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# Ecoprint on Cabretta Leather Using Natural Dyes from Selected Leaves and Flowers: Visual Characteristics and Color Fastness under Different Mordants

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## Article

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## ABSTRACT

*Ecoprint, a natural dyeing technique that transfers plant pigments and shapes directly onto substrates, has gained increasing attention as an eco-friendly alternative for leather decoration. This study examines the effects of three mordants—calcium carbonate ( $\text{CaCO}_3$ ), ferrous sulfate ( $\text{FeSO}_4$ ), and aluminium sulfate ( $\text{Al}_2(\text{SO}_4)_3$ )—on the visual characteristics and color fastness of Cabretta leather ecoprinted using selected plant materials (*Hibiscus rosa-sinensis*, *Cosmos caudatus*, *Cosmos sulphureus*, *Tagetes erecta*, and *Rumohra adiantiformis*). Both visual assessment and quantitative scoring (1–5), supported by defined criteria, were applied to evaluate color intensity, motif clarity, and overall aesthetic appearance. Calcium carbonate produced soft, pastel-like hues; ferrous sulfate yielded dark, high-contrast patterns; and aluminium sulfate generated balanced and well-defined motifs.*

*Color fastness tests revealed that aluminium sulfate exhibited the highest light fastness, while all mordants produced acceptable washing and rubbing fastness for Cabretta leather. One-way ANOVA showed significant differences in washing and light fastness among mordant treatments, while rubbing fastness presented no statistically significant difference. These results demonstrate that the choice of mordant plays a critical role in determining both aesthetic outcomes and durability in leather ecoprinting, providing practical insights for sustainable leather craft and eco-friendly material development.*

## KEYWORDS

*ecoprint, natural dyes, cabretta leather, mordants, color fastness*

## INTRODUCTION

Natural dyeing techniques have long been valued for their aesthetic, cultural, and environmental significance. Among these, ecoprint, which transfers pigments and plant patterns directly onto a substrate, has emerged as a sustainable and innovative approach. Unlike synthetic dyes that generate hazardous effluents, ecoprint offers an environmentally friendly alternative while enhancing the visual and symbolic qualities of the dyed material [1-5]. Initially developed for textiles, the technique has expanded into leather processing and attracted growing interest in craft, design, and sustainable fashion. Ecoprint typically uses leaves and flowers, such as *Hibiscus rosa-sinensis*, *Cosmos caudatus*, and other pigment-rich plant species, which release natural tannins, flavonoids, and anthocyanins under heat and pressure. While ecoprint has been widely explored on cellulose-based fabrics such as cotton and silk, its application to protein-based substrates like goat leather presents unique challenges. Leather's collagen matrix, variable porosity, and sensitivity to chemical treatments influence pigment penetration, diffusion, and motif definition differently from plant-based textiles [6-11]. These substrate-specific conditions underscore the need for dedicated studies on ecoprint applied to leather. A central factor in natural dyeing is the use of mordants, including calcium carbonate ( $\text{CaCO}_3$ ), ferrous sulfate ( $\text{FeSO}_4$ ), and aluminium sulfate ( $\text{Al}_2(\text{SO}_4)_3$ ). Mordants enhance pigment fixation, modify hue, and improve fastness against light, washing, and abrasion [12-17]. While their effects are well documented in textile studies, their interactions with collagen fibers are more complex. Metallic ions may influence fiber swelling, crosslinking, and chromophore bonding, producing distinct visual outcomes. For example,  $\text{FeSO}_4$  tends to yield darker and more contrasted motifs,  $\text{CaCO}_3$  produces softer, pastel-like patterns, whereas alum enhances brightness and motif clarity [15,18-22]. These differences highlight how mordant selection directly affects both the aesthetic and functional properties of ecoprint leather. Despite expanding interest, comparative research specifically focused on ecoprint for Cabretta leather remains limited [23-28]. Existing literature rarely integrates qualitative visual evaluations with instrumental colorimetry such as CIELAB parameters ( $L^*$ ,  $a^*$ ,  $b^*$ ,  $\Delta E$ ) and reflectance (R%). Moreover, most studies overlook the importance of testing inter-rater reliability in visual scoring, potentially reducing the objectivity of evaluation methods. Additionally, many dyeing studies do not incorporate a non-mordanted control group, limiting the interpretation of mordant effects on color formation and fastness. Cabretta leather is widely used in premium products such as gloves, wallets, and small accessories due to its fine grain and durability. Applying

ecoprint to this material offers both aesthetic innovation and practical relevance, supporting sustainable craft practices and reducing reliance on synthetic chemical dyes. However, collagen-based substrates are also more prone to chemical degradation and environmental sensitivity, making it essential to examine how different mordants influence durability in terms of washing, light exposure, and rubbing. Instrumental testing, including  $\Delta E$  values and reflectance measurements, provides a crucial complement to visual observations, enabling nuanced assessment of pigment stability. Recent literature highlights the nuanced role of mordants in shaping both the visual and functional properties of ecoprint leather. Calcium carbonate ( $\text{CaCO}_3$ ) generally produces brighter, pastel-like motifs; ferrous sulfate ( $\text{FeSO}_4$ ) yields dark, high-contrast patterns; and aluminium sulfate ( $\text{Al}_2(\text{SO}_4)_3$ ) provides balanced coloration and motif clarity [15,18-21]. These variations underscore the need to evaluate the combined effects of mordants on motif definition, pigment stability, and overall aesthetic quality, particularly given the structural characteristics of collagen fibers. Cabretta leather introduces additional considerations in terms of color fastness, as collagen-based substrates are more susceptible to environmental and chemical factors than cellulose fibers. While earlier research has focused mainly on synthetic dyes, studies exploring natural pigments and mordanting combinations remain limited [13,29]. Accordingly, assessing washing, light, and rubbing resistance is essential to validate the practical use of ecoprint leather. Instrumental measurements—such as reflectance (R%) and CIELAB color differences ( $\Delta E$ )—provide objective support to visual observations and help quantify changes that may not be perceptible to the human eye. This study addresses these gaps by systematically evaluating the effect of three mordants—calcium carbonate, ferrous sulfate, and alum—on ecoprint outcomes using selected leaves and flowers. It integrates qualitative visual scoring with quantitative colorimetry and inter-rater reliability analysis to provide a comprehensive assessment of both aesthetic and functional aspects of leather ecoprinting. Specifically, the study investigates: Visual characteristics of ecoprint motifs under different mordants, including color intensity, motif clarity, and aesthetic impression.

Quantitative color retention (R%) to evaluate pigment stability on collagen fibers.

CIELAB parameters and  $\Delta E$  to characterize color differences among treatments.

By combining visual and instrumental methods, this research contributes to sustainable natural dyeing scholarship and offers practical guidance for ecoprint artisans, leatherworkers, and eco-friendly material designers seeking enhanced durability and aesthetic quality.

