at retanning stage as dyeing agent in chrome tanned goat or wet blue goat leather.

Materials and methods

Materials and chemicals

Raw goat leathers were purchased from local markets. Then, those raw leathers were processed in our laboratory drum for making chrome tanning leather. After two weeks later, those wetted undyed chrome tanned goat leathers were used for dyeing of leathers with various concentration of aqueous tea extracts without or with mordants. Dried tea leaves were collected from local open market Mirpur-2, Dhaka, Bangladesh. All chemicals utilized for chrome tanning were of commercial grade. Ferrous sulphate and Copper sulphate used for dyeing process with tea extract as mordants of laboratory grade. Dried jambolan (*Syzygium cumini*) leaves powder and taro (*Colocasia esculenta*) leaves were collected from nearby local sources; those plants extracts were used as natural mordants.

Instrumentations

Hot dried tea leaves extracts were cooled and filtered by vacuum pump filtration with whatman (125 mm Dia) filter paper. Dried jambolan (*Syzygium cumini*) leaves were grinded in automated grindermachine (FRITSCH Pulverisette 2, Germany). Dye exhaustion was measured by UV/Vis Spectrometer SPECORD 210 plus, analytikjena, Germany. Hanging Wodden Experimental Drum (Model: GJOB2, drum size: 1300 × L 1000 (mm), speed: 4 rpm (made as customer's request), motor Power: 1.5 KW-4P (AC 220V, 50 Hz), drum door: 400 × 400 (mm), company: Yancheng Shibiao Machinery Manufacturing Co. Ltd, origin: Jiangsu, China) was utilized for chrome tanning of raw goat leather and dyeing of chrome tanned leather. For Chroma experiments, Colorimeter (Model: CR400, Brand: Konica Minolta, Origin: Japan) was used. Thermal properties (TGA) were investigated by a Simultaneous Thermal Analyzer (STA, 449 F3 Jupiter®; NETZSCH GmbH, Germany). The SEM analysis was done by SEM (JSM-7610F, JEOL, USA).

Preparation of extract from tea leaves

The required amount of dried tea leaves in three 500 mL beaker was poured in water (1:10w/v) at temperature 100^{o}C with continuous stirring by magnetic bar for 30 minutes, 40 minutes, and 60 minutes, respectively, filtered and cooled. Part of the extract was filtered through Whatman (125 mm Dia) filter paper and 100 mL of filtrate was used for the determination of percentage of total soluble containing in aqueous extracts by evaporating in an oven at 105ºC. From this procedure, three dye extract concentrations were found.

FTIR of Crude tea extract

FTIR spectrum of optimum dye extract was measured using Perkin-Elmer Fourier Transform Infrared Spectrometer contained a diamond attenuated total refection accessory (Frontier, Perkin-Elmer, UK; Software: Spectrum Version 10.4.4). The spectrum was measured from range 600-4000 cm^{-1} at a scan rate 4 cm^{-1}.

Procedure of dyeing

After chrome tanning, the tanned leather samples were piling for two weeks, then washed, dyed with aqueous tea extracts (pH= 6.8) of various concentrations (10%, 12%, 15%) at 30°C using the process given in Table I and II. Then, those processed liquors were collected and examined for exhaustion of tea dye from aqueous extracts.

