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Coloring: The Role of Cork Oak as a Novel Bio-Mordant in Enhancing the Colorfastness and Durability of Natural Dyes.

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1. Introduction

The most common products for hair colouring are formulated with synthetic dyes. However, such products can in the long-term damage the hair structure and might cause irritant and allergic reactions. Recently, interest in the use of natural dyes has been growing, as herbal dyes are easily biodegradable, sustainable, eco-friendly, non-toxic, low-cost, easily available, and widely used in food, medicine, perfumes, and textile industries. Various parts of the plant can be used, such as the roots, leaves, twigs, stems, heartwood, bark, and wood. Moreover natural dyes may lend several functional properties, such as antibacterial, antifungal, UV protective and aromatic properties.

Despite these benefits, natural dyes often present challenges such as low dye uptake, poor fastness, uneven dyeing, difficulty in the reproduction of shades, long extraction processes, and seasonal variations [1]. To overcome the poor substantivity and fastness properties of natural dyes, textile fabrics are traditionally treated with substances called mordants.

The word “mordant” is derived from the French verb *mordre* which means “to bite” [2]. Mordants play an important role in the process of dyeing fibers with natural dyes by improving the adhesion of dyes to fibers, thereby forming long-lasting and stable color complexes. There are three mordanting methods: pre-mordanting, meta-mordanting, and post-mordanting [3]. The most commonly used mordants include various chemical salts made, particularly those based on aluminum; iron salts are often used as a post-dye treatment to darken colors; and copper salts are used either as post-mordant or a pre-mordant.

However, traditional mordants used in natural dyeing have often been associated with risks to the environment [4]. Furthermore, the treatment of transition metal mordants on the human hair may result in the accumulation of iron and copper, which may cause photooxidative damage to dyed hairs [4,5]. In response of these concerns, as a more environmentally friendly alternative to conventional metallic mordants, recent research has been exploring the use of biomordants, derived from natural sources including agro-industrial waste and by-products.

The purpose of this study was to investigate the properties of the biomordant Cork oak (*Quercus suber L.*) extract as an effective alternative to metallic mordants. To investigates the optimization of natural colorants with biomordant, three plants extracts have been used: Logwood bark (*Haematoxylum campechianum L.)*, Rhamnus Frangula (*Frangula alnus Mill.*), Red Sandalwood (*Pterocarpus santalinus L.)*. Futhermore, the plants were tested using the tradionali mordant such as tannic acid (10%), aqueous solutions of ferrous sulfate (5%) and copper sulfate (5%) or tested in synergi using mordant in combination. The plants extracts been diluted in water in different proportions and applied at an acidic pH of 5.5-6-5.

2. Materials and Methods

- *Hair tresses*: Three types of hair tresses (yak, salt and pepper, bleached human hair) with a total length of 6 cm and 0,3 g weight were used in this work.

- *Mordating Agents*:

- Tannic Acis (10%)
- Solution of Ferrous sulfate (5%)
- Solution of Copper sulfate (5%)
- Cork Oak (*Quercus suber L.*) extract (10%)

- *Plant materials and dyeing application*: Three powder plant extracts, Logwood bark (*Haematoxylum campechianum L.)*, Rhamnus Frangula (*Frangula alnus Mill.*), Red Sandalwood (*Pterocarpus santalinus L.*, and Cork Oak (*Quercus suber L.*) were used during this study, and their characteristics are tabulated in Table 1.

Each of the powdered plant extracts had been diluted in distilled water at 40-45°C in a 1:2 ratio until fine pastes were obtained. They have been tested at acidic pH levels. The pH of pastes were determined by using a digital pH meter. The three types of hair tresses (yak, salt and pepper and bleached human hair) were treated for 45'. Futhermore, the plants were tested using the tradiotional mordant such as tannic acid (10%), aqueous solutions of ferrous sulfate (5%) and aqueous solution of copper sulfate (5%) or tested in synergy by combining mordants. Finally, hair tresses were washed with water and dried with a hairdryer.