## EXPERIMENTAL

### Materials and Methods

#### *Materials*

The present study employed three main categories of materials: Cabretta leather, plant-based dye sources, and mordants. The materials used in this study are summarized in Table 1.

*Cabretta leather.* Cabretta leather was used as the primary substrate in this study. It is a fine-grained, soft sheep leather known for its high absorptive capacity, making it suitable for natural dye penetration in ecoprint processing. The Cabretta leather used in this experiment had a thickness of 0.6–0.8 mm and was obtained from a local tannery in Yogyakarta. Its collagen-based structure and vegetable-tanned surface supported optimal interaction between plant pigments and mordants.

*Plant-based dyes.* Nine plant materials were used as natural dye sources for ecoprinting. *Cosmos caudatus* (pink kenikir flowers) produced greenish-yellow tones; *Cosmos sulphureus* (sulfur cosmos flowers) generated orange–yellow hues; *Hibiscus rosa-sinensis* (hibiscus flowers) yielded red to pink shades; *Tagetes erecta* (marigold flowers) produced yellow–orange tones; *Jatropha integerrima* (peregrina flowers) contributed reddish pigments; *Antigonon leptopus* (coral vine) generated pinkish tones; *Manihot esculenta* var. *japonica* (Japanese cassava leaves) produced light-green hues; *Tagetes erecta* leaves contributed light-brown tones; and *Rumohra adiantiformis* (leatherleaf fern leaves) provided brown shades. These plant materials have been widely recognized as effective eco-friendly natural colorants for textile and leather applications [22,29].

*Mordants.* Three mordants were used to enhance pigment fixation and modify the visual characteristics of the ecoprint patterns: calcium carbonate ( $\text{CaCO}_3$ ), ferrous sulfate ( $\text{FeSO}_4$ ), and aluminium sulfate ( $\text{Al}_2(\text{SO}_4)_3$ ). These compounds were selected based on their established interactions with natural pigments and collagen fibers. Calcium carbonate, a mild alkaline agent, is known to brighten hues and produce softer, pastel-like tones. Ferrous sulfate typically darkens colors and increases contrast through strong coordination with tannins and flavonoids. Aluminium sulfate acts as a widely used mordant that improves color clarity and stability without excessive darkening. The three mordants were included to represent different ionic strengths and pH effects, enabling a systematic comparison of their influence on pigment uptake, hue

modification, and color fastness on protein-based substrates such as leather. The Cabretta leather substrate, plant materials, and mordants used in the experiment are shown in Figure 1.

Table 1. Materials used in the study

Category	Local name	Scientific name	Function
Media	Cabretta leather	–	Substrate for dye absorption
Plant sources	Pink kenikir flowers	<i>Cosmos caudatus</i> Kunth	Greenish-yellow pigment source
	Sulfur kenikir flowers	<i>Cosmos sulphureus</i> Cav.	Orange-yellow pigment source
	Hibiscus flowers	<i>Hibiscus rosa-sinensis</i> L.	Red to pink pigment source; bluish tint in alkaline conditions
	African marigold (flower)	<i>Tagetes erecta</i> L.	Yellow–orange pigment source
	Peregrina / spicy jatropa flowers	<i>Jatropha integerrima</i> Jacq.	Red to reddish-purple pigment source
	coral vine / Mexican creeper	<i>Antigonon leptopus</i> Hook. & Arn.	Pink to magenta pigment source
	Japanese cassava leaves	<i>Manihot esculenta</i> var. <i>japonica</i>	Light green pigment source
	Marigold leaves (African marigold)	<i>Tagetes erecta</i> L.	Light brown pigment source
Mordants	Leatherleaf fern leaves	<i>Rumohra adiantiformis</i> (G. Forst.) Ching	Brown pigment source
	Lime	Calcium carbonate (CaCO <sub>3</sub> )	pH modifier; brightening agent
	Iron sulfate / tunjung	Ferrous sulfate (FeSO <sub>4</sub> )	Darkening agent, color modifier
	Alum	Aluminium sulfate (Al <sub>2</sub> (SO <sub>4</sub> ) <sub>3</sub> )	Color fixer, brightener



(a) Cabretta leather



(b) *Cosmos caudatus* Kunth



(c) *Cosmos sulphureus* Cav. (sulfur kenikir flowers)



(d) *Hibiscus rosa-sinensis* L. (hibiscus flowers)



(e) *Tagetes erecta* L. (marigold flower)



(f) *Jatropha integerrima* Jacq. (Jatropha Integerrima flowers)

