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Green Application of Isolated Colorant from Neem Bark for Mordant-Coated Wool: Optimization of Dyeing and Mordanting for Shade Development

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Abstract: This study aimed to assess the effectiveness of utilizing a tannin-based natural brown colorant from neem bark for dyeing wool under microwave treatment, specifically evaluating its coloring efficiency. The colorant was extracted in a methanol solution that had been acidified both before and after being subjected to microwave treatment for up to 6 min. The dyeing variables were optimized to create new shades of dye with desirable fastness properties, and sustainable chemical and bio-mordants ranging from 1 to 10 g/100 mL were employed. Through experimentation, it was determined that when an unirradiated acidic methanolic extract (AME) with a salt concentration of 3 g/100 mL was applied onto wool fabric (RWF) and subjected to microwave treatment for 4 min, it resulted in a high color yield. This was achieved by heating the solution to 65 °C and allowing it to remain in contact with the fabric for a duration of 65 min. Favorable color characteristics were achieved when utilizing a pre-chemical mordant of 5% Fe and a post-chemical mordant of 5% Fe. In contrast, the utilization of 7% henna as a pre-bio-mordant in combination with 3% turmeric extract as a meta-bio-mordant resulted in favorable color characteristics. The study concludes that microwave treatment exhibits outstanding sustainable efficacy in isolating colorants from neem bark powder for wool dyeing. Incorporating bio-mordants further enhanced the process's sustainability and eco-friendliness.

Keywords: bio-anchors; green extraction; microwave radiation; neem; sustainability; tannin



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1. Introduction

Synthetic dyes are organic substances synthesized from coal tar, benzene, toluene, and their other derivatives [1]. These dyes are considered cheaper, brighter, and colorfast and can be applied easily onto a matrix [2]. Direct dyes, reactive, azo dyes, basic dyes, acidic dyes, etc., are the common synthetic dyes applied in the paper, leather, food, wood, and textile industries [3]. In addition to these benefits, these dyes have numerous disadvantages, such as water pollution, the production of carcinogenic chemicals, and hazardous waste as

industrial effluents [4]. From an application point of view (color range, low cost, and bright shading), these dyes are widely available and exhibit beautiful properties, yet they may cause dermatologic and genotoxic effects on humans and other living beings [5,6]. Hence, due to such acute and lethal effects of these synthetic dyes, the revival of natural dyes in all fields is underway around the globe.

Several advantages make natural dyes an excellent choice and a better alternative to synthetic dyes. The benefits of using natural dyes have attracted people to their frequent use in all fields. These dyes are readily biodegradable and compatible with the environment [7]. These dyes are non-toxic, non-carcinogenic, and non-allergic to the skin. Most natural dyes are also sustainable and Ayurvedic [8]. These dyes produce a variety of soothing colors with or without the aid of different types of mordants [9,10]. These dyes play a vital role in the environment's cleanliness and help maintain the eco-balance of the globe. These dyes are considered antioxidant, insect repellent, flame retardant, deodorant, antibacterial, etc. [11]. These dyes are not only used in the textile industry but also have wide applications in food, flavors, cosmetics, electronic industries (dye-sanitized solar cells, DSSC), etc. [12]. It is also seen that natural dyes used in dyeing techniques in various fields of art maintain their vitality for many years. It is a fact that the natural dyes used in wall paintings, ceramics, and fabrics obtained in ancient excavations have survived to the present day by preserving their clarity as that of the first day. Today, the use of natural dyes, which has started to be preferred especially in artistic printing techniques, is becoming more and more widespread day by day. Especially in the last 10–15 years, theories about the use of natural dyes in the coloring of textiles have significantly changed the view of natural dyeing methods and techniques, leading to the emergence of different perspectives. Today, there are more advanced and versatile natural screening methods using advanced protection.