Table I. Formulation of chrome tanning process of raw leathers

Process	Reagents	Temperature (°C)	Drum Rotation Time/ (minutes)	Remarks and pH
Soaking	400% H_2O 3% Wetting agent (surfactant) 0.1% Busan 40L (Bacteside)	30	Overnight	Draining out water after soaking
Liming	80% H_2O 2% Liming Oxilary (Binkale A)	30	15	
	2% Na_2S 2% Lime	30	10	
	60 (resting time)			
	2% Na_2S 2% Lime	30	2 days	
	Drain Out and wash with 100% H_2O			
Deliming	75% H_2O 2% Ammonium Sulphate 0.5% Sodium Metabisulphate	30	60	Check leather by cutting and pH (≤7.5)
Bating	2% Bating agent 2% Wetting agent (Surfactant)	30	60	
Pickling	80% H_2O 8% Common Salt (NaCl)	30	10	
	0.5% Formic Acid	30	30	
	1.1% H_2SO_4	30	120	Check leather pH (≤3.5)
Tanning	4% Chrome 0.5% OCS (Chrome stable fat)	30	60	
	4% Chrome 0.5% Sodium formate	30	120	Check leather by cutting whether Cr penetrates through leather
	1.5% R-RF (Relugan RF)	30	60	
	1% Sodium Bicarbonate	30	45	Check leather by cutting and pH=3.5-4
	0.1% Busan 30L (Fungesite)	30	15	
	Drain out and piling those leather stuffs for two weeks			

There were two types of mordants used for dyeing process. One is natural mordant and another is inorganic mordant. Natural mordants were dried jambolan (*Syzygium cumini*) leaves powder water extracts and taro (*Colocasia esculenta*) leaves mesh water extracts; whereas copper sulphate and Ferrous sulphate used as inorganic mordant. Those mordants were used before dyeing step. For dyeing process, 1% inorganic mordants and 5% natural mordants were used.

Table II. Formulation of dyeing of chrome tanned goat leathers

Process	Reagents	Temperature (°C)	Drum Rotation Time/ (minutes)	Remarks and pH
Washing	200% H_2O 0.2% LD 600 (Wetting agent)	30	10	Draining out water after washing
Neutralization	80% H_2O 2% NG (Neutral Syntan) 0.5 Sodium Formate	30	45	Check leather by cutting and pH=4.5-4.9
Retanning	100% H_2O 3% RS3 (Resin Syntan)	30	20	Check bath after treartment & pH (≤5)
	Mordants (1% or 5%)	30	10	
	Tea Dye (10%, 12% and 15%)	30	20	
	4% R-D (Relugan-D) 1% NFO (Natural Fat) 4% OS (Tenogen OS) 2% AN (Syntan AN) 4% Mimosa (for sample 2 and 6) 2% Quebracho (for sample 2 and 6)	30	90	
	1.5% Formic Acid	30	20	
Fat liquoring	80% H_2O	50	1	
	4% SLP (Anionic Fat) 3% SL335 (Anionic Fat) 1% NFO (Natural Fat)	50	60	
	0.2% Busan 30L (Fungesite) 1% Formic Acid	50	25	
	0.2% Cationic Fat	50	5	
	Drain out and wash with 100 H_2O			

In table II, the dyeing processes of chrome tanned goat leather were followed washing, neutralization, retanning and fat liquoring stages. At retanning stage, the dyeing of leather tea extarcts was occured.

Processing after leather dyeing

After completion of dyeing with fat liquoring treatment, first those leather stuffs were air dried for 7 days. Then, those were air dried in hang drier for 3 days, shaved in shaving machine, troughed in toughing machine and ironed with ironing machine. At last, those leather stuffs were used for Chroma experiments for color measurements. After various dyeing process of leathers, leather colors were compared with leather color chart (Table III). Those colors of leather samples are shown in Figure 2.

Conditioning of dyed leather

Leather samples for analytical and physical testing were cut from official sampling position (Krishnamoorthy *et al.* 2013). After that, those samples were measured for color values.

Analysis of leather after dyeing

Tensile strength properties

Tensile strength properties were measured by Tensile tester equipment (Model No.: STM 566, Company: SATRA, Kettering, UK).

Fastness properties

Fastness properties were collected using instruments such as Bally Finish Tester/Color Fastness Tester (Model No.: STM 643-B, Company: SATRA TECHNOLOGY, Castro Engineering Pvt. Ltd., Dasnagar, Howrah, India) and Verivide (See in Truth), Color Fastness Cabinet (CF 60), UK using D65 lamp for light fastness measurements.