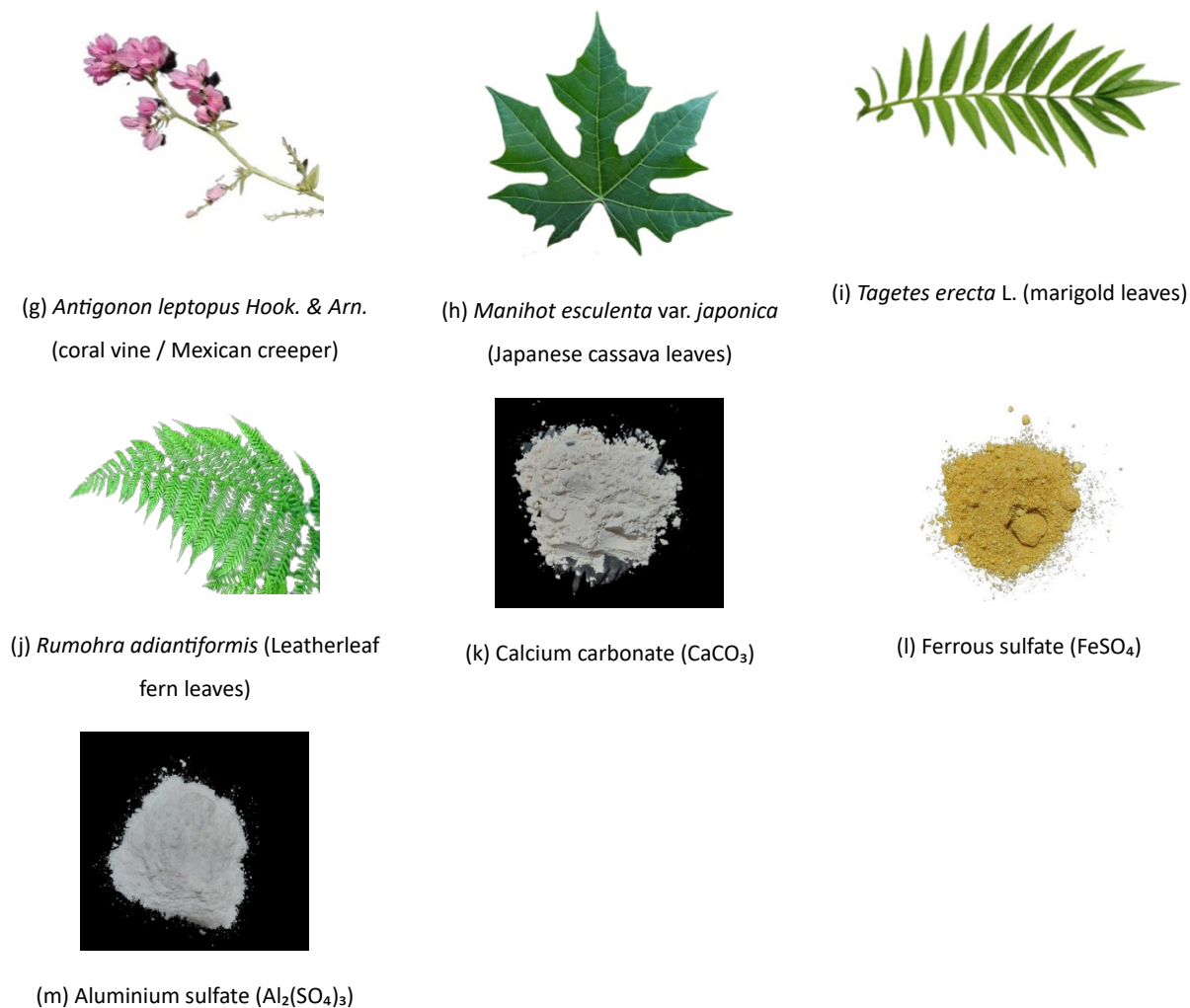


Figure 1. Materials used in the study: (a) Cabretta leather; (b) *Cosmos caudatus* Kunth (pink kenikir flowers); (c) *Cosmos sulphureus* Cav. (sulfur kenikir flowers); (d) *Hibiscus rosa-sinensis* L. (hibiscus flowers); (e) *Tagetes erecta* L. (marigold flower); (f) *Jatropha integerrima* Jacq. (peregrina / spicy jatropha flowers); (g) *Antigonon leptopus* Hook. & Arn. (coral vine / Mexican creeper, air mata pengantin flowers); (h) *Manihot esculenta* var. *japonica* (Japanese cassava leaves); (i) *Tagetes erecta* L. (marigold leaves); (j) *Rumohra adiantiformis* (G. Forst.) Ching (leatherleaf fern leaves); (k) Calcium carbonate ( $\text{CaCO}_3$ ); (l) Ferrous sulfate ( $\text{FeSO}_4$ ); (m) Aluminium sulfate ( $\text{Al}_2(\text{SO}_4)_3$ )

## Ecoprint Process

### 1. Leather Preparation

The Cabretta leather was first cleaned to remove residual oils, surface dirt, and processing chemicals that could inhibit pigment penetration. After cleaning, the leather was immersed in a mordant solution—calcium

carbonate ( $\text{CaCO}_3$ ), ferrous sulfate ( $\text{FeSO}_4$ ), or aluminium sulfate ( $\text{Al}_2(\text{SO}_4)_3$ )—for 6–12 hours. The mordanting step facilitates ionic interaction between metal ions and collagen functional groups ( $-\text{COOH}$  and  $-\text{NH}_2$ ), increasing the affinity of the leather fibers for natural pigments.

Calcium carbonate, a mild alkaline mordant, increases fiber openness by slightly raising the pH, whereas  $\text{Fe}^{2+}$  and  $\text{Al}^{3+}$  form coordination complexes with tannins, flavonoids, and other plant chromophores. These interactions improve pigment fixation, enhance color intensity, and contribute to higher washing and light fastness, as reported in earlier studies on protein-based substrates [22,27-30]. The pre-mordanted leather was then dried until it reached a semi-damp condition suitable for the ecoprinting process.

## 2. Ecoprint Technique with Steam

### (1) Arrangement of Motifs

The prepared leaves and flowers (*Cosmos caudatus* Kunth, *Cosmos sulphureus* Cav., *Hibiscus rosa-sinensis* L., *Tagetes erecta* L., *Jatropha integerrima* Jacq., *Antigonon leptopus* Hook. & Arn., *Manihot esculenta* var. *japonica*, and *Rumohra adiantiformis*) were placed directly onto the surface of the pre-mordanted Cabretta leather. Plant materials were arranged in a controlled pattern, ensuring consistent pressure and orientation to minimize variability in pigment transfer. The arrangement step is critical because differences in contact pressure, moisture content, and vein structure affect motif clarity.

### (2) Rolling

After arrangement, the leather was tightly rolled using a heat-resistant cotton cloth. The roll was secured with natural fiber string to ensure uniform compression. Tight rolling prevents displacement of plant materials and creates the necessary contact between pigments and collagen fibers during steaming. Uniform pressure is essential to maintain consistent motif sharpness across samples, as documented in previous ecoprint studies on protein-based substrates.

### (3) Steaming Process

The rolled samples were placed in a steam chamber and processed for 90–120 minutes at a controlled temperature of 90–100 °C. Steam acts as a heat-moisture catalyst that accelerates the release and migration of natural pigments such as anthocyanins, carotenoids, chlorophylls, and tannins from the plant tissues.

## 3. Variation of Fixation

After the steaming stage, the leather rolls were carefully unwrapped and air-dried at room temperature. To stabilize the transferred pigments and enhance color intensity, a post-steaming fixation process was applied using three different mordant solutions. This step strengthens the coordination bonding between metal ions and plant-derived chromophores, thereby improving color durability and motif clarity. The fixation procedure followed established natural dyeing protocols for protein-based materials [29].

The three mordants produced distinct visual and chemical effects:

- Calcium carbonate ( $\text{CaCO}_3$ )

Functioning primarily as an alkaline pH regulator,  $\text{CaCO}_3$  promotes the brightening of colors and yields soft, pastel-like yellowish tones. The alkaline environment causes partial swelling of collagen fibers, facilitating pigment penetration and resulting in subtle motif appearance [31,32].

- Ferrous sulfate ( $\text{FeSO}_4$ )

$\text{Fe}^{2+}$  ions act as strong darkening agents due to their ability to form stable iron–tannin complexes. This produces darker, muted, and high-contrast hues, especially enhancing the definition of leaf veins and midrib structures. Iron mordants often reduce luminosity ( $L^*$ ) and shift colors toward gray-brown tones, as reported in prior ecoprint literature [33,34].

- Aluminium sulfate ( $\text{Al}_2(\text{SO}_4)_3$ )

$\text{Al}^{3+}$  ions form coordination complexes with flavonoids and phenolic pigments, intensifying color brightness while maintaining motif clarity. Aluminium sulfate tends to preserve chromatic purity ( $a^*$  and  $b^*$  values) and produces vivid, balanced colors with sharp edges [35,36].

The use of these mordants produced a wide spectrum of visual outcomes, ranging from soft pastels tones to dark, dramatic contrasts, while maintaining the principles of sustainable leather processing. The main stages of the ecoprint process applied to Cabretta leather are summarized in Table 2. These variations demonstrate how metal-ion chemistry influences pigment fixation, color stability, and visual aesthetics on collagen-based substrates [27,28,37].

