

# APPLICATION OF KERMES OAK (*QUERCUS COCCIFERA*) IN LEATHER DYEING: A NATURAL AND SUSTAINABLE APPROACH

### COBAN Semiha Sultan<sup>1</sup>, ORK EFENDIOGLU Nilay<sup>1</sup>, MUTLU Mehmet Mete<sup>1</sup>

<sup>1</sup>Ege University, Engineering Faculty, Leather Engineering Department, 35100, Izmir, Turkiye

Corresponding author: Mutlu, Mehmet Mete, E-mail: mete.mutlu@ege.edu.tr

Abstract: Leather, as an organic material, has been used and continuously developed since ancient times, maintaining its value through various dyeing and finishing techniques. Although the widespread use of synthetic dyes led to a decline in the use of natural dyes, recent concerns about environmental impact and health hazards have renewed interest in natural alternatives. This study explores the dyeing potential of kermes oak (Quercus coccifera), a species of the Fagaceae family native to the Mediterranean region of Anatolia, as a sustainable natural dye source for leather. The effects of dyeing temperature (60°C, 80°C, and 100°C) and mordant concentration (0.5%, 1%, and 1.5%) on the color and color strength (K/S) of leather were systematically investigated. The most intense and uniform coloration was achieved at 100°C with a 1.5% mordant ratio. The results highlight the effectiveness of kermes oak in producing aesthetically pleasing and environmentally friendly dyeing results. Overall, this study demonstrates the potential of natural dyes to serve as viable, eco-conscious alternatives to synthetic dyes in leather production, contributing to sustainable practices in the leather industry.

Key words: Leather, dyeing, kermes oak, Quercus coccifera, natural dye, coloration

## 1. INTRODUCTION

Until the mid-nineteenth century, natural dyes represented the sole source of coloration for textiles and garments across the globe [1]. These dyes were not only utilized in textile applications but also extensively used in the dyeing of leather and furs, as evidenced by various studies [2], [3]. These practices show how natural dyes served as essential agents for coloration in multiple materials until synthetic dyes were introduced.

Natural dyes are renewable, biodegradable, and non-toxic, making them a promising alternative for reducing the environmental impact of synthetic dyes [4], [5]. However, despite these ecological benefits, natural dyes often exhibit limitations in terms of color vibrancy, shade diversity, and durability, particularly concerning fastness to light, washing, and rubbing. To address these challenges, the use of mordants—substances that facilitate dye fixation on fibers and enhance fastness properties—plays a crucial role [6].

For centuries, one of the primary plant sources of natural dyes used in leather dyeing has been the Kermes oak (*Quercus coccifera*). It belongs to *Quercus* genus (Fagaceae), which comprises over 200 species, in the temperature areas of Northern hemisphere and tropical mountains and found in Southern Europe from Portugal to Turkey [7]. Previous researches demonstrated that Kermes oak can be effectively used as a natural dye for coloring textile fabrics [8] and wool [9].



Despite the extensive use of Kermes oak as a natural dye in textile applications, no studies have been identified in the literature regarding its potential for leather dyeing. Given the growing demand for sustainable and eco-friendly alternatives in the leather industry, this study aims to explore the feasibility of using Kermes oak as a natural dye for leather. Furthermore, the research examines the influence of key dyeing parameters, specifically temperature and mordant ratio, on color uniformity and strenght. By addressing these aspects, the study seeks to contribute to the development of environmentally sustainable dyeing methods for leather.

## 2. EXPERIMENTAL

## 2.1 Material

Twelve crust sheep leathers from the Metis breed were used in this study. The root woods of *Quercus coccifera* (Kermes oak) (Figure 1.a) were collected from the forest region of Hatay, located in the southern part of Turkey (humid climate, 36°45′ N, 36°26′ E) during November 2024. The harvested woods were air-dried at room temperature and subsequently ground into sawdust (Figure 1.b). The experimental design can be seen in Table 1.