Dye exhaustion

Dye concentration in spent liquor was measured using the absorbance value at 280 nm (λmax) by UV–visible spectro photometer (Yoo and Jeon, 2012; Chen *et al.* 2022). Using the following equation, the dye exhaustion was measured:

$$\%\text{Dye exhaustion}=[(Co–Cs)/Co] \times 100$$

Where, Co is the concentration of dye offered and Cs is the concentration of dye in the spent liquor.

Color measurement

Chrome tanned goat leather (undyed) and leathers dyed with dried tea leaves aqueous extract with or without mordants were introduced to color intensity measurements. The color intensity parameters L*, a*, b*, c* and h were examined using Milton Roy Colormate HDS spectrophotometer for the chrome tanned goat leather samples. Color quantification was measured using the Commission InternationaledelEclairage (CIE) system of color measurement with 10° standard observer data. Here, L*, a*, b*, c* and h represent the clarity, the chromatic component red-green axis, yellow-blue axis, color saturation and tonality (angle) of color (Sivakumar *et al.* 2009).

Dyeing characteristics evaluation

The color intensity of chrome tanned goat leather samples was evaluated using the difference of values of undyed chrome tanned goat leather and chrome tanned goat leathers dyed with various dyeing agents. The negative values of DL*, Db*, Dc*, Dh and positive value Da* show the reasons for more color intensity of dyeing of leathers (Selvi *et al.* 2013; Sivakumar *et al.* 2009; Vedaraman *et al.* 2017; Ashrafi *et al.* 2018).

Thermogravimetric analysis

The thermal properties (TG) of prepared leather (only dye extracts and control sample) were determined by a simultaneous thermal analyzer (STA, Model: 449 F3 Jupiter®; Brand: NETZSCH GmbH) from room temperature to 600°C at a 10 K/min heating rate in an inert atmosphere (N_2 gas at 40 mL/min).

SEM Analysis

Dyed leather samples (only dye extracts and control sample) cross-section area was assessed using FESEM through placement on conducting carbon type sampling technique. The photographs were obtained by operating the FESEM (JSM-7610F, JEOL, USA) at an accelerating voltage of 10 keV with 1000 × magnification.

Results and discussion

FT-IR Spectrum of tea extract

FTIR spectrums of crude tea extract is shown in Figure 1. A wide peak is shown at 3314 cm^{-1} corresponding to O–H and N–H stretching modes vibrations of polyphenolic compounds. Peaks at 2186 cm^{-1}, 1690 cm^{-1} are indicated C=C asymmetric and symmetric stretching modes of vibration

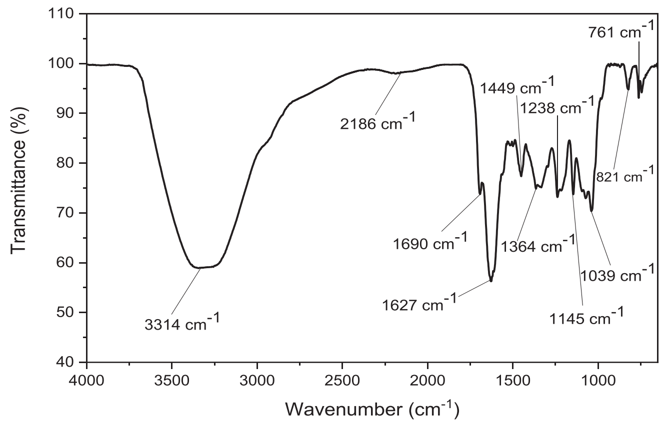

Fig. 1. FTIR spectrum of crude tea extract

in aromatic ring respectively. The C=O stretching modes of vibration in polyphenolic compounds is shown at 1627 cm^{-1}. –CH2 and C–OH bending modes of vibrations are shown at 1449 cm^{-1} and 1364 cm^{-1}, respectively. C–O–C and C–C stretching modes of vibrations are shown at peaks 1238 cm^{-1}, 1145cm^{-1}, 1039 cm^{-1} and 821 cm^{-1}, 761 cm^{-1}, respectively. So, FTIR spectra of tea extract shows peaks for carboxylic acid, polyphenols and amino acid as main functional groups (Brza *et al.* 2020; Pavia; *et al.* 2015).