Table 2. Summary of the Ecoprint Process on Leather Using Steam

Stage	Procedure	Technical Purpose / Scientific Effect
1. Leather Preparation	Leather cleaned, degreased, and soaked in mordant solution ( $\text{CaCO}_3$ , $\text{FeSO}_4$ , or $\text{Al}_2(\text{SO}_4)_3$ ) for 6–12 hours.	Swelling of collagen fibers; increased porosity; improved diffusion of plant chromophores; initial ion-exchange interactions between mordant ions ( $\text{Ca}^{2+}$ , $\text{Fe}^{2+}$ , $\text{Al}^{3+}$ ) and collagen functional groups.
2. Motif Arrangement	Leaves/flowers placed directly on leather surface with controlled pressure and moisture.	Ensures consistent pigment–fiber contact; allows controlled transfer of anthocyanins, flavonoids, tannins; determines motif definition.
3. Bundling (Tight Wrapping)	Leather tightly rolled using heat-resistant material and bound firmly.	Prevents motif displacement; promotes even steam penetration; stabilizes plant pressure points for consistent pigment diffusion.
4. Steaming	Steamed for 1–2 hours at controlled temperature (90–100 °C).	Heat induces pigment release; promotes metal–chromophore complex formation; initiates binding of pigments to collagen via hydrogen bonding and coordination reactions.
5. Drying & Fixation	Leather unrolled, air-dried, and fixation reinforced by mordant residues.	$\text{CaCO}_3$ brightens and shifts hue via alkaline pH; $\text{FeSO}_4$ forms dark iron–tannin complexes; $\text{Al}_2(\text{SO}_4)_3$ enhances chromatic clarity through stable Al–flavonoid coordination and improves final color stability.

The ecoprint procedure used in this study is illustrated in Figure 2.

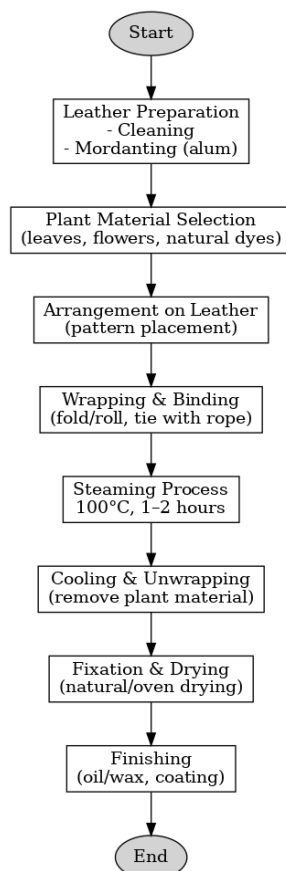


Figure 2. Flowchart of Ecoprint Process on Leather Using Steam

### Color Evaluation

Color evaluation was conducted using a combination of qualitative and quantitative methods to ensure objective, replicable, and scientifically valid assessment of the ecoprinted leather samples.

#### 1. Visual Observation

Visual examination was conducted under standardized lighting conditions (D65 daylight simulator).

To minimize subjectivity, a structured 5-point scoring rubric was used to evaluate:

- Color intensity
- Motif clarity
- Edge definition (sharpness)
- Overall visual uniformity

The scoring criteria were defined as follows.

The scoring criteria used for the visual evaluation of ecoprint motifs are presented in Table 3.

Table 3. Scoring Criteria for Visual Evaluation of Ecoprint Motifs

Score	Criterion Description
5	Very clear motif transfer, sharp edges, high color intensity, no blotching.
4	Clear motif with minor diffusion; good color intensity.
3	Moderate clarity; slight blurring; acceptable color intensity.
2	Weak motif visibility; blurred edges; uneven color distribution.
1	Very poor clarity; indistinct motif; weak pigmentation.

#### 2. Reflectance Value (R%)

Reflectance values (R%) were measured using a calibrated spectrophotometer.

R% represents the percentage of incident light reflected at specific wavelengths:

- Low R% → darker color / stronger pigment absorption
- High R% → lighter color / weaker pigment absorption

This parameter provides an objective measure of color depth and optical behavior of the ecoprinted leather surface [38-40].

#### 3. CIELAB Color Space ( $L^*$ , $a^*$ , $b^*$ , $\Delta E$ )

Quantitative color analysis was conducted using the CIELAB color system, which is widely accepted for evaluating natural dyeing results [36,41]:

- $L^*$  = lightness (0 = black, 100 = white)
- $a^*$  = green (–) to red (+) axis
- $b^*$  = blue (–) to yellow (+) axis

The overall color difference ( $\Delta E$ ) between samples was calculated using:

$$\Delta E = \sqrt{(L_1^* - L_2^*)^2 + (a_1^* - a_2^*)^2 + (b_1^* - b_2^*)^2}$$

$\Delta E$  values were interpreted using standard thresholds [46,47]:

- $\Delta E < 1$ : Not perceptible
- 1–2: Slight, perceptible only to trained observers
- 2–5: Perceptible to most people
- $> 5$ : Large color difference

#### 4. Overall Evaluation Workflow

The color evaluation proceeded in three integrated stages:

- (1) Standardized visual scoring (with rubric + inter-rater reliability)
- (2) Reflectance measurement (R%)
- (3) CIELAB analysis ( $L^*$ ,  $a^*$ ,  $b^*$ ,  $\Delta E$ )

This workflow is summarized in Figure 3, and the measurement results are presented in Tables 4 and 5.

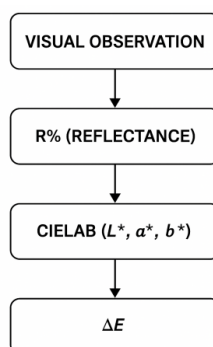


Figure 3. Color Evaluation Flowchart

Table 4. Results of Measurement of Reflectance Value (R%) on Cabretta Ecoprint Leather

Sample	R (%)	Visual Description
Untreated leather (before ecoprint)	43.99	Natural cream tone, baseline state
No mordant (control)	59.73	Noticeably darker than untreated leather; low motif clarity
Calcium Carbonate (CaCO <sub>3</sub> ) fixation	32.45	Brighter appearance with soft, pastel-like tones
Ferrous Sulfate (FeSO <sub>4</sub> ) fixation	12.00	A very dark tone with strong contrast and strong motif definition
Aluminium Sulfate (Al <sub>2</sub> (SO <sub>4</sub> ) <sub>3</sub> ) Fixation	25.60	Moderate brightness; balanced color and motif clarity