- *Application of the plant extract on bleached human hair*: Previously, to the vegetable process, the light brown hair tresses were bleached only one time with commercially available bleaching products. The mixing ratio of bleaching powder and developer was 1:2 in a non-metallic bowl. After mixing the bleaching cream, it was immediately applied with a brush to each sample. After 30 min processing time, the hair tresses were rinsed off under warm water for 2 min and then, the hair tresses were dried with a hair dryer. Afterwards, the hair tresses were dyed with extract powder plants.

The research is divided into three phases:

i. Application of the New Bio-Mordant: Initially, each natural dye was tested with tannic acid, a commonly used biomordant to improve color stability. Subsequently, tannic acid was replaced with Cork oak extract, and the results were compared to assess the effectiveness of cork as a biomordant.

ii. Combination with Traditional Metal Mordants: Cork extract was combined with traditional metal mordants like ferrous or copper sulfate. Four tests were done for each plant: two with tannic acid and a metal mordant, and two with cork extract and a metal mordant.

iii. Synergy Between Bio-Mordants: The plant extracts were also tested with a combination of tannic acid and cork extract to investigate their synergistic effects.

Table 1: Plant extracts used for hair dye applications and their characteristics

Name of the plant extract (Family)	Common name	Part used	Phytochemicals	Dyeing application at room temperature	Ref.
Haematoxylum campechianum L. (Fabaceae)	Logwood Bark	Bark	Lignans, Flavonoids, Tannins (Hematoxylin, Homoisoflavonoids)	Dilution: water:extract (1:2) pH 5.5-6-5	**[6]**
Pterocarpus santalinus L. (Fabaceae)	Red Sandalwood	Bark	Flavonoids, Tannins, Terpenoids, Phenolic compounds (Pterocarpol)	Dilution: water:extract (1:2) pH 5.5-6-5	**[7]**
Frangula alnus Mill. (Rhamnaceae)	Rhamnus Frangula	Bark	Anthraquinones, Phenolic compounds (Emodin, Frangulin)	Dilution: water:extract (1:2) pH 5.5-6-5	**[8,9]**
*Quercus suber L.**	Cork Oak	Bark	Phenolic compounds, Lignin, Tannins, Polysaccharides (Ellagitannins)	small amounts *(around 10.0%)*	**[10]**

** Cork Oak (Quercus suber L.)*

The bark of the species *Quercus suber* L., popularly known as cork, is a natural material obtained from the outer bark of an oak tree. This is an economic and ecological species in Mediterranean Basin countries. Cork oak extracts are primarily composed of aliphatic and phenolic compounds (including terpenes such as friedeline and betulinic acid), tannins (specifically ellagitannins), and phenolic acids (such as gallic, ellagic, and protocatechin). Suberin, lignin, and polysaccharides are other components of Cork Oak (*Quercus suber L.*). Suberin is the most prevalent component of Cork (about 40% m/m), exhibiting notable properties such as antioxidant, anti-inflammatory, free radical scavenging, enzyme inhibition, and antimicrobial activities. Textile dyeing is another potential field of application for Cork extracts. Furthermore, waste generated during the manufacture of Cork goods has great potential and may be applied in a variety of uses, including in cosmetic and topical treatments.

3. Results

Application of the New Bio-Mordant

Using Logwood bark (*Haematoxylum campechianum L.)* with tannic acid resulted in a brown color, while the use of Cork extract (*Quercus suber L.)* produced a more solid brown shade. Red Sandalwood (*Pterocarpus santalinus L.)* with tannic acid yielded an orange color, while Cork extract enhanced the intensity of the color. For Rhamnus Frangula (*Frangula alnus Mill.*), tannic acid produced a yellow color, whereas Cork extract resulted in a more vibrant yellow. See Figures 1 a) b) c), below. The intensity of colour by these plants were observed to be more powerful when the hair tresses were bleached in all cases.

a)

b)

c)

Figure 1: Comparative color results on hair tresses using plant extracts with tannic acid and Cork extract (a) *Haematoxylum campechianum L. (b) Pterocarpus santalinus L.(c) Frangula alnus Mill.*

Combination with Traditional Metal Mordants

Using Sandalwood with both metal mordants - a 5% ferrous sulfate solution or 5% copper sulfate solution, resulted in dark, gray-violet shades. However, replacing tannic acid with Cork extract while using metal mordants produced the same color but with more brilliance and

intensity. Longwood with metal mordants yielded gray-brown tones, while Cork extract with metal mordants resulted in brighter shades without altering the hue. For Rhamnus Frangula, metal mordants produced yellow shades, but when combined with Cork extract, a more intense yellow was achieved. All the results are reported in Figure 2. The intensity of colour by these plants were observed to be more powerful when the hair tresses were bleached in all cases.

