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Enhanced Natural Dyeing and Antibacterial Properties of Cotton by Physical and Chemical Pretreatments

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Abstract: In this study, pomegranate peel extract was used to dye cotton fabrics. Generally, natural dyes have low affinity for cellulosic fibers and therefore need treatment for attachment. Air atmospheric plasma was applied to lower the concentrations of mordants needed and enhance dyeability. The outcomes showed that the residual concentrations of metallic ions decreased when applying the plasma treatment. ATR-FTIR and SEM analysis were performed to evaluate the effect of plasma modification on the surface chemistry and topography of the cellulosic fibers. The traces of metals released from finished fabric were quantified using ICP OES spectrometry. Color fastnesses (wash, rubbing and light) were evaluated to assess the mordanting step effect and the durability of the color. All tested fastnesses were good to very good. The concentrations of metal ions in finished fabrics were 32.4 for Fe, 0.28 for Al, 0.29 for Cr and 1.21 for Cu. Plasma treatment can be considered an efficient strategy to reduce the concentration of toxic metallic mordants.

Keywords: air-atmospheric plasma; mordant; natural dye; cotton; antibacterial activity

1. Introduction

The textile finishing industry is moving towards cleaner production strategies since it is under increasing social pressure to lower the use of hazardous chemicals [1], especially substances classified as carcinogenic, mutagenic and allergenic [2]. Recent studies have developed new nontoxic, sustainable, ecofriendly, and cost-effective processes [3]. New restrictions related to health hazards and environmental impacts have resulted in the replacement of hazardous substances by safer alternatives [4,5]. Recently, the use of natural dyes has been an emerging trend for textile finishing. These dyes are derived from plants, animals and minerals [6,7] and they offer the advantage of being safe, ecofriendly and sustainable [8,9]. However, it is well known that they have a very low attachment to cotton, the most used natural fiber in the world [10,11].

Textile fibers and especially cellulosic ones do not present sufficient affinity for numerous natural dyes, which are nonsubstantive [12]. Cellulosic fibers, when immersed in the dyeing bath, generate ionized hydroxyl groups characterized by a negative charge, whereas most natural dyes are anionic when immersed in water. The negative charge on the cotton fabric is responsible of the repellency between fibers and dye [13].

Cotton, a carbohydrate polymer that is made up of cellulose, is the most used fiber in day-to-day life. It is a soft polymer. Cotton fibers can hold more water than their weight and they are considered as absorbent and breathable materials.

The application of natural dyes for cotton dyeing is always associated with the application of a pretreatment to improve dye uptake and fastness [14]. The fixing substances, called mordants, contain heavy metals such as iron, copper, chromium, aluminum, cobalt or nickel [15]. After dyeing, the residual amounts of such chemicals in wastewater cause serious health and environmental problems [16]. Mordants are able to form an insoluble complex with the dye used to intensify and fix the color [17]. Colorimetric parameters such as color strength (K/S), lightness (L), blueness-greenness (b) and redness-yellowness (a) significantly vary relying on the used mordant and the mordanting method [18].

The data generated from a literature survey demonstrated significant enhancement of color strength during the dyeing step when using mordants [19].

Meena et al. reported that the color yield of cotton fabrics, dyed with pomegranate peel increased with rising mordant concentration [20]. Ibrahim et al. showed that mordants are important to fix the natural dye extracted from the plant Bisham (*Commiphora gileadensis*) [21]. Moniruzzaman et al. investigated the influence of mordant concentration on the dyeing efficiency of cotton fabric using tea extract and found that a darker shade was obtained when using a mordant concentration of 3% [22]. Mulec et al. carried out cotton dyeing with turmeric extract. The results revealed the effect of mordants in enhancing the color yield of dyed fabrics [23]. Other researchers reported that natural dyeing of cotton fabrics using metal mordants leads to a significant amelioration in color depth [15].

Generally, metallic mordants are considered as hazardous chemicals. Metal ions are known to induce organ damage. They are considered as human carcinogens according to the U.S. Environmental Protection Agency, and the International Agency for Research on Cancer [24,25]. However, the degree of danger depends on the concentration of the mordant. This is the parameter that defines the hazards it presents [15]. The amount of mordant left in wastewater or released from finished fabrics has to be considered according to the limit values of ecological standards [26].

Some strategies have been proposed to reduce the toxic effects of metal mordants while keeping their efficiency, such as using auxiliaries or pretreating textile materials by physical or chemical treatments such as UV, ultrasound and microwave [15].