From a sustainable point of view, textile researchers, traders, and other associated companies are trying to standardize the extraction process to obtain stable and strong shades with high extraction yields [13,14]. These methods involve traditional and modern tools for extracting colorants from plant sources [15]. Previously, researchers tried to employ two methods, i.e., either to improve the extraction process by using the Soxhlet method, immersion method, supercritical method [16], acidic and alkali extraction, soaking method [17], adsorption methods, etc., or to modify the fabric surface by using techniques such as cationization, mercerization, and bio-polishing [18]. These tools either cause degradation of the colorant's ability to interact firmly with fabric or are expensive in terms of the solvent, energy, and money required to give acceptable results [17]. However, the role of novel tools such as gamma radiation, ultrasonic radiation, and microwave irradiation is receiving more attention because of their high treatment speed and energy- and cost-effective nature in the field of isolation of natural products and their application in various fields such as textiles, cosmetics, pharmaceuticals, leather, food, and flavors [19–21].

Being a clean, rapid, and even heating source, the microwave (MW) is one of the modern tools in synthetic chemistry, particularly in the field of dye synthesis and its application [22]. Because of its time-, cost-, chemical-, and energy-effective nature, it is one of the best tools for this application in natural dye industries [23]. This radiation works through the charges in the liquid or the conducting ions in the solid to shift the energy into heat, driving the process compared to conventional heating [24]. During dyeing, microwave radiation uniformly heats the substrate from core to surface, which causes uniform coloration in a very efficient manner, giving excellent results [21,25]. Microwave radiation (MW), the commercially viable heating source, is found everywhere at the domestic and industrial levels. In the world of natural products, this conventional heating source adds value to the isolation yield without any damage to the actual nature of a potent molecule [26–28]. Hence this unique mode of action requires less solvent consumption, time, labor, and money and lower temperatures [29].

Bio-mordants are another type of anchor where the functional biological isolate acts as the bridging agent between the fabric and the dye. Their application involves extra H-bonding to develop a new and firm tint when applied onto the fabric before, after, and

during the dyeing [30]. The application of biological anchors is a newly introduced novel technique to obtain excellent tint with the desired fastness properties and make the process cleaner, sustainable, and soothing [31]. Another advantage is that these potent molecules exhibit excellent therapeutic, Ayurvedic, and attractive characteristics such as antibacterial, antiviral, anti-inflammatory, and antioxidant properties, which upon the dyeing, make the dyed fabric more beneficial to the global community [32,33]. Bio-mordants are part of medication systems in Greece, China, India, etc., so these are natural blessings that add value to sustainability and give new dimensions to the eco-friendly approach of the natural dyeing process [34]. Chemical mordanting is an old conventional art of fixing color onto fabrics. The salts of Al^{+3} (Alum) and Fe^{+2} (FeSO_4) are commonly used as sustainable anchors for this purpose, while the salts of Cu^{+2} , Cr^{+3} , Co^{+2} , and Sn^{+2} are also used [35]. However, due to toxicity, using Cr^{+3} , Co^{+2} , Sn^{+2} , and Cu^{+2} salts is under strict observation. Mordants cause either the brightening of the shades or the darkening of the tint, depending upon the nature of the fabrics and the dye [36]. Among all mordants, aluminum (Al) is the safest to use and cheapest to buy, whereas chrome (Cr) and Tin (Sn) are the most expensive mordants. However, mordants can reduce the fabric's tenacity [6].

Neem bark (*Azadirachta indica*) is a source of a natural red-brown dye belonging to the Meliaceae family. It is one of nature's blessings and is a powerful home remedy for many health-related problems [37]. Its bark extracts have excellent antifungal, antiviral, antibacterial, anti-inflammatory, antimalarial, antidiabetic, and anti-allergic characteristics [38]. Its extract also exhibits anti-UV, insect repellent, antiarthritic, antipyretic, hypoglycemic, and antistress characteristics. Due to such excellent benefits, it has a good place in Ayurvedic, Greek, and Chinese systems of medicine [39–41]. Proteinous wool fiber has keratin as its significant functional moiety [42,43]. In its helical structure, the amino acids are linked through peptide linkage to form different proteins. Wool fiber, being amorphous, is more sorbent and hypoallergenic. The amido linkage of the wool keratin is responsible for the bonding with the functional sites of the dye [44–46].