Figure 2. Comparative color results on hair tresses dyed with Logwood (*Haematoxylum campechianum L.*) (2a, 2d), Red Sandalwood (*Pterocarpus santalinus L.*) (2b, 2e), and Rhamnus frangula (*Frangula alnus Mill.*) (2c, 2f) using different mordants. Each pair shows the effect of tannic acid versus Cork extract combined with (2a, 2b, 2c) ferrous sulfate and (2d, 2e, 2f) copper sulfate.

Synergy Between Bio-Mordants

The plant extracts were also tested in combination with two biomordants, tannic acid and cork extract, to investigate their synergistic effects. In all cases, the combination of these bio-mordants demonstrated a clear enhancement in color intensity. Logwood resulted in a deep brown, Sandalwood in a vibrant orange, and Rhamnus frangula in a vivid yellow, as shown in Figures 3 a) b) c).These findings suggest that tannic acid and cork extract work in synergy, improving both the color intensity and stability of the dyes.

a) b) c)

Figura 3. Color results on hair tresses dyed with a) Logwood (*Haematoxylum campechianum* L.), b) Red Sandalwood (*Pterocarpus santalinus* L.), and c) Rhamnus frangula (*Frangula alnus Mill.*) (*Frangula alnus* Mill.) using the combination of two bio-mordants (Tannic Acid + Cork Extract). Enhanced intensity and stability of the dyes were observed.

4. Discussion

The use of the biomordant Cork oak combined with plant extracts enhanced the fixation of natural colorants on the hair fibres by forming of some complex compounds with the dyes. These interactions are based on various types of bonds such as hydrogen, ionic or covalent bonds. Furthermore, the fixation of these biomordants-dyes complexes compounds on the hair fibres were more effectively when the hair tresses used were previously bleached. It is suggested that the sulfonic acid groups, which increased on hair surfaces after chemical bleaching, interacted with the mordant-dye complexes, leding to stronger cross-links between keratinous fibres and the colouring complexes.

The colours strength of the vegetable dyed tresses correlates with the bleaching process. The highest colour strength and intensity were obtained when hair tresses were previously bleached. It is suggested that the sulphonic acid groups, which increase after bleaching hair tresses, increase the amount of dye uptake and makes the hair fibres more accessible for colour treatments.

The experiments showed that Cork extract worked effectively with traditional metal mordants like ferrous sulfate and copper sulfate, producing positive results. This indicates that Cork extract can potentially replace metal mordants, offering similar or improved color intensity and fixation. The findings suggest that Cork extract could be a viable alternative to traditional metal mordants in natural dyeing processes.

The use of the combination of two bio-mordants, tannic acid with cork extract, demonstrated enhanced intensity and stability of the dyes. This improvement is likely due to the formation of biomordant-dye complex compounds, which interact with the functional groups of keratin

in the hair fibers. These interactions contribute to a stronger bond between the dye and the hair, resulting in more vibrant and longer-lasting color.

5. Conclusion

This study highlights the potential of Cork Oak extract as an effective and sustainable bio-mordant for natural hair dyeing. Rich in aliphatic and phenolic compounds, including terpenes (friedeline and betulinic acid), tannins (ellagitannins), and phenolic acids (gallic, ellagic, and protocatechin), Cork Oak interacts with plant pigments to form complexes that bond with the keratin in hair. These complexes improve the intensity and stability of the resulting colors, offering a promising, eco-friendly approach to enhancing the colorfastness and richness of natural hair dyes. This research contributes to the development of more sustainable and effective solutions for the natural dyeing industry, with potential applications in both cosmetic and textile industries.

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