Fig. 1: The woods (a) and sawdust (b) of kermes oak

Table 1: The layout of the experimental design						
Code	Mordan Ratio (%)	Temp. (°C)				
A1	non-mordant	60				
A2	non-mordant	80				
A3	non-mordant	100				
B1	0.5	60				
B2	0.5	80				
B3	0.5	100				
C1	1	60				
C2	1	80				
C3	1	100				
D1	1.5	60				
D2	1.5	80				
D3	1.5	100				



Copper sulfate, obtained in technical grade from BOR-KIM Chemicals, was used as a mordant due to its minimal influence on altering the inherent color of the leather during the dyeing process. As a fatliquoring agent, a high lightfastness, electrolyte-stable, and colorless sulfone oil from TFL was employed.

Color measurements were conducted using a Konica Minolta CM-3600d spectrophotometer under a CIE D65 light source and a  $10^{\circ}$  standard observer angle.

#### 2.1 Methods

#### 2.1.2 Extraction Process and Extraction Yield

Dried *Quercus coccifera* (Kermes oak) sawdust (40 g) were weighed and boiled in 1 liter of water at 90°C for 1 hour to initiate extraction. Following the boiling process, the mixture was allowed to stand at room temperature for 24 hours to enhance the extraction efficiency. Afterward, the solution was filtered to obtain the extract. The resulting extract was transferred into a glass dish and dried in a hot-air oven at  $100 \pm 2$ °C until complete evaporation of the water content, ensuring the residue reached a constant weight. The total solid content was determined by weighing the remaining residue in the dish. The extraction yield was calculated using the following equation (1):

Evaporation Residue 
$$\left(\frac{mg}{L}\right) = (A - B) * 1000/V$$
 (1)

A = Total weight of the evaporation dish and solid residue (mg)

B = Weight of the empty evaporation dish (mg)

V = Volume of the sample (mL)

#### 2.1.3 Leather dyeing

The initial weights of the crust leathers were recorded prior to processing. The recipe applied to the crust leathers is presented in Table 2. The temperatures (60, 80, and 100  $^{\circ}$ C) and mordant concentrations (0.5, 1, and 1.5%) used in the process are specified in the recipe.

Process	Amount (%)	ChemicalsTemp. (°C)		Time (min.)
Rewetting	100	Water	50	
	0.5	Wetting agent (Nonionic)		60
Washing	100	Water	45	
Dyeing	60	Water	60/80/100	
	2	Dye auxiliary material (Ethoxylated fatty amine sulphate)		15
	5	Dye (Kermes oak extract)		60
	0.5/1/1.5	Mordant (Copper Sulfate)		45
	3	Fatliquor agent (Combination of synthetic fatliquors and esters)		40
	2	НСООН		30 (10*3)
Washing	100	Water	30	45 (3*15 min)

Table 2: Dyeing recipe

#### 2.1.4 Determination of color measurement

A Konica Minolta CM-3600d spectrophotometer was used to quantitatively assess the color differences of leathers dyed using different ratio of mordants and temperatures. Color measurements were conducted in accordance with the CIE  $L^*$ ,  $a^*$ ,  $b^*$  color system. In this system, the  $L^*$  value



represents the lightness of the color, ranging from 0 (black) to 100 (white). The  $a^*$  value indicates the position of the color on the red-green axis, where negative values correspond to green and positive values to red. Similarly, the  $b^*$  value denotes the position on the blue-yellow axis, with negative values indicating blue and positive values indicating yellow.

To ensure accuracy and consistency, color measurements were taken from five distinct points on the leather surface using the standard measurement area of the device, and the mean of these readings was calculated.

Additionally, the color strength (K/S) values were calculated at the maximum absorption wavelength ( $\lambda$ max = 400 nm) using the Kubelka–Munk equation (2). K is the scattering coefficient, S is the absorption coefficient, and R is the reflectance. R is the decimal fraction of the reflectance of dyed leather, R = 1.0 at 100% reflectance. This approach allows for an objective evaluation of the dyeing efficiency and color performance of the processed leathers.