The aim of the current study was to use microwave treatment for the isolation of colorant (tannin) from neem bark and surface scaling of wool for maximum sorption. Additionally, plant-derived biomolecules were included as eco-mordants to develop new colorfast tints.

2. Materials and Methods

2.1. Collection of Materials

Neem bark powder, in conjunction with various bio-mordants, including pomegranate (*Punica granatum* L.) peels, *Acacia nilotica* bark, turmeric (*Curcuma longa*) rhizomes, and henna (*Lawsonia inermis*) leaves, were procured from the local market in Faisalabad. The aforementioned samples were subjected to fine grinding to achieve powders of uniform particle size via a 25-mesh sieve. Chemical mordants, including $\text{Al}^{+3}(\text{SO}_4)_3$, Fe^{+2} (FeSO_4), Cu^{+2} (CuSO_4), Co^{+2} (CoCl_2), tannic acid (T.A.), Ni (NiSO_4), Sn (SnCl_2), and Cr (CrCl_2), were acquired from Paragon Dyes and Chemicals, Faisalabad.

2.2. Extraction and Irradiation Process

The present study utilized an acidified methanol extraction medium to isolate natural dye from neem bark powder effectively. Specifically, 4 g of finely prepared neem bark powder was heated with 100 mL of distilled water at the boiling point for 45 min, filtered, and stored. A methanolic extract from an acidic medium, utilizing a 1% HCl solution ($v/v\%$), was obtained using the same procedure. One part of the acidified methanol extracts and woolen fabric were exposed to a domestic M.W. irradiator and heated for 6 min at high power. In the first series, the unirradiated woolen fabric (NRWF) was dyed with unirradiated neem bark extract (NRE) at a temperature of 65 °C. For comparative purposes, irradiated woolen fabric (RWF) was dyed with irradiated neem bark extracts (RE) at 65 °C for a duration of 45 min.

2.3. Optimization of Dyeing and Mordanting Conditions

Response surface methodology was employed as a statistical approach to create a central composite design consisting of 32 experiments. Various parameters, as listed in Table 1, were utilized for computer simulation to generate the design for the different trials.

Table 1. Levels for optimization of dyeing conditions.

Radiation Used	Powder Amount (g)	Volume (mL)	Dyeing Temperature (°C)	Dyeing Time (min)	Table Salt (g/100 mL)
Microwave	2	10	25	25	0
	4	20	35	35	1
	6	30	45	45	3
	8	40	55	55	5
		50	65	65	7
	10	60	75	75	9
		70	85	85	10

2.4. Mordanting Process

The demand for colorfast gamuts has led to a global interest in utilizing eco-friendly chemicals in natural dyeing. The present study utilized the electrolytes of Al, Fe, Cu, Co, Sn, and tannic acid. To prepare the Al-mordant solution, 1, 3, 5, 7, 9, and 10 g of $\text{Al}_2(\text{SO}_4)_3$ were dissolved in 100 mL of water and utilized at a temperature of 65 °C for a duration of 65 min, with the fabric to acidified methanolic extract value maintained at 1:25. The same process was carried out before and after dyeing, utilizing iron, Cu, Sn, Co salt, and tannic acid at predetermined concentrations. In order to carry out comparative studies, plant extracts were utilized as a natural source of biomolecules. Pomegranate peels, acacia bark, turmeric rhizomes, and henna leaves were ground and sieved up to 25 mesh. The resulting extracts were used to prepare bio-mordants by boiling 1–10 g with 100 mL of water for 65 min. The resulting filtrates were utilized before and after dyeing at a temperature of 65 °C for a duration of 65 min, with the bio-mordant to fabric ratio maintained at 25:1.