Plasma is an ecofriendly initiating system to achieve cotton surface modification without changing its inner properties. It is a good alternative to the use of chemicals for the activation of textile surfaces. Plasma treatment is a technique with low environmental pollution, low maintenance costs, and is a quick and simple process with an energy and water-saving effect with high productivity [27,28]. Compared to conventional wet methods, air plasma treatment reduces the use of chemicals, energy and water. The elimination of chemicals and water is cost-effective [29]. Materials treated with plasma show changes in surface properties resulting in enhanced attachment of metals on the surface. The attachment of metal ions on the surface is an important issue. In fact, when the textile is in contact with liquids or with the skin, the metal ions leach from the surface causing undesirable effects and leading to serious consequences, especially when the leached concentrations are high [30].

During this study, we aimed to investigate an eco-friendly physical treatment using air atmospheric plasma to enhance uptake of metal ions while keeping good coloring and antibacterial properties using pomegranate peel extract, which is known as a good natural dye for textile dyeing [31–33]. By enhancing the metal ions uptake using a plasma physical treatment, their concentrations in wastewater are reduced. Indeed, the concentrations of metal ions discharged from the finished fabric must be in the safe zones to avoid human health risks. In addition, metals are known to exhibit antibacterial activities. They were applied in medicine before antibiotics were discovered. The rapid increase in resistance of bacteria, fungi and viruses has brought into focus the lack of new antimicrobial agents.

To avoid the transmission of infectious diseases, there has been a revival of interest in the application of metals as antimicrobial agents.

To our knowledge, little research has been done on air-atmospheric plasma and mordants preparation for dyeing.

2. Materials and Methods

2.1. Materials

Pomegranate peels were collected and impurities were removed by washing with water. After washing, the peels were dried at room temperature (25 ± 2 °C) for 72 h and powdered using a grinder. An aqueous extract of powdered pomegranate peel was used as a natural dye for finishing cotton fabrics. Commercially available woven cotton fabrics were used. Alum ($KAl(SO4)_2\ 12H_2O$), potassium dichromate ($K_2Cr_2O_7$), Iron sulfate hydrate ($FeSO_4\ 7H_2O$) and copper sulphate hydrate ($CuSO_4\ 5H_2O$) were kindly provided by Chimitex plus company. Analytical grade chemicals were employed for the entire experiments.

2.2. Plasma Treatment

A plasma machine Coating Star (Ahlbrandt System, Lauterbach, Hessen, Germany) was used to treat cotton fabrics before mordanting and dyeing steps. Plasma discharge was created at atmospheric pressure by a mechanism composed of three ceramic electrodes. The parameters of the plasma machine were adjusted as following: ambient air as working gas, a frequency of 30 kHz, electrical power of 1 kW, inter-electrode distance of 1.5 mm and an electrode length of 0.5 m. The power was adjusted to 1000 W and the treatment speed to 2 $m \cdot min^{-1}$. Each side of the fabric (30 cm × 30 cm) was treated twice.

2.3. Cotton Surface Characterization

To examine the effects of plasma treatment, the wettability of cotton samples was evaluated using a Drop Shape Analyzer-DSA25. The contact angle of the immediate contact with ultrapure water (2 µL) was recorded. The presented values are an average of three measurements.

To monitor the difference between cotton samples before and after plasma treatment, attenuated total reflectance Fourier-transform infrared (ATR-FTIR) spectra were recorded using a commercial ATR-FTIR attachment (Spectrum Two™ FTIR, Perkin Elmer, Waltham, Massachusetts, United States). Spectra were recorded between 4000 cm^{-1} and 400 cm^{-1}, resolution = 4 cm^{-1}, 400 scans. The peak area at 3271 cm^{-1} was calculated for the untreated and plasma treated samples using the software OriginPro 2019.

Morphological changes of different cotton samples were evaluated using a pressure-controlled scanning electron microscope (JEOL JSM5400). Cotton fabric samples (5 mm × 10 mm) were mounted on the specimen stabs and then coated with a thin film of gold using a sputtering method.

2.4. Dyeing Process

Cotton fabric dyeing was carried out using an Ahiba Datacolor machine using a bath ratio of 40:1. After adding the fabrics, the dyeing baths were heated to 100 °C and held for 45 min. The beakers of the machine were used for controlling fabrics and the dyeing bath throughout the dyeing process. Subsequently, the cotton samples were washed with water to remove residual dye and dried at room temperature.