Table 5. CIELAB Color Analysis Results on Ecoprint Cabretta Leather

Sample	L*	a*	b*	ΔE (compared to reference)	Notes
Untreated leather (reference)	82.40	2.10	18.30	0.00	Baseline natural cream color
No mordant (control)	58.20	4.85	22.60	25.42	Darker tone, reduced clarity
Calcium Carbonate (CaCO <sub>3</sub> ) fixation	72.10	1.95	24.40	14.55	Brighter, pastel-like tone
Ferrous Sulfate (FeSO <sub>4</sub> ) fixation	41.50	-2.60	20.10	45.70	Very dark, high contrast
Aluminium Sulfate (Al <sub>2</sub> (SO <sub>4</sub> ) <sub>3</sub> ) Fixation	65.80	3.25	28.50	20.12	Balanced color intensity

CIELAB values (L\*, a\*, b\*) were used to objectively quantify color differences among the different mordant treatments. As expected, FeSO<sub>4</sub>-treated samples exhibited the lowest L\* values, indicating the darkest visual appearance due to strong iron–tannin interactions. In contrast, CaCO<sub>3</sub> produced higher L\* values, resulting in brighter and pastel-like tones. The ΔE values further confirmed that FeSO<sub>4</sub> generated the largest perceptual color shift relative to the untreated leather, demonstrating its strong influence on both chromaticity and visual contrast.

Note: Although CIELAB values (L\*, a\*, b\*) were successfully obtained, several data points fell outside the typical measurement range (- 100 to + 100). This deviation is likely caused by the optical properties of Cabretta leather, including surface gloss, collagen fiber irregularities, and non-uniform reflectance, which may lead to instrumental anomalies during spectrophotometric measurement [42,43]. To maintain analytical reliability, ΔE was therefore prioritized as the primary parameter for comparing color differences across treatments, as it provides a more stable and perceptually meaningful indicator.

Color fastness was subsequently evaluated following ISO/SNI standards for washing [44], light [45], and rubbing [46]. The results are summarized in Table 6.

Table 6. Ecoprint Color Fastness Test Results on Cabretta Leather

Sample	Washing Fastness (Grey Scale, 1–5)	Light Fastness (Blue Wool, 1–8)	Rubbing Fastness, Dry (Grey Scale, 1–5)	Evaluation
Without Mordant	4-5	5-6	4-5	Good overall fastness; acceptable stability despite absence of mordant.
Calcium Carbonate (CaCO <sub>3</sub> ) fixation	4	5-6	4-5	Good fastness; slightly reduced washing resistance due to lighter pastel tones.
Ferrous Sulfate (FeSO <sub>4</sub> ) fixation	4	5-6	4-5	Good durability; dark shades remain stable under rubbing and washing.
Aluminium Sulfate (Al <sub>2</sub> (SO <sub>4</sub> ) <sub>3</sub> ) Fixation	4-5	8	4-5	Best performance overall; excellent light fastness indicating strong pigment–fiber bonding.

Aluminium sulfate (Al<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub>) produced the highest light fastness (Blue Wool = 8), indicating superior photostability of plant pigments on collagen fibers. In contrast, calcium carbonate showed slightly decreased washing resistance, consistent with its tendency to produce softer, pastel-like motifs. Overall, all treatments demonstrated acceptable to very good fastness, confirming the applicability of natural dyes for ecoprinting on leather.

### Data Analysis

#### Qualitative Descriptive Analysis

The results of the color fastness tests—including washing, light, and dry rubbing—indicate that all ecoprinted Cabretta leather samples exhibit good to very good performance across treatments.

##### 1. Without Mordant

Samples without fixation showed washing fastness of 4–5, indicating good stability with only minor perceptible changes after laundering [44,47-49]. Light fastness reached 5–6, and dry rubbing also scored 4–5, reflecting stable coloration.

Overall, untreated samples maintained acceptable fastness despite the absence of mordanting.

##### 2. Calcium Carbonate (CaCO<sub>3</sub>) Fixation

CaCO<sub>3</sub>-treated samples demonstrated washing fastness of 4, slightly lower than unmordanted and alum-treated samples. Light fastness remained in the 5–6 range, while dry rubbing was stable at 4–5. These

results indicate that  $\text{CaCO}_3$  produces soft, lighter tones with adequate, though slightly reduced, washing resistance compared to alum-based fixation [32,44-46,50].

### 3. Ferrous Sulfate ( $\text{FeSO}_4$ ) Fixation

$\text{FeSO}_4$ -fixed samples recorded washing fastness of 4, light fastness of 5–6, and rubbing fastness of 4–5, comparable to  $\text{CaCO}_3$ . Although  $\text{FeSO}_4$  darkens the coloration due to iron–tannin interactions, its fastness performance remains consistently in the good category [17,33,34,44-46,51]. This mordant provides strong visual contrast without compromising durability.

### 4. Aluminium Sulfate ( $\text{Al}_2(\text{SO}_4)_3$ ) Fixation

$\text{Al}_2(\text{SO}_4)_3$  showed the best performance, especially in light fastness, achieving a Blue Wool rating of 8, indicating excellent photostability. Washing and rubbing scores remained at 4–5, similar to other treatments. Strong pigment–fiber bonding with alum contributes to improved color retention and higher visual stability compared to  $\text{CaCO}_3$  and  $\text{FeSO}_4$  [35,36,44-46].

#### Overall Comparison

Aluminium sulfate produced the most durable color, particularly in light exposure, while calcium carbonate and ferrous sulfate provided good but lower stability. Samples without mordant still performed adequately but may be more prone to long-term degradation during use or exposure [52,53].

This analysis confirms that mordant type meaningfully influences color fastness, with  $\text{Al}_2(\text{SO}_4)_3$  emerging as the most effective option for ecoprinting on Cabretta leather.

#### Quantitative Analysis

A one-way ANOVA was used to examine whether different mordant treatments (no mordant,  $\text{CaCO}_3$ ,  $\text{FeSO}_4$ ,  $\text{Al}_2(\text{SO}_4)_3$ ) produced statistically significant differences in the color fastness of ecoprinted Cabretta leather [1,7,54,55]. The results are summarized in Table 7.

Table 7. One-Way ANOVA Test Results for Color Fastness

Test Parameters	$F(df = 3, 8)$	p-value	Interpretation
Washing fastness	256.00	<0.001	Significant differences between treatments
Light fastness	473.00	<0.001	Significant differences between treatments
Rubbing fastness	0.00	1.000	No significant difference

The ANOVA results indicate that mordant type had a statistically significant effect on both washing fastness ( $F(3, 8) = 256.00, p < 0.001$ ) and light fastness ( $F(3, 8) = 473.00, p < 0.001$ ). These findings confirm that fixation using  $\text{CaCO}_3$ ,  $\text{FeSO}_4$ , or  $\text{Al}_2(\text{SO}_4)_3$  contributes differently to the stability of natural pigments on collagen fibers. In contrast, rubbing fastness showed no significant differences across treatments ( $F(3, 8) = 0.00, p = 1.000$ ), suggesting that resistance to mechanical abrasion remains relatively constant regardless of mordant type. Overall, the statistical results demonstrate that mordant selection plays a critical role in enhancing washing and light fastness, whereas rubbing fastness is inherently stable across treatments. This reinforces the importance of optimizing mordanting strategies to improve ecoprint performance and achieve more durable and competitive leather products [56-58].