Concentration of dried tea leaves aqueous extract

Dried tea leaves aqueous extracts were contained 10%, 12% and 15% tea extract concentration. 15% aqueous tea extract concentrate solution was used with 1% inorganic mordant for dyeing. Another 5% two natural mordants were comibed with same concentrate aqueous extract solution of tea for dyeing.

Table III. Color comparison of various leather dyeings

Leather Sample No.	Dyeing solutions descriptions	Color after dyeing with chrome tanned goat leather
1	No dyeing solutions were added (considering as control)	Azure
2	Mimosa and Quebracho as coloring agent	Tan
3	Only 10% tea dye extracts as coloring agent	Tan
4	Only 12% tea dye extracts as coloring agent	Soze
5	Only 15% tea dye extracts as coloring agent	Tan
6	10% Tea dye extracts with Mimosa and Quebracho as coloring agent	Soze
7	15% Tea dye extracts with 5% dried jambolan (*Syzygium cumini*) leaves powder water extracts as natural mordant	Tan
8	15% Tea dye extracts with 5% taro (*Colocasia esculenta*) leaves mesh water extracts as natural mordant	Soze
9	15% Tea dye extracts with 1% $FeSO_4.6H_2O$ as inorganic mordant	Burlywood
10	15% Tea dye extracts with 1% $CuSO_4$. $5H_2O$ as inorganic mordant	Bronze

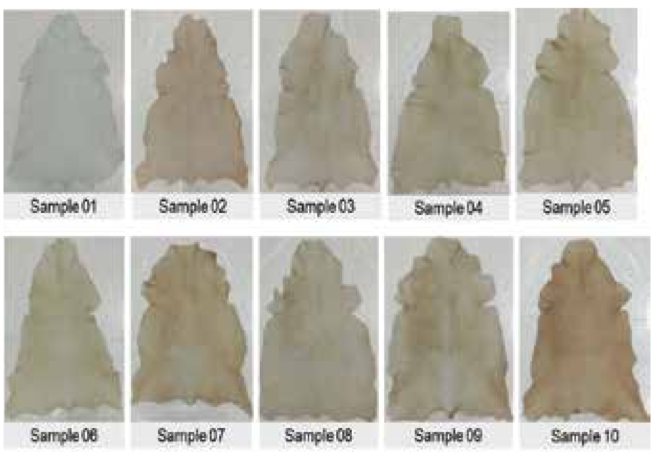

Fig. 2. Physical appearances of chrome tanned goat leathers dyeing from sample 1 to 10

Dyeing with and without mordant

The color values of control (sample 1), chrome tanned goat leathers dyeing with aqueous tea extract with various concentrations with or without mordants were given in Table 4 and Table 5. In all cases, inorganic mordants $CuSO_4$. $5H_2O$ or $FeSO_4.6H_2O$ and natural mordants dried jambolan (*Syzygium cumini*) leaves powder water extracts or taro (*Colocasia esculenta*) leaves mesh water extracts were utilized before dyeing treatment as pre-mordant applications. Using increasing amounts of natural dyes, the color of chrome tanned goat leather was deeper gradually (Table III, IV, and V and Figure 2). Before dyeing treatments, different mordants usage gave different colors and color values. After treatment with tea extracts the L* values were found decreasing with increasing concentrations, which was a sign that tea dyes treated with chrome tanned goat leathers were darker than undyed control sample 1. However, comparing mimosa and quebracho as dyeing agents solely for sample 2 and sample 6 where 10% tea dye extracts along with additional mimosa and quebracho as dyeing agents, the L* value was increasing for sample 6, which was an anomaly. May be it can be happened for the competition of different dyeing compounds at dyeing stages. The h values of tea dyes extracts treated leathers were decreasing compared to the undyed sample1, which were another signs of the colorings. But for sample 6, the h value was increasing which might be considered another anomaly for the decreasing trend of h values. In colorimetry, the "h" value, or hue angle, can differ between measurements due to several factors, including variations in surface texture or glossiness. Glossy or textured surfaces can scatter light differently than matte surfaces, affecting the measured hue (Ly *et al.* 2020).