2.5. Evaluation of Color Characteristics of Dyed Fabrics

Subsequently, the color attributes of the dyed fabrics were evaluated utilizing the Spectra flash SF 600 at the Department of Applied Chemistry, Government University Faisalabad, Pakistan. The dyed fabrics' colorfastness properties were examined per ISO standards to determine the efficacy of the bio-mordants compared to the chemical mordants. The ISO 105 CO3 method was used for washing fastness evaluation in a Rota wash, while ISO 105 BO2 was employed for light fastness evaluation in a fadometer. The ISO 105 X-12 method was used for crocking (rubbing) fastness evaluation, and the ISO 105 E04 method was employed for perspiration fastness evaluation in a perspirometer. Lastly, dry cleaning fastness was evaluated utilizing the ISO 105 D01 method at the Department of Chemistry, Government College University Faisalabad, Pakistan.

3. Result and Discussion

Sustainable technologies are now attracting people, research organizations, and industrial sectors due to their cost-, energy-, and time-effectiveness in nature [47]. Their role in natural products is much appreciated [25]. Their rapid action and use of less time, solvent, and energy have improved their yield of natural products (colorants) from crude materials with suitable results [48,49]. The same role of sustainable technologies, i.e., microwave radiation, was observed in our studies when neem bark was used to isolate reddish-brown natural pigment (tannin) for wool dyeing. The results obtained after treatment with radiation, as shown in Figure 1, show that the treatment of both the fabric and the extract should

be conducted for 4 min before dyeing, whereas after dyeing, the highest yield has been observed. Three factors play their roles side by side, i.e., (i) name of the fabric, (ii) name of extract, and (iii) medium selected. These rays are eco-friendly and act uniformly on plant material by creating solvent polymerization [24]. The energy referring to solvent molecules when they collide with the plant's outer wall causes its rupture and the evolution of functional materials due to rapid mass transfer kinetics [25,26]. Upon treatment, at 4 min, the maximum bioactive mass (tannin) from neem bark was developed, and upon application, tannin imparts dark shades onto wool (Figure 1). The solvent role is also much appreciable because acidified methanol can quickly produce the maximum tannin level through the solid–solvent interaction (SSI) up to a high extract level (K/S = 4.1165). Also, wool, a natural protein, contains amido linkage for interaction with dye-binding sites, hence the addition of acid to organic molecules to obtain an excellent yield. The third factor is the irradiation of fabric, which has been found to be significant for its surface tuning. Chemically, these rays do not disturb its nature, but physically, they scratch the fiber surface, and where the peeled surface is used for the sorption of colorant, a high yield is obtained [50,51]. Our previous studies have confirmed that MW irradiation has no potential to alter the chemistry of fiber but only modifies the wool surface physically to obtain maximum yield [52,53]. Hence, it was concluded that extracts and fabric should be subjected to MW irradiation for up to 4 min.

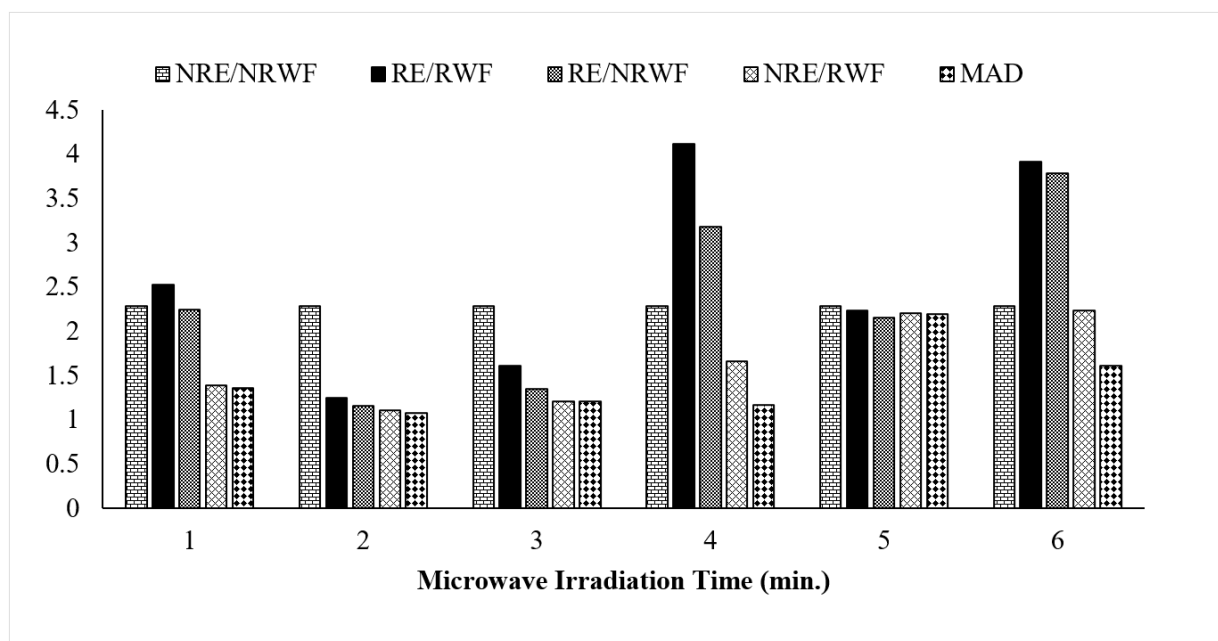


Figure 1. Influence of microwave radiation for isolation of colorant from neem bark in acidified methanolic extract for isolation of colorant in an aqueous medium (c) and its dyeing of wool.

Two-way ANOVA was applied to observe the radiation rate on the extract and fabric together or alone. The results revealed in Table 2 show that the model used is fit ($p = 0.04$), the choice to use MW treatment (MWT) for both fabric and extract is also significant, and treatment time (MW = 4 min) ($p = 0.012$) is of higher significance ($p = 0.002$).

The extract obtained from the selected powder amount was subjected to MW treatment for up to 4 min after utilization, furnishing a high yield. Low powder amounts (2 g/100 mL) do not give a high colorant yield, whereas above 4 g/100 mL, other molecules present in the extract play their role and, during dyeing, disturb the shade strength. The maximum yield with level shade is obtained when 4 g of neem powder is subjected to extraction with 100 mL of acidic methanol medium (AMM) followed by treatment for up to 4 min. Similarly, 40 mL of extract obtained from 4 g/100 mL of acidified methanol also developed a dark shade upon treatment for up to 4 min. Thus, it is clear that 40 mL of AAM from 4 g

powder after 4 min of MW heating can be used to obtain a high yield. The utilization of 4 g powder for extract and the value for dyeing revealed that the MW is a cost-effective tool. In wool dyeing, the nature of the extract is essential because wool contains amido units, which work well under acidic conditions to interact with the extract. Under acidic conditions, the wool's amino group is positively charged, so the -OH from the dye should attach firmly to yield a high yield [54].

Table 2. Two-way ANOVA design for optimization of irradiation and extraction of colorant from neem bark for wool dyeing under microwave radiation.

Source	Type III Sum of Squares	Df	Mean Square	F	p-Value
Corrected Model	13.185	9	1.465	4.217	0.004
Intercept	126.958	1	126.958	365.478	0.000
Sample course	5.909	4	1.477	4.252	0.012
Time	7.276	5	1.455	4.189	0.009
Error	6.948	20	0.347		
Total	147.091	30			
Corrected Total	20.133	29			

The results in Figure 2 show that applying an acidified methanolic extract with a pH of 3 to fabric yields a high yield. Above that (pH > 3), the nature of the colorant is also disturbed, as is the acidic group of wool amido linkage, which also loses its affinity to bind firmly [55]. Adding salt is also beneficial for wool dyeing because the salt adds value by exhausting the colorant from the medium toward the fabric [19]. A low amount cannot perform the activity, whereas a high amount creates exhaustion, resulting in aggregation or poor fixation upon the finishing process. Many colorants are stripped, and low yield is observed. Hence, acidified methanol is extracted at pH 3 with 3 g/100 mL of salt (OWF). After treatment with MW rays for up to 4 min, it yields excellent color strength when employed onto surface-tuned fabric. Adding auxiliaries has always been helpful because these additives reduce the dyeing time of wool and the dyeing temperature during the natural dyeing process. It has been found that dyeing of MW-treated fabric at 65 °C for 65 min, using 40 mL of an acidified methanol extract (3 pH) containing 3 g/NaCl of sodium sulfate, gave a high yield. For low contact levels, the dyeing rate is low, whereas the above-selected (65 °C/65 min) desorption rate is favorably rated, and in both cases, after washing, a low yield is found [56]. Hence, the results displayed in Figure 2 show that MWT of the fabric and extract also reduced contact levels, which are the properties of the energy- and time-effective nature of MW treatment.

Mordanting is the art of developing colorfast shades using plant colorants. Previously, chemical mordants such as a salt of Al, Fe, Cu, Ni, Sn, and tannic acid have been used [45]. However, owing to toxicity, salts of Cu, Ni, Sn, and Co are under strict observation, and only low amounts are allowed to be used to develop colorfast shades. Microwave treatment of fabrics and extracts before mordanting has made it possible to use less toxic anchors to develop colorfast shades [24]. The results in Table 3 show that 40 mL of 7 g/100 mL Cu, 3 g/100 mL of Sn, and 1 g/100 mL T.A. salt developed a colorfast shade with a reddish-yellow hue. However, 5 g/100 mL of Al, Co, and Fe developed high strength before dyeing with a dark reddish-yellow hue.

Similarly, after dyeing with 5 g/100 mL of Fe and T.A., the dye developed an excellent shade, whereas during dyeing with 5% T.A. and 3% Fe salt, the dye developed a dark reddish shade (Table 3). Hence, using all chemical anchors for tannin from neem bark, only salt of Al, Fe, and T.A. is suitable to develop dark reddish-brown shades on the wool fabric surface. The metal develops a coordinate bond when interacting with dye as a bridging agent on fabric [57,58]. The particular type of binding creates new shades with excellent fastness. Low amounts of mordants do not form stable complexes on fabric, whereas too much anchoring forms dye complexes in the form of aggregates, hence failing to sorb onto

fabric evenly [59]. It is only the optimal amount of metal (5 g/100 mL) depending upon its reduction power that adds value in coloration by furnishing a dark colorfast gamut. Hence, the overall salt of Al, Fe, and tannic acid has an excellent affinity for neem-bark-based tannin for wool to furnish colorfast shades under the selected dyeing and radiation conditions. Despite the use of toxic salt as a fixer, the concept of using a plant extract is now gaining fame [31]. The reasons behind using pollution-free fixers are as follows: one is that a plant extract has excellent biological characteristics, and the other is that new shooting shades with excellent fastness are obtained [19]. Most plants have phenolics that utilize the -OH group to bind with the -OH of the colorant and -CONH of wool; their extra H-bonding develops a colorfast gamut [8]. The results reveal that among bio-mordants used, henna after dyeing, and a 5% turmeric extract was notable; during dyeing, a 3% turmeric extract developed dark reddish-yellow shades. Compared to chemical fixers, plant extracts have the highest yields with dark reddish-yellow hues.

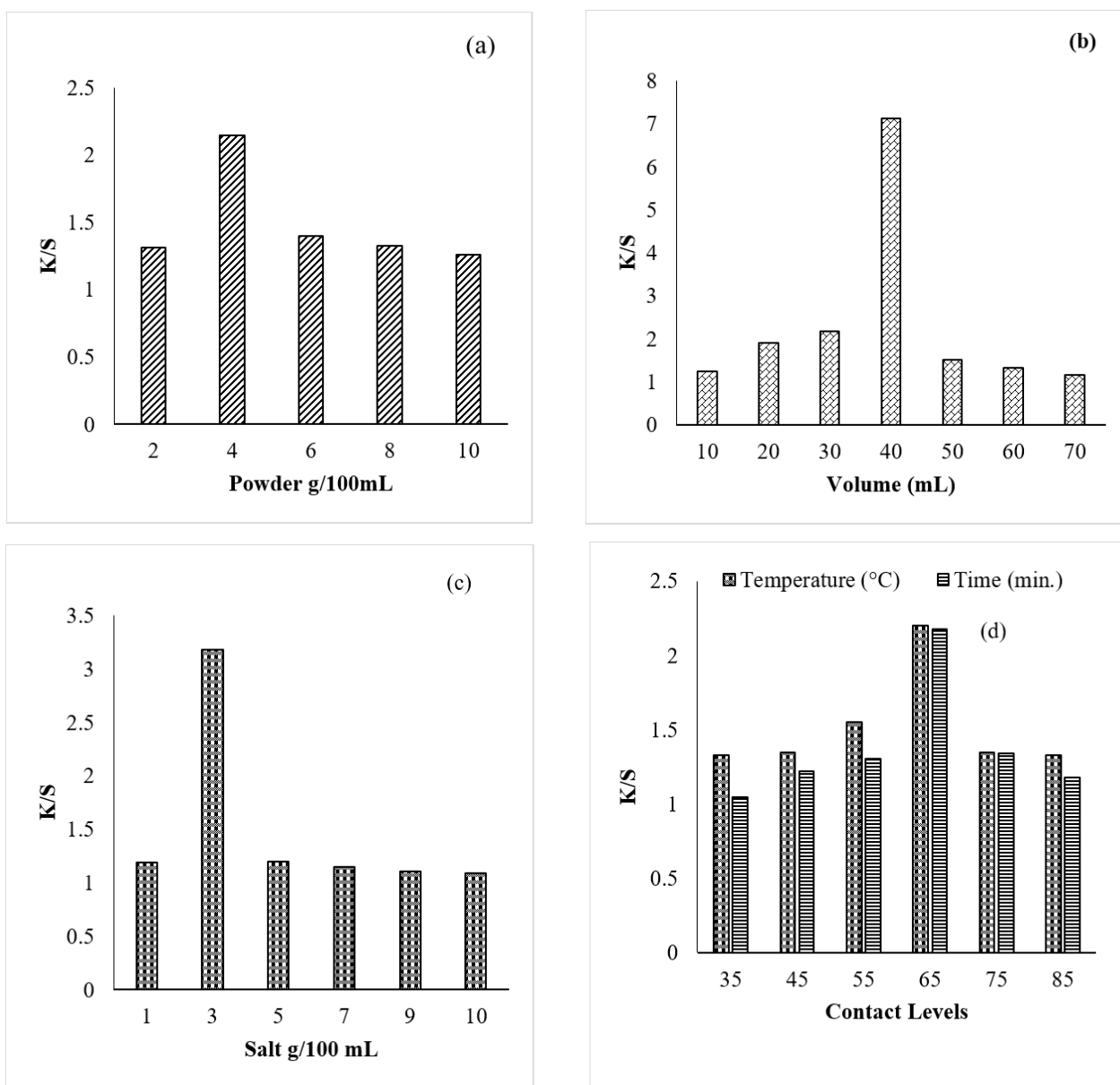