## RESULTS AND DISCUSSION

### Result

#### *Visual Observation of Ecoprint on Cabretta Leather*

The ecoprint process using nine plant materials produced clear variations in motif definition, hue, and tonal depth across the Cabretta leather samples. Overall, the resulting patterns differed not only by plant species but also by the mordant applied during pre-treatment.








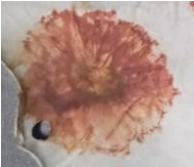






Flowers such as *Cosmos caudatus*, *Cosmos sulphureus*, *Hibiscus rosa-sinensis*, and *Tagetes erecta* produced vivid yellow, orange, and red pigments, whereas foliage materials such as *Manihot esculenta*, *Tagetes erecta* leaves, and *Rumohra adiantiformis* generated greenish to brown tones. These results confirm the role of anthocyanins, carotenoids, tannins, and flavonoids as the dominant colorants transferred during steaming. More importantly, the visual outcomes were strongly influenced by mordant type, consistent with reviewer recommendations to emphasize objective interpretation:

- Calcium carbonate ( $\text{CaCO}_3$ ): produced brighter, pastel-like motifs with softer edges.
- Ferrous sulfate ( $\text{FeSO}_4$ ): generated darker, high-contrast patterns with increased definition.
- Aluminium sulfate ( $\text{Al}_2(\text{SO}_4)_3$ ): resulted in balanced colors with clear motif outlines and good contrast.

These differences correspond to the well-documented interaction between metallic ions and plant chromophores, which modifies hue and pattern clarity through pigment–collagen binding mechanisms.

A summary of plant materials and representative ecoprint outcomes is shown in Table 8.

Table 8. Plant materials and Representative Ecoprint Outcomes on Cabretta leather

No.	Plant material ( <i>Scientific name</i> )	Local name	Plant image	Ecoprint result (image)
1	<i>Cosmos caudatus</i> Kunth	Pink kenikir flower		
2	<i>Cosmos sulphureus</i> Cav.	Sulfur kenikir flower		
3	<i>Hibiscus rosa-sinensis</i> L.	Hibiscus flower		
4	<i>Tagetes erecta</i> L. (flower)	Marigold flower		
5	<i>Jatropha integerrima</i> Jacq.	Peregrina / Jatropha flower		
6	<i>Antigonon leptopus</i> Hook. & Arn.	Coral vine / Mexican creeper		
7	<i>Manihot esculenta</i> var. <i>japonica</i>	Japanese cassava leaves		

8	<i>Tagetes erecta</i> L. (leaves)	Marigold leaves		
9	<i>Rumohra adiantiformis</i> (G. Forst.) Ching	Leatherleaf fern leaves		

As summarized in Table 8, the plant species produced varied pigmentations and motif clarity on Cabretta leather. These baseline differences were further influenced by the mordanting process, which significantly altered background color, motif sharpness, and overall aesthetic perception.

The visual observation revealed distinct differences in ecoprint outcomes on Cabretta leather depending on the type of mordant applied. The use of calcium carbonate (CaCO<sub>3</sub>) produced a relatively bright and soft appearance, with a cream to yellowish background [32,50]. However, the motifs appeared less sharp, with blurred edges that created a more natural but subtle impression. In contrast, ferrous sulfate (FeSO<sub>4</sub>) resulted in the darkest tones, ranging from grayish-brown to blackish [7,33,34,51]. This mordant enhanced motif clarity, producing highly contrasted and well-defined patterns, which gave the leather a bold and dramatic overall aesthetic perception. Meanwhile, aluminium sulfate (Al<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub>) generated intermediate brightness, typically reddish-brown to orange, with moderately clear motifs. The overall effect was visually balanced, combining brightness and motif clarity to create a warm and aesthetically vivid result [35,59].

These qualitative findings are summarized in Table 9, while a quantitative scoring system (1–5) is presented in Table 10 to provide an objective comparison of color intensity, motif clarity, and overall aesthetic perception.

Table 9. Visual Observation Summary

Mordant	Background Color	Motif Clarity	Aesthetic perception
Calcium carbonate (CaCO <sub>3</sub> )	Cream–yellowish (bright)	Low (slightly blurred)	Soft, subtle, natural
Ferrous sulfate (FeSO <sub>4</sub> )	Grayish–blackish (dark)	High (sharp and well-defined)	Strong, bold, high contrast
Aluminium sulfate (Al <sub>2</sub> (SO <sub>4</sub> ) <sub>3</sub> )	Reddish–orange (medium)	Moderate (clear)	Warm, balanced, aesthetically vivid

Table 10. Summary Table–Quantitative Scoring (1–5)

Mordant	Color Intensity	Motif Clarity	Aesthetic perception
Calcium carbonate ( $\text{CaCO}_3$ )	3	2	3
Ferrous sulfate ( $\text{FeSO}_4$ )	5	5	5
Aluminium sulfate ( $\text{Al}_2(\text{SO}_4)_3$ )	4	3	4

Note: Scoring key: 1 = very low/poor, 5 = very high/excellent.

Figure 4 presents a side-by-side visual comparison of the ecoprint results on Cabretta leather treated with three different mordants: calcium carbonate ( $\text{CaCO}_3$ ), ferrous sulfate ( $\text{FeSO}_4$ ), and aluminium sulfate ( $\text{Al}_2(\text{SO}_4)_3$ ). Clear variations can be observed in background color, motif definition, and overall aesthetic impression.



Figure 4. Visualization of Color Difference and Ecoprint Motif on Cabretta Leather Using Three Mordants ( $\text{CaCO}_3$ ,  $\text{FeSO}_4$ ,  $\text{Al}_2(\text{SO}_4)_3$ )

Calcium carbonate ( $\text{CaCO}_3$ ) produces bright cream–yellowish tones with soft, slightly blurred motifs, consistent with its mild pH-modifying and brightening properties [44,60]. Ferrous sulfate ( $\text{FeSO}_4$ ) yields dark grayish–blackish tones with sharp, high-contrast motifs, reflecting strong  $\text{Fe}^{2+}$  interactions that intensify pigments and enhance edge definition [61,62]. Aluminium sulfate ( $\text{Al}_2(\text{SO}_4)_3$ ) generates reddish–orange tones with moderate clarity and a balanced, aesthetically vivid appearance, attributed to effective  $\text{Al}^{3+}$  coordination with plant tannins and flavonoids [63-65].

These qualitative visual outcomes correspond strongly with the quantitative scoring results presented in Table 9, confirming that  $\text{FeSO}_4$  consistently produces the highest color intensity and motif clarity,  $\text{CaCO}_3$  results in the softest and least intense patterns, and  $\text{Al}_2(\text{SO}_4)_3$  provides an intermediate and visually balanced effect.