Table 4. Color values of chrome tanned goat leathers after dyeing

Sample No.	L*	a*	b*	c*	h
1	70.17 ± 0.31	0.4 ± 0.02	2.12± 0.26	2.26 ± 0.26	79.77 ± 0.37
2	56.27 ± 0.20	6.14 ± 0.09	15.72 ± 0.08	17.13 ± 0.14	65.93 ± 0.36
3	57.92 ± 0.81	5.22 ± 0.15	16.80 ± 0.47	17.72 ± 0.84	72.12 ± 0.35
4	56.33 ± 0.57	3.22 ± 0.10	16.39 ± 0.40	16.70 ± 0.39	78.86 ± 0.26
5	52.21± 0.25	5.20± 0.22	20.04 ± 0.45	20.70 ± 0.46	75.53 ± 0.26
6	60.57± 0.23	2.39± 0.07	16.59± 0.27	16.76± 0.32	81.79± 0.13
7	54.15± 1.09	5.89± 0.17	19.22± 0.17	20.01± 0.16	72.78± 0.52
8	55.10± 0.07	4.58± 0.04	16.27± 0.07	17.02± 0.08	74.04± 0.10
9	52.17 ± 0.49	5.32 ± 0.09	18.71 ± 0.09	19.45± 0.06	74.12± 0.41
10	51.21± 1.06	7.35± 0.06	21.74± 0.39	22.91± 0.32	70.30± 0.20

Table 5. Color value differences according to control sample 1

SL. No	Sample No.	Color value differences according to control sample 1				
		DL*	Da*	Db*	Dc*	Dh
1	2	-13.90± 0.30	6.75± 0.07	13.50± 0.17	14.97 ± 0.20	-12.85±0.32
2	3	-12.13± 0.61	4.82 ± 0.10	14.58 ± 0.36	15.46 ± 0.55	-7.15 ± 0.31
3	4	-13.74 ± 0.48	2.82 ± 0.07	14.17± 0.33	14.44± 0.32	-0.91 ± 0.31
4	5	-17.86± 0.32	4.80 ± 0.13	17.82 ± 0.35	18.44 ± 0.36	-4.34± 0.31
5	6	-9.50± 0.31	1.98 ± 0.15	14.17 ± 0.25	14.50 ± 0.27	2.01 ± 0.25
6	7	-15.82± 0.74	5.49± 0.11	16.94± 0.21	17.75± 0.21	-6.89 ± 0.44
7	8	-13.87 ± 0.23	4.28± 0.04	14.15 ± 0.16	14.76 ± 0.17	-5.73 ± 0.23
8	9	-17.90± 0.44	4.92± 0.07	16.59± 0.17	17.19 ± 0.16	-5.65 ± 0.39
9	10	-18.86±0.72	6.95 ± 0.05	19.52 ± 0.32	20.65 ± 0.29	-8.47 ± 0.28

The differences of color values gave the dyeing characteristics according to Table 5. The decreasing trends of DL*, Db*, Dc*, Dh values and the increasing trend of value Da* showed the reasons for more color intensities and darkness of color respectively for dyeing of leathers. The most prominent considering facts for deep colorings of leathers were decreasing of DL* values. Because it's more negative values (Table V) defined the colors were darker. Then, the Da* values increasing trends took the second place for color darkness measurements because of it reddish color trends. The Db*, Dc* and Dh values color decreasing trends considered to be another reasons for color intensities at last stage (Selvi *et al.* 2013; Sivakumar et al 2009; Vedaraman *et al.* 2017; Ashrafi *et al.* 2018). According to those considerations, the color intensities can be followed an increasing trend order following for sample 10, sample 9, sample 5, sample 7, sample 8, sample 2, sample 4, sample 3, and sample 6, respectively (Table 4 and Table 5). From those interpretations showed that inorganic mordants applications gave the highest intensities of leather coloring rather than natural mordants. Another fact was vital that, using mimosa and quebracho as dyeing agents along with tea extracts at retanning stage did not enhance color intensity, rather decreasing trend following. The same things happened for applications of natural mordants with tea extracts at retanning stage. In general, 15% tea extracts along with inorganic mordants gave better results compared to other options.