 $K/S = (1 - R)^2 / 2R$ 

3. RESULTS AND DISCUSSIONS

#### 3.1. Extraction Yield

A total of 1000 mL of kermes oak extract was obtained through aqueous boiling, and the extraction yield was determined to be 14.57% based on the dry matter content of the plant material.

#### **3.2.** Color Measurement Findings

The color measurement results for the leathers are provided in Table 3. According to the CIE Lab color system, the  $L^*$  values range from 60.82 to 70.41, indicating a general trend towards lighter shades, especially in the A and B group samples. Samples A1 and A2 have the highest  $L^*$  values (70.17 and 70.41, respectively), suggesting they are the lightest among the group. In contrast, sample D3 shows the lowest  $L^*$  value (60.82), reflecting a relatively darker appearance.

The  $a^*$  values, which represent the red-green axis, ranged from 1.09 to 6.47 across the dyed samples and increase progressively from group A to group D. A1 and A2 have values close to 1, whereas C3 and D3 reach over 6. Given that undyed crust leather typically exhibits low  $a^*$  values due to its slightly greenish undertone, the observed increase after dyeing suggests a clear shift toward the red axis. This trend is consistent with the inherent reddish hue of kermes oak extract, which enhanced the red component in the leather, contributing to a warmer and more vibrant appearance.

Since undyed crust leather typically displays a slightly bluish-green undertone, the increase in  $b^*$  values observed in all dyed samples — ranging from 12.42 to 17.32 — indicates a noticeable shift toward the yellow axis after dyeing. This shift reflects the influence of the reddish hue of kermes oak extract, which effectively counteracted the cool base tone of the crust leather.

Regarding the K/S values, which are associated with the color depth or saturation, a notable increase is observed from A1 (1.16) to D3 (2.57). This gradual enhancement suggests that the leather samples in later groups (especially D3 and C3) have more intense and deeper coloration, potentially due to higher dye uptake, which is likely influenced by the increased mordant ratio and temperature during the dyeing process.

As observed in the pseudo colors, the dyed leathers exhibit increasingly darker shades with rising dyeing temperatures. This visual change corresponds with higher K/S values, confirming enhanced dye uptake. The results highlight the significant influence of temperature on the aesthetic outcome, demonstrating the potential of kermes oak to produce rich and appealing tones through controlled dyeing conditions.

(2)



Code	$L^*$	<i>a</i> *	b*	K/S	Pseudo Color
A1	70.17	1.09	13.84	1.16	
A2	70.41	1.15	14.04	1.23	
A3	68.13	2.90	14.50	1.45	
B1	70.03	2.04	12.52	1.17	
B2	68.85	3.29	14.91	1.32	
B3	64.94	5.14	16.62	1.77	
C1	70.09	2.32	12.75	1.17	
C2	66.92	3.55	14.98	1.61	
C3	62.51	6.47	17.32	2.18	
D1	69.54	1.17	12.42	1.18	
D2	65.72	4.03	15.69	1.78	
D3	60.82	6.47	17.01	2.57	

Table 3: The color measurement results

Figure 2 illustrates the effect of mordant concentration (0-1.5%) on the K/S values at three different dyeing temperatures (60 °C, 80 °C, and 100 °C). The results indicate that increasing the temperature significantly enhances the interaction between the dye and the collagen fibers, leading to higher color strength values. At 60 °C, K/S values remain nearly constant regardless of the mordant ratio, suggesting limited dye uptake due to insufficient thermal activation. At 80 °C, a moderate increase in K/S values is observed as the mordant ratio increases, particularly beyond 1%, indicating improved dye fixation. The most pronounced effect is seen at 100 °C, where the K/S values rise sharply with increasing mordant ratios, reflecting enhanced dye affinity and deeper shade formation. These findings demonstrate that higher dyeing temperatures, in combination with appropriate mordant ratio, facilitate better dye-fiber interactions and result in more intense coloration.