During this study, a simultaneous mordanting method was applied (dyeing and mordanting carried out in the same bath). Three-percent mordant was used at 40 °C for 20 min before adding the natural dye (Figure 1).

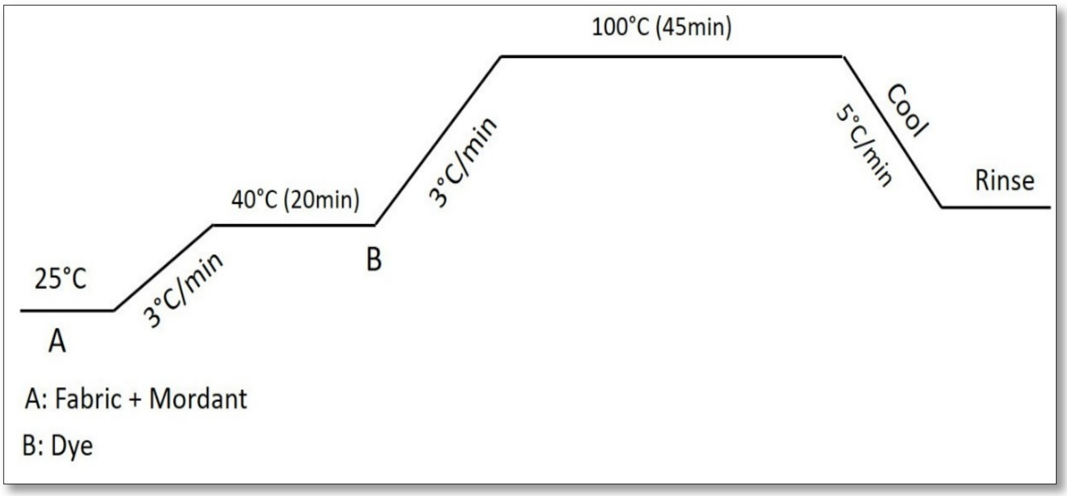


Figure 1. Simultaneous mordanting dyeing process.

2.5. Concentrations of Metal Ions in Dyed Fabrics

Before the experiments, prepared samples were dried for 24 h at 65 °C. Metal ions were extracted from the fabrics using an artificial sweat solution prepared according to the ISO 3160/2 standard as follows: 17.5 g NH_4Cl, 20 g NaCl, 5 g CH_3COOH, and 15 g lactic acid were dissolved in 1 L of ultrapure water. The pH was regulated at 4.7. A mass of 1 g of each sample was mixed with 30 mL of artificial sweat solution for 24 h. Then, the solutions were filtered and analyzed by ICP OE (Perkin Elmer, Norwalk, CT, USA) [34]. The operating conditions were adjusted as follow: RF power 1450 W, plasma gas flow rate 15 L min^{-1}, auxiliary gas flow rate 0.2 L min^{-1}, sample flow rate 1.5 mL min^{-1}, Read delay (s) 50 s, replicates 3 and plasma gas: Argon.

2.6. Assessment of Natural Dye Attachment

Natural dye attachment was evaluated based on colorimetric data using a laboratory machine (Data Color 650®, Datacolor, Suzhou, China) under the following parameters: illuminant D65 and 10° standard observer. Color parameters (L, a, b, and K/S) were evaluated. The parameters L, a, b and K/S refer to lightness-darkness, red-green share yellow-blue share and the color yield, respectively.

2.7. Color Fastness

For assessment of color fastness levels of dyed fabrics, standard methods were used. Color fastness (washing, rubbing and light) were carried out by using scales of color change according to ISO 105-C06 (washing), ISO 105-X12 (dry and wet rubbing) and ISO 105-B02 (light).

2.8. Antibacterial Activity

The antibacterial activity of dyed samples was evaluated according to the ASTM E2149 standard. *Staphylococcus aureus* (*ATCC 6538*) and *Escherichia coli* (*ATCC 8739*) bacteria strains were used during experiments. A mass of 1 g of the dyed samples was placed in a contact with bacteria (3×10^5 UFC/mL). Treated and control samples were shaken with the bacterial suspension for 1 h at 200 rpm. Then, 0.1 mL of prepared suspension was distributed over a petri dish. All petri dishes were incubated for 24 h at 37 °C. Finally, the formed bacteria colonies were counted. Results are presented as viable colonogenic cell numbers in CFU/mL.