Mordant $CuSO_4$. $5H_2O$ treatment with tea dye extracts (sample 10) showed the highest value of b* which was an indication that this leather was more yellowish compared to other leathers. Whereas the undyed leather sample 1 gave the lowest b* value compared to other leathers trending to bluish color. The L* values were decreasing and a* values were increasing meaning the darkness and redness of leather colors, which indicated leathers coloring values increasing. In these cases, the L* and a* values of tea extracts treated leathers compared to undyed leather sample 1 were decreasing and increasing respectively according to Table 4 and Table 5, which were clear indications of increasing trends of colors. Four samples showed the higher 'a*' values compared to sample 1, and they were sample 10, sample 2, sample 7 and sample 9. The 'L*' values of leathers treated with tea extracts along with mordants $CuSO_4$. $5H_2O$, $FeSO_4.6H_2O$ and two other natural sources mordants gave lower values representing the darkness of colors shade increasing. However, the variations of color shades and intensities of colors can be clearly interpreted from the values L*, a*, b*, c*, h, their differences of values DL*, Da*, Db*, Dc*, Dh and from Table IV and Table V and Figure 2.

Dye exhaustion

The dye exhaustion for chrome tanned leather dyeing was decreased with increasing concentration after 12% tea dye extracts. Here, the dye exhaustion for 12% tea extract concentrated solution was found highest 84.68 ± 0.06%. Whereas, dye exhaustion for 15% and 10% concentrated tea extracts solutions were found 82.86 ± 0.11% and 84.55 ± 0.08% respectively. Here, dye penetration rate for 15% tea extracts was lowest from UV absorbance at 280 nm.

Color fastness tests of dyed leather samples

Color rub fastness test is an indicator for quality of leather dyeing. Dry rub, wet rub, perspiration and light fastness tests of the leather samples were measured according to norm of SATRA PM 08 (Patil *et al.* 2014). Here, all leather dyeing of tea extracts are shown almost same result compared to sample 1 except 15% tea dye extracts (Table 6). These fastness properties results indicate that 12% tea extracts is optimum condition for leather dyeing comparing with standard sample 1. For Mordants, sample 9 and sample 10 gave the better results.

Tensile strength properties of dyed leather samples

Tensile strength properties of leathers are depicted in Table 7. Tea extracts are shown comparable results with commercial mimosa and Quebracho tannin according to tensile strength properties considering tear strength, tensile strength and elongation at break (%) of leathers. Among them, 12% tea extracts of leather dyeing show the best result for tensile strength properties comparing with standard sample 1. For Mordants, sample 7 and sample 8 gave the better results.

Table 7. Tensile strength properties of leather

Leather Sample No.	Tensile strength (N/mm^2)	Elongation at break (%)
1.	13.74 ± 0.41	37.19 ± 0.24
2.	24.11 ± 0.34	40.05 ± 0.21
3.	13.83 ± 0.18	35.51 ± 0.27
4.	18.75 ± 0.25	36.10 ± 0.61
5.	16.85 ± 0.19	42.32 ± 0.14
6.	19.80 ± 0.26	33.59 ± 0.12
7.	15.28 ± 0.31	25.91 ± 0.17
8.	13.94 ± 0.27	36.19 ± 0.08
9.	10.40 ± 0.30	38.19 ± 0.09
10.	12.78 ± 0.22	26.46 ± 0.15

TGA of dyed leather

The TGA thermograms of four distinct samples, sample 1, sample 3, sample 4, and sample 5 throughout a temperature range of 30°C to 600°C are shown in Figure 3, reflects the change of mass over temperature increment.