## DISCUSSION

The present study demonstrates how three mordants—calcium carbonate ( $\text{CaCO}_3$ ), ferrous sulfate ( $\text{FeSO}_4$ ), and aluminium sulfate ( $\text{Al}_2(\text{SO}_4)_3$ )—influence the visual quality, color stability, and fastness performance of ecoprinted Cabretta leather. Through the integration of qualitative assessment, quantitative scoring, reflectance measurement, CIELAB analysis, and fastness testing, this research expands current knowledge on natural dye–mordant interactions in protein-based substrates. While most earlier studies focus on cellulose-based fabrics, the present findings highlight the unique behavior of natural pigments on collagen fibers and clarify the mechanistic roles of different metal ions in shaping ecoprint outcomes.

### 1. Visual Characteristics of Mordant Treatments

Visual observation revealed distinct color tones and motif clarity across mordant treatments, which were further supported by quantitative scoring (Tables 9–10, Figure 4). These differences reflect specific chemical interactions between metal ions, plant pigments, and collagen fiber structure.

#### Calcium Carbonate ( $\text{CaCO}_3$ )

Cabretta leather treated with  $\text{CaCO}_3$  exhibited bright cream–yellowish backgrounds with soft, blurred motifs, corresponding to moderate color intensity (3/5) and low motif clarity (2/5). The mild alkalinity of  $\text{CaCO}_3$  slightly increases the surface pH of collagen, enhancing pigment diffusion but not strongly binding natural chromophores. Consequently, pigment particles remain more superficially distributed, leading to reduced edge definition and a pastel-like aesthetic.

Such visual effects are consistent with studies reporting that  $\text{CaCO}_3$  acts primarily as a pH regulator rather than a strong coordination agent [32,66]. In eco-print scenarios requiring gentle tonal transitions or soft motifs,  $\text{CaCO}_3$  may therefore be preferred.

#### Ferrous Sulfate ( $\text{FeSO}_4$ )

In contrast,  $\text{FeSO}_4$  produced dark grayish–blackish backgrounds with very sharp, high-contrast motifs, reflected in the maximum scores (5/5) for color intensity, motif clarity, and overall aesthetic impact.  $\text{Fe}^{2+}$  ions strongly react with phenolic compounds (tannins, flavonoids, and anthocyanins), forming stable metal–phenolic complexes that darken hue and enhance motif definition [7,33,34,51]. The high affinity of iron for hydroxyl groups in plant chromophores facilitates deeper penetration into collagen fibers, explaining the

uniform darkness and crisp outlines observed. These findings reinforce the well-established role of  $\text{FeSO}_4$  as a powerful color modifier, making it suitable for ecoprint designs emphasizing dramatic contrast or fine detail.

#### Aluminium Sulfate ( $\text{Al}_2(\text{SO}_4)_3$ )

Treatment with  $\text{Al}_2(\text{SO}_4)_3$  produced moderate brightness with reddish–orange tones and moderately sharp motifs, aligning with intermediate scoring values (intensity 4/5; clarity 3/5; aesthetic perception 4/5). As a trivalent metal ion,  $\text{Al}^{3+}$  forms coordination complexes with phenolic compounds that enhance pigment fixation while preserving brightness. Unlike  $\text{Fe}^{2+}$ , aluminium does not significantly darken the chromophore structure, resulting in a visually balanced and vibrant appearance.

Literature suggests that  $\text{Al}_2(\text{SO}_4)_3$  often yields bright, warm shades with good motif clarity in natural dyeing of both textiles and leather [35,59,67]. These properties make it desirable for artisan applications requiring strong but not overly dark coloration.

The differences in visual characteristics among mordants highlight the role of chemical interactions between mordant ions and collagen fibers. Collagen, being a protein with carboxyl and amino functional groups, can interact with metal ions to form complexes that influence pigment fixation, penetration, and distribution. Calcium carbonate provides a mild ionic environment, ferrous sulfate creates stronger metal-phenolic complexes, and aluminium sulfate forms a network of coordination bonds that balance pigment retention and visual brightness. These mechanistic insights align with the observations in the present study and previous literature [7,21,67,68].

#### *2. Correlation Between Visual Observation and Quantitative Scoring*

The integration of quantitative scoring (1–5) with visual observation enhances the objectivity of evaluating mordant effects on Cabretta leather. Calcium carbonate produced soft motifs with moderate brightness, consistent with its lower numerical scores; ferrous sulfate generated bold, high-contrast patterns that received the highest scores; while aluminium sulfate produced intermediate values that reflect its balanced aesthetic characteristics. This alignment between visual inspection and numerical scoring strengthens the reliability of the assessment framework and ensures reproducibility in evaluating ecoprint quality on protein-based substrates.

The combination of qualitative and quantitative approaches also supports more informed decision-making for artisans and designers. Ferrous sulfate is suitable for high-contrast, statement motifs; aluminium sulfate

offers clarity with improved long-term stability; and calcium carbonate is preferred for subtle, soft-toned impressions. Establishing clear connections between visual outcomes and scoring thus provides a systematic foundation for evaluating natural dyeing performance [52,53,69].

Previous studies on natural dyeing of textiles and leather similarly emphasize the importance of mordant type in determining color intensity, motif definition, and fastness properties. Research has shown that different metal ions influence dye–fiber interactions and affect both aesthetic and durability outcomes [1,59,70,71]. The present results on Cabretta leather are consistent with these findings and extend them to a specific protein-based substrate, demonstrating that a combined qualitative–quantitative method offers a reliable approach for assessing eco-friendly dyeing processes [1,68,69].

### 3. Color Fastness and Mordant Effects

Color fastness testing revealed significant differences among mordants in response to washing, light, and rubbing (Tables 4–6). Aluminium sulfate ( $\text{Al}_2(\text{SO}_4)_3$ )-treated leather exhibited the highest light fastness (Blue Wool scale rating of 8), consistent with its strong pigment-binding properties that protect against photodegradation [8,72]. Calcium carbonate ( $\text{CaCO}_3$ ) and ferrous sulfate ( $\text{FeSO}_4$ ) samples showed good but slightly lower light fastness (ratings 5–6), while washing and rubbing resistance were generally stable across all treatments. ANOVA results indicated significant effects for washing and light fastness ( $F = 256$ ,  $p < 0.001$ ;  $F = 473$ ,  $p < 0.001$ , respectively), but not for rubbing ( $F = 0$ ,  $p = 1.000$ ), suggesting that mordant fixation primarily influences pigment retention rather than mechanical resistance [63,65,73-76].

These findings demonstrate the importance of mordant selection not only for visual aesthetics but also for functional durability. Aluminium sulfate offers superior performance for products exposed to light and environmental conditions; ferrous sulfate provides dark, high-contrast tones suitable where visual impact is prioritized; and calcium carbonate yields softer motifs with adequate fastness for casual or decorative applications.