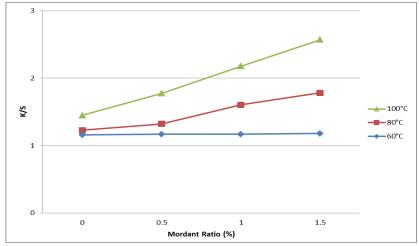


Fig. 2: The effect of mordant ratio and dyeing temperature on K/S of leather dyed with kermes oak



#### 4. CONCLUSIONS

This study demonstrated the potential of kermes oak as a sustainable and natural dye source for leather dyeing. The results indicated that variations in mordant concentration and dyeing temperature significantly affected the color tone and uniformity of the dyed leathers. Higher mordant ratios and elevated temperatures generally enhanced color depth, suggesting improved dye-fiber interaction under these conditions. Moreover, the leathers dyed with kermes oak exhibited visually appealing and homogeneous coloration, highlighting the applicability of this natural dye in aestheticdriven leather applications.

In addition to its technical performance, the use of kermes oak contributes to environmentally responsible leather production by reducing dependence on synthetic dyes and petrochemical-based auxiliaries, which are often associated with ecological toxicity and wastewater pollution. As a locally available, plant-based resource, kermes oak represents a low-impact alternative that aligns with circular economy principles and supports the transition toward greener manufacturing practices in the leather industry.

These findings provide a valuable basis for the development of environmentally friendly dyeing techniques in the leather industry. Future studies may focus on optimizing process parameters for industrial scalability, investigating the long-term stability of the dyed leather, and evaluating the use of kermes oak in combination with other natural mordants or auxiliaries. This research contributes to the advancement of sustainable leather processing practices by promoting the use of bio-based, locally available resources.

#### REFERENCES

[1] D. Cardon, "*Natural dyes: Sources, tradition, technology and science*", London: Archetype Publications Ltd, 2007.

[2] S. C. Lee, E. C. Shin, and W. J. Kim, "Dyeing properties of natural leather using red natural dyes", Journal Society of Leather Technologists and Chemists, vol. 98 (6), pp. 252–258, 2014.

[3] A. E. Musa, B. Madhan, W. Madhulatha, and S. Sayeed, "*Coloring of leather using henna natural alternative material for dyeing*", Journal American Leather Chemists Association, vol. 104 (104), pp. 183–190, 2009.

[4] M. Odero, A. Kiprop, I. K'owino, M. Arimi, B. Maiyo, "Optimization of dyeing process parameters for dyeing of cotton fabric using extracts of Aloe succotrina", Annals of the University of Oradea Fascicle of Textiles, Leatherwork, vol. 21 (2), pp. 63-68, 2020.

[5] M. D.Cocîrlea, A. Coman, L.-F. Popovici, S. Oancea, D. Coman, "Potential use of biodyes for green coloring of medical textiles", Annals of the University of Oradea Fascicle of Textiles, Leatherwork, vol. 25 (2), 2024.

[6] J. Che, X. Yang, "A recent (2009–2021) perspective on sustainable color and textile coloration using natural plant resources", Heliyon, vol. 8 (10), 2022.

[7] D. Sohretoglu, A. Kuruüzüm-Uz, A. Simon, and M. Dékány, "*New secondary metabolites from Quercus coccifera L.*" Records of Natural Products, vol. 8 (4), pp. 323–329, 2014.

[8] N. Baaka, A. Mahfoudhi, and M. F. Mhenni, "Tannin-rich natural dye extracted from kermes oak (Quercus coccifera L.): Process optimization using response surface methodology (RSM)", Journal of Natural Fibers, vol. 16 (8), pp. 1209–1220, 2019.

[9] N. Baaka, "Sustainable Dyeing of wool fabric using kermes oak (Quercus Coccifera L) as source of natural colorant", Journal of Natural Fibers, vol. 19 (1), pp. 37–45, 2022.