Table 6. Color fastness tests (Grey scale rating)

Leather Sample No.	Dry rub fastness	Wet rub fastness	Perspiration fastness	Light fastness
1.	4/5	4/5	4/5	4/5
2.	4/5	4/5	3/4	4/5
3.	4/5	4/5	3/4	4/5
4.	4/5	4/5	3/4	4/5
5.	4/5	3/4	3/4	4/5
6.	4/5	4	4	4/5
7.	4/5	4	3/4	4/5
8.	4/5	4	3/4	4/5
9.	4/5	4	4	4/5
10.	4/5	4	4	4/5

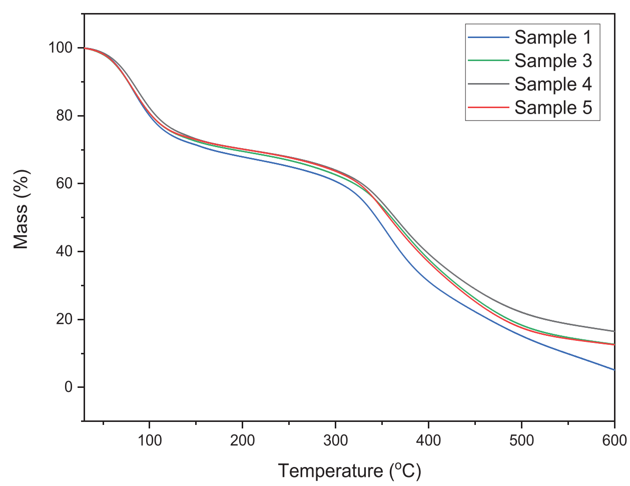

Fig. 3. TGA of tea dyeing of chrome tanned goat leather

Below 150°C, all samples first show a modest mass loss, which is the result of physically adsorbed water and volatile components evaporating. The thermal breakdown of organic components is indicated by a more significant mass loss that occurs when the temperature rises above this threshold, usually between 200°C and 400°C.

While sample 4 (12% dye extract) keeps the most residual mass at higher temperatures, suggesting greater thermal resistance, sample 1 exhibits the largest overall mass loss, indicating inferior thermal stability. The decomposition behaviors of the intermediate samples 3 and sample 5 show modest stability.