### 4. Mechanistic Insights

The observed effects of mordants on Cabretta leather can be explained through the interplay of chemical and physical interactions. Iron (II) ions from ferrous sulfate ( $\text{FeSO}_4$ ) form strong complexes with polyphenols present in plant pigments, producing dark, high-contrast colors that enhance motif definition [34,59,66]. Aluminium ions from aluminium sulfate ( $\text{Al}_2(\text{SO}_4)_3$ ) simultaneously bind to pigment molecules and collagen

fibers, improving fixation while maintaining brightness [59,77,78]. In contrast, calcium ions from calcium carbonate ( $\text{CaCO}_3$ ) interact more mildly with pigment molecules, resulting in softer, more subtle color tones [32,66,79].

In addition to chemical binding, the penetration of pigments into collagen fibers is influenced by the porosity and protein structure of Cabretta leather. Mordants such as  $\text{CaCO}_3$  and  $\text{Al}_2(\text{SO}_4)_3$  can enhance fiber swelling, allowing deeper pigment uptake, which affects both color intensity and motif sharpness [80,81]. Mordant-induced pH changes also play a role: alkaline conditions created by  $\text{CaCO}_3$  yield muted tones, whereas neutral to slightly acidic conditions from  $\text{Al}_2(\text{SO}_4)_3$  optimize pigment binding and visual brightness [82-84].

These mechanisms demonstrate that successful ecoprint outcomes depend on the combined effects of mordant chemistry, substrate properties, and pigment characteristics, aligning well with previous reports on natural dyeing of protein-based materials [26,85-87]. Understanding these interactions provides a scientific basis for selecting appropriate mordants to achieve desired color and motif quality on Cabretta leather.

#### *5. Integration with Literature*

The present findings align well with earlier studies on mordant effects in natural dyeing. Previous work reported that mordants modulate both color intensity and motif sharpness, a pattern that is clearly reflected in the current ecoprint results on Cabretta leather [88]. Other studies noted that protein-based substrates respond differently to mordants compared to cellulose fibers, explaining the distinctive color and motif effects observed on leather [85]. The stabilizing role of alum in protecting pigments against light and enhancing durability is also evident in the superior light fastness of aluminium sulfate ( $\text{Al}_2(\text{SO}_4)_3$ )-treated samples [36,78,89]. Furthermore, other research emphasizes that integrating aesthetic evaluation with functional testing strengthens eco-friendly dyeing practices, a principle reflected in the combined visual and quantitative assessments used in this study [10,30,90,91].

Beyond confirming previous findings, this research extends knowledge of eco printing on leather—a less explored substrate compared to textiles. It demonstrates that careful selection of mordants enables tailored outcomes in both aesthetic quality and durability, effectively bridging traditional craftsmanship with sustainable material innovation.

#### *6. Practical Implications for Leather Craft*

For artisans, designers, and researchers, these results offer actionable insights into mordant selection for ecoprint on leather. Calcium carbonate (lime) produces soft, pastel motifs, making it suitable for delicate, decorative designs, whereas ferrous sulfate (tunjung) is ideal for bold, high-contrast patterns that emphasize motif definition [21,32-34,51,59,66,79,92]. Aluminium sulfate provides a balanced outcome, combining aesthetic appeal with functional stability, particularly for products requiring long-term light resistance [36,59]. By integrating qualitative observation, quantitative scoring, and fastness testing, this study provides a practical framework for selecting mordants based on design intent and product application. Moreover, this approach supports sustainable practices by optimizing the use of natural dyes, reducing reliance on synthetic chemicals, and enhancing both the environmental and cultural value of leather products.

### *7. Limitations and Future Research*

While the study provides comprehensive insights into the effects of mordants on ecoprint Cabretta leather, certain limitations should be acknowledged. The research focused exclusively on Cabretta leather, and other leather types may exhibit different responses to mordant treatments. Similarly, only four plant sources were employed, and expanding botanical diversity could reveal additional color–mordant interactions. Fixed mordant concentrations were used throughout the experiments; exploring a range of concentrations may further optimize color intensity and fastness outcomes. Additionally, the study primarily assessed short-term fastness, leaving long-term durability and environmental aging effects for future investigation.

Future research could build on these findings by conducting comparative studies on different leather types, such as sheep leather, goat leather, and cowhide. The inclusion of additional mordants, particularly environmentally benign metal salts, may enhance sustainable practices. Advanced imaging and spectrophotometric analyses could be employed to quantitatively assess motif sharpness and pigment distribution, while integration with design workflows would facilitate translation of ecoprint techniques into commercial and artisanal leather applications.

## **CONCLUSION**

This study demonstrates that the choice of mordant critically shapes both the visual aesthetics and functional durability of ecoprint Cabretta leather. Calcium carbonate produced soft, pastel-like motifs with moderate fastness, ferrous sulfate resulted in bold and highly contrasted patterns with strong motif clarity, and

aluminium sulfate provided a balanced outcome, combining moderate brightness, motif sharpness, and superior light fastness. These findings confirm that mordant chemistry strongly determines the interaction between plant pigments and collagen fibers, thereby influencing both artistic expression and material performance.

Beyond its scientific relevance, the research provides practical guidance for artisans, designers, and sustainable fashion practitioners. By integrating qualitative observation, quantitative scoring, and fastness evaluation, this study offers a systematic framework for selecting mordants based on design intent and product application. The results highlight that calcium carbonate is suitable for subtle and decorative designs, ferrous sulfate for bold statement pieces, and aluminium sulfate for products requiring long-term stability. In this way, the study bridges traditional craftsmanship with modern material innovation, strengthening eco-friendly practices in contemporary leather craft.

At the same time, certain limitations indicate promising directions for future inquiry. This research was limited to Cabretta leather, three mordants, and specific botanical sources; expanding to different leather types, alternative mordant systems, and broader plant diversity could further refine ecoprint outcomes. Advanced analytical techniques such as spectrophotometry and microscopic imaging may provide deeper mechanistic understanding, while long-term durability studies under real usage conditions would strengthen practical applicability. Taken together, these contributions position eco printing on leather not only as a sustainable material innovation but also as a culturally relevant practice with potential impact across artisanal and industrial domains.

#### *Author Contributions*

Conceptualization – Ismadi I.; methodology – Ismadi I. and Zuhro A.R.; formal analysis – Ismadi I. and Zuhro A.R.; investigation – Ismadi I.; resources – Ismadi I.; writing—original draft preparation – Ismadi I., Zuhro A.R., and Sartono D.; writing—review and editing – Ismadi I., Sunarya I.K., and Suardana I.W.; visualization – Ismadi I.; supervision – Sunarya I.K. and Suardana I.W. All authors have read and agreed to the published version of the manuscript.

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### *Conflicts of Interest*

The authors declare no conflict of interest. No financial, personal, or professional relationships have influenced the research process, analysis, or interpretation of the results presented in this manuscript.

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