3. Results and Discussion

3.1. Surface Characterization

ATR-FTIR spectra of the samples before and after plasma treatment are presented in Figure 2. Qualitative analysis showed the characteristic peaks of cellulose [35]. The broad bands at 3271 cm^{-1} corresponds to hydroxyl groups (O-H) in cellulose [36]. The peak at 2896 cm^{-1} is characteristic of stretching vibration of C-H in cellulose molecules [37].

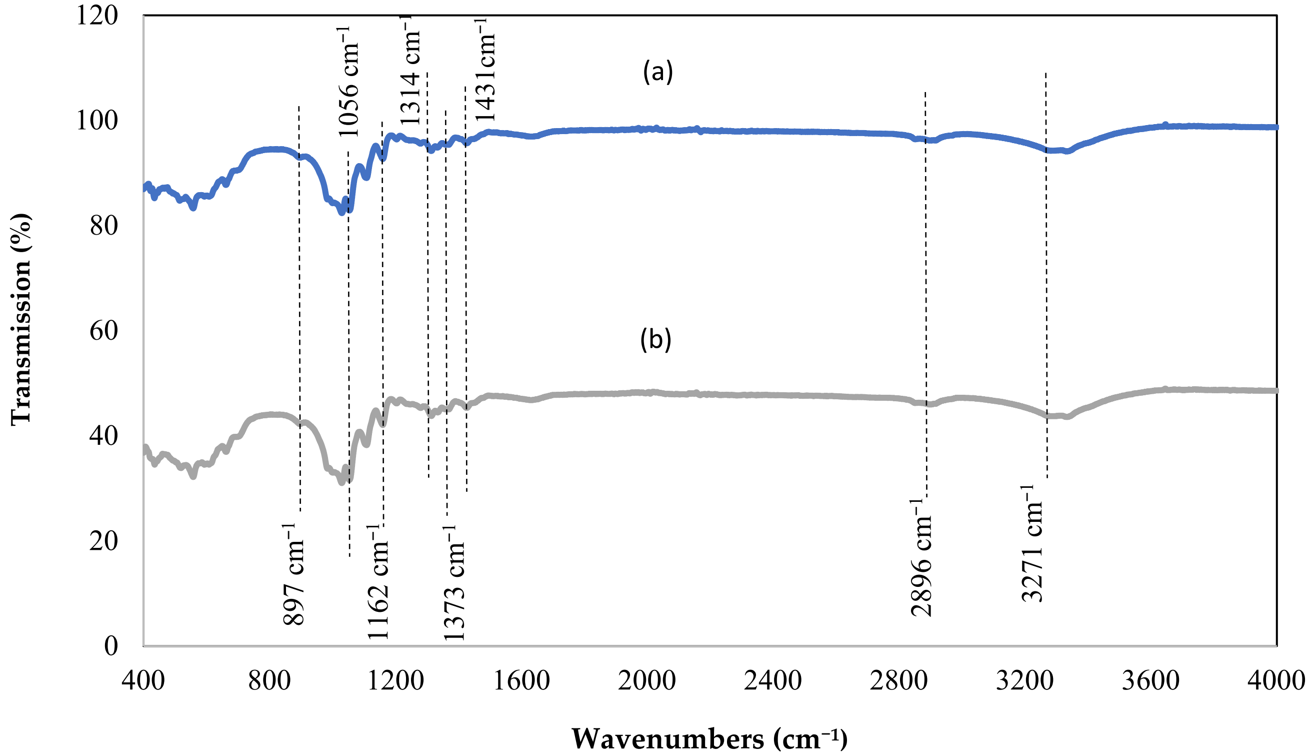


Figure 2. ATR-FTIR spectra of (**a**) untreated cotton and (**b**) plasma treated cotton.

Characteristic peaks at 1431 and 1373 cm^{-1} are attributed to bending vibrations of methyl and ethyl groups ν(CH_3) and ν(CH_2) [38]. The peaks at 1314, 1162–1056 and 897 cm^{-1} are assigned to C–O stretching of ether, C–O–C stretching, and glycosidic C-H deformation, respectively [39,40]. The spectrum of cotton fabrics before plasma treatment did not show any significant difference when compared to the untreated fabric. This outcome is due to the high penetration depth (around 1 μm) of ATR-FTIR analysis. This depth cannot detect a fine surface modification produced by plasma treatment. However, there was a difference between both spectra in terms of peak intensity at 3271 cm^{-1}.

Table 1 show that plasma surface activation generates additional C–OH groups. This finding confirms the results of previous investigations [41].

Table 1. ATR-FTIR analysis: peak area.

Sample	3271 cm^{-1}
Untreated	1.75
Plasma treated	2.13

In general, under air atmospheric plasma, CH/C-C groups decreased while the oxygen groups increased. Thus, it is evident from ATR-FTIR analysis that the plasma treatment increased the oxygen functional groups, which made the cotton surface more active.

The wettability of the cellulosic fibers measured as water contact angle indicated the hydrophilic behavior of the untreated cotton and cotton treated with plasma. Initial contact angle values were considered to evaluate the wettability of different cotton fabrics. Figure 3 shows that plasma-treated cotton fabric had a lower contact angle compared to that of untreated fabric. This result proves that plasma treatment enhances the hydrophilicity of the treated fabric. This result is in agreement with the ATR-FTIR results. The plasma treated cotton fabric had higher percent oxygen. The surface was richer in functional groups associated with a hydrophilic property (C-O and C-OH).

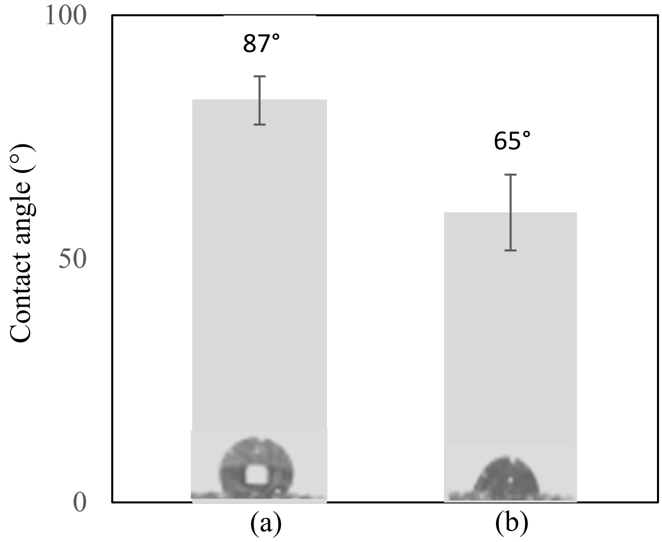
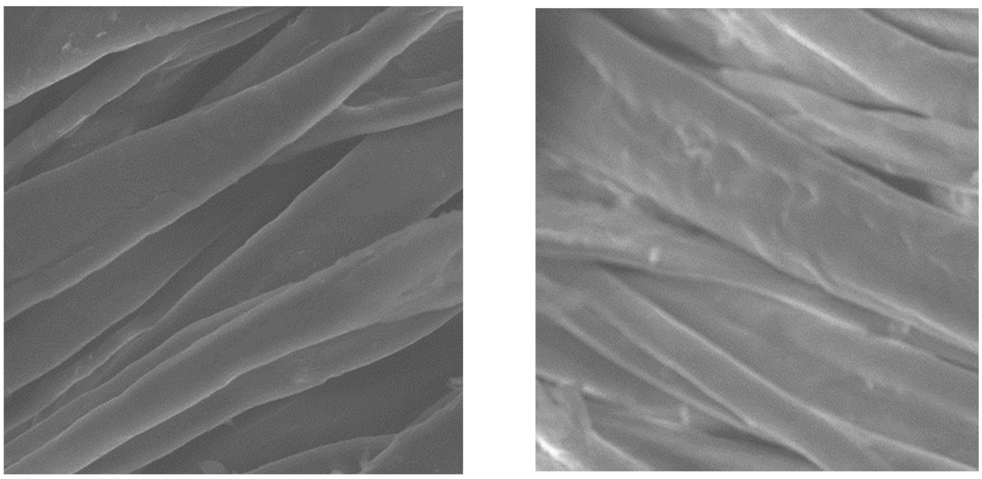


Figure 3. Contact angle values of (**a**) untreated cotton and (**b**) plasma treated cotton.

The normal spiral structure of cotton fibers was evidenced by SEM images (Figure 4). The surface of the untreated sample appeared relatively smooth (Figure 4a). However, the plasma treated surface was rougher than that of the untreated sample (Figure 4b). No significant differences were visible except for the more distinct macrofibrilar structure of cellulosic fibers.



(**a**) (**b**)

Figure 4. SEM images of (**a**) untreated cotton and (**b**) plasma-treated cotton.

3.2. Trace Metal Ions in Finished Samples

During this study, the mordants used were aluminum sulfate, iron sulfate, potassium dichromate and copper sulfate. Metals extraction was conducted using a standard for fabrics used in direct contact with the human skin. The results are presented in Table 2. Results were compared with previous studies. The contents of trace element of Al, Cr and Cu range from 0.11–1.58 µg/mL, 0.01–0.32 µg/mL, and 0.05–1.95 µg/mL, respectively [26]. These concentrations are below the concentration limits set by ecological standards such as Öko Tex [26,34,42]. There are no any limit values for the element Fe, since the use of this metal is not prohibited [43].

Table 2. Metals concentrations expressed as µg of metal/mL.

Mordanted Sample	Element	Concentration	
		Plasma treated	Untreated
Iron sulfate	Fe	32.4 ± 2.4	69.3 ± 0.4
Aluminum sulfate	Al	0.28 ± 0,04	1.11 ± 4.2
Potassium dichromate	Cr	0.29 ± 0.11	0.36 ± 1.4
Copper sulfate	Cu	1.21 ± 0.53	2.14 ± 25

± Standard deviation.

3.3. Colorimetric Parameters

Table 3 shows the influence of cotton treatment on dyed samples. A difference in colorimetric parameters was detected. This was due to the effect of different mordants. The uses of copper sulfate and iron sulfate affected the shades of the dyed fabrics. Many flavonoids can coordinate various metal ions to form metal complexes due to the presence of oxo and hydroxyl groups [44]. The formed complex acts as a binding agent and thus produces deeper color on the fabric [45]. It is clear from the values presented in Table 2 that metal mordants affected the colorimetric data (L, a and b).

Table 3. Colorimetric data of cotton samples.

Sample	K/S	L	a	b
Without mordant	1.37	78.24	2.78	18.03
Iron sulfate	0.85	64.94	0.43	0.99
Aluminium sulfate	0.78	83.71	0.43	19.68
Potassium dichromate	0.24	87.76	−0.27	11
Copper sulfate	1.93	71.69	2.94	20.88

The different obtained colors were expressed in CIELAB (Figure 5); L expresses lightness while a and b parameters define the red/green and yellow/blue value, respectively. A value movement in the direction +a indicates a tendency toward reddish hue. For the b axis, a value movement in the direction + b represents a shift toward yellow. Values moving in the directions -a and -b reflect a tendency towards greenish and bluish shades, respectively. The L scale (degree of lightness) is also presented in Figure 5. Lightness varies from 0 (black) to 100 (white).

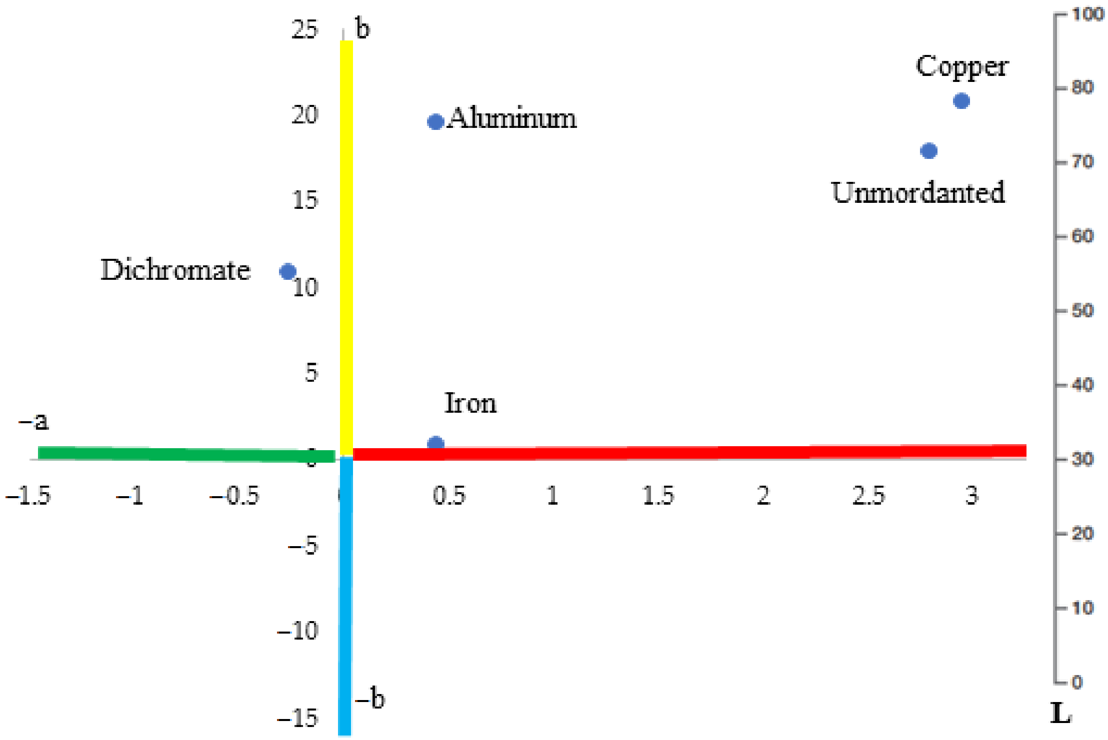


Figure 5. The L, a and b values expressed in CIELAB (CIE, International Commission on Illumination).

3.4. Color Fastness

The results of washing fastness are shown in Table 4. The untreated sample showed a poor color fastness rating. For all mordanted samples, the color change rating was 4–5. The treated cotton fabrics exhibited very good washing fastness.

Table 4. Color fastness.

Sample	Washing	Rubbing (Dry)	Rubbing (Wet)	Light
Without mordant	3	4	4	2
Iron sulfate	4–5	4–5	4–5	2–3
Aluminium Sulfate	4–5	4–5	4–5	2–3
Potassium dichromate	4–5	4–5	4–5	2–3
Copper sulfate	4–5	4–5	4–5	2–3

The rubbing fastnesses (dry and wet) of all mordanted fabrics were very good. Untreated dyed samples exhibited good fastness for dry and wet rubbing properties. Concerning the color fastness to light, the unmordanted sample showed a rating of 2. This result indicates a poor light fastness property. Mordanted samples displayed light fastness ratings of 2–3, which means light fastnesses were slightly improved when using mordants.

The mordant-dye complex has excellent cross-linking performance onto the cotton surface. So, an almost equivalent color fastness was observed for natural dyed cotton as with synthetic dye. However, the natural dye had an inferior light fastness property compared to synthetic dyes [46].

3.5. Antibacterial Activity

Antibacterial activity results after 1 h of contact are shown in Table 5. The counting test results of mordanted samples showed better activity against bacteria, causing bacterial reduction. It is known that metals such as aluminum, iron and copper are effective agents in reducing the growth of a wide range of microorganisms.

Table 5. Antibacterial activity results.

Sample	CFU/mL	
	(*E. coli*)	**(*S. aureus*)**
Without mordant	126.67 ± 8.62	142.11 ± 3.42
Iron sulfate	11.14 ± 5.05	35.14 ± 2.61
Aluminium sulfate	29.2 ± 5.75	38.2 ± 4.12
Potassium dichromate	31.33 ± 1.17	51.21 ± 3.21
Copper sulfate	21 ± 2.79	42 ± 1.64

± Standard deviation.

The mechanism of bacterial inhibition consists of the entry of metal ions into the metabolic system of bacteria with consequent formation of secondary metabolites. These secondary metabolites contain metal ions and are toxic to the organism [47]

Heavy metals cause a change in bacterial cell enzyme activities, growth inhibition and inhibition of replication. These mechanisms lead to cell lysis [48].

4. Conclusions

In this work, an aqueous extract of pomegranate peel was used for dyeing cotton fabrics. To improve the attachment of mordants metal ions, plasma treatment of cotton samples was used as a safe and ecofriendly pretreatment. Contact angle, ATR-FTIR, and SEM analysis showed the effects of plasma treatment on the surface morphology and chemistry of cotton fibers. Colorimetric data were evaluated. The results prove that the attachment of pomegranate peel dye was enhanced using metal mordants. Mordanting with aluminum sulfate, potassium dichromate, ferrous sulfate and copper sulfate enhanced the color yield and fastness properties of dyed samples.

The air atmospheric plasma treatment showed an effect on dying ability of cotton with natural dye and mordant. This was due to an etching effect and the introduction of functional groups on the surface of cotton fibers that enhanced mordant absorption into the plasma treated fibers. Plasma pretreatment is a promising method to lower the impact of toxic metal ions usually released after natural dyeing processes.

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