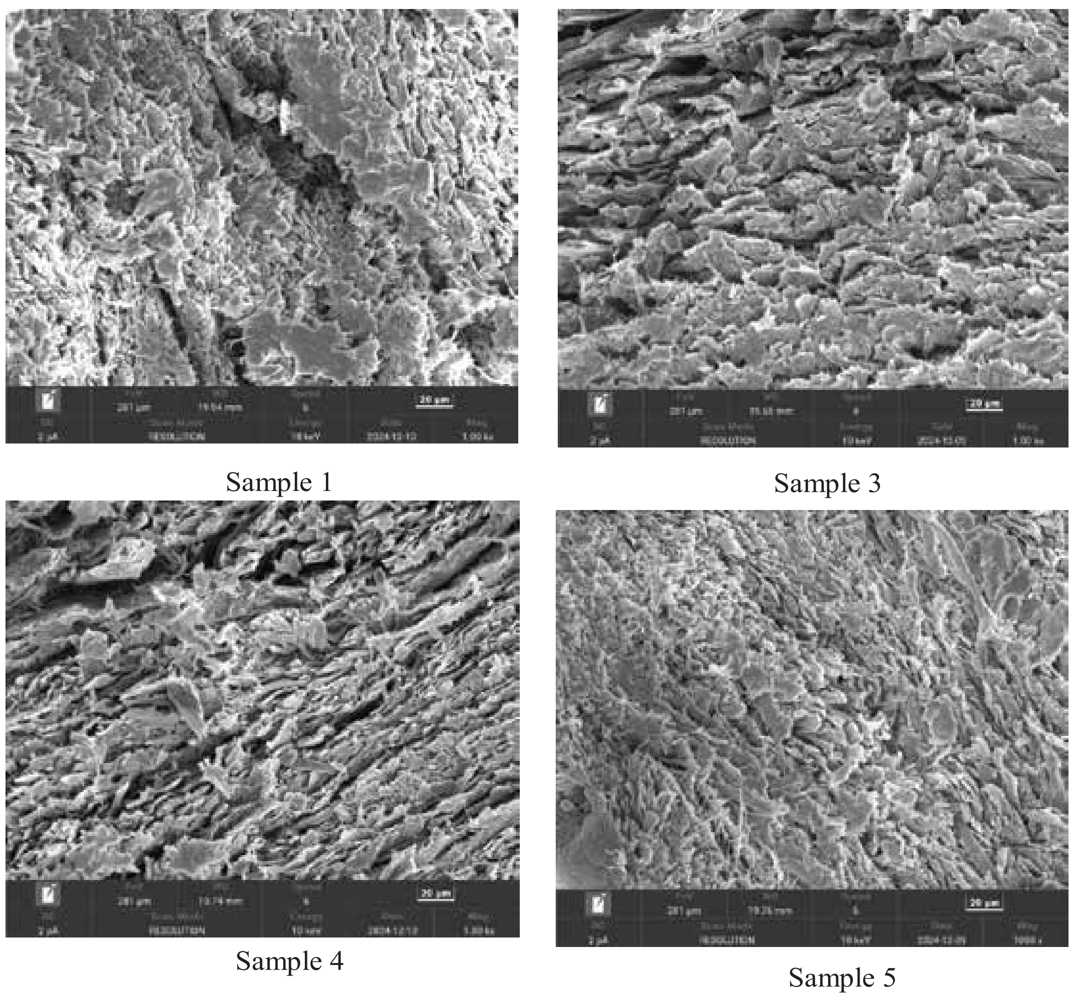

Fig. 4. SEM images of tea dyeing of chrome tanned goat leather

SEM Analysis of dyed leather

SEM images of dyed leather are shown in Figure 4. Sample 1 (control) represents with visible fibril detachment and irregular gaps corresponding to undyed leather. While sample 4 (12% dye extracts) shows a relatively compact and intact fibrous collagen matrix, with distinct fiber bundles oriented parallel to the surface. The fibers are well aligned and closely packed, indicating that the highest dye penetration may have occurred without major disruption to the collagen network. Sample 3 and Sample 5 shows slightly open fibrous structure compared to Sample 4, which could be attributed to the slightly less absorption of dye molecules.

Conclusions

Dried tea leaves aqueous extracts of various concentrations with or without mordants had been used as dyeing agents for color enhancement of chrome tanned goat leathers. Among them inorganic mordants especially 1% CuSO4. $5H_2O$ with 15% tea dye extract gave the highest result for color enhancement of chrome tanned goat leathers. However, using natural mordants had increased the positive value of Da* but decreased the negative value of DL* which indicated that the color shade going to more reddish color but color intensity decreasing. The dye exhaustion was decreasing with increasing concentration of tea extracts after 12%. However, the color intensities of chrome tanned goat leathers was slightly increased with increasing concentration of tea extracts up to 15%. The conditions for pH and temperature for tea dyeing were 6.8 and 30^{o}C, respectively. Tensile strength properties were increased with increasing dye concentration up to 12%, after that decreasing trend was shown. Tensile strength properties measurements may be an indication that those extracts can be used as tanning materials as well as dyeing materials at retanning stage at certain concentration. Therefore, further study will be needed for those processes also. Comparing to color values, physical and thermal strength, and microscopic image, 12% tea dye extract applied leather was best without mordant usage. In addition, 15% tea dye extract with 1% copper (II) sulphate mordant showed better dyeing qualities comparing other treatment with mordants. Hence, it can be concluded that aqueous tea extracts dyeing can be another option for environment friendly dyeing and producing value added products which have more aesthetic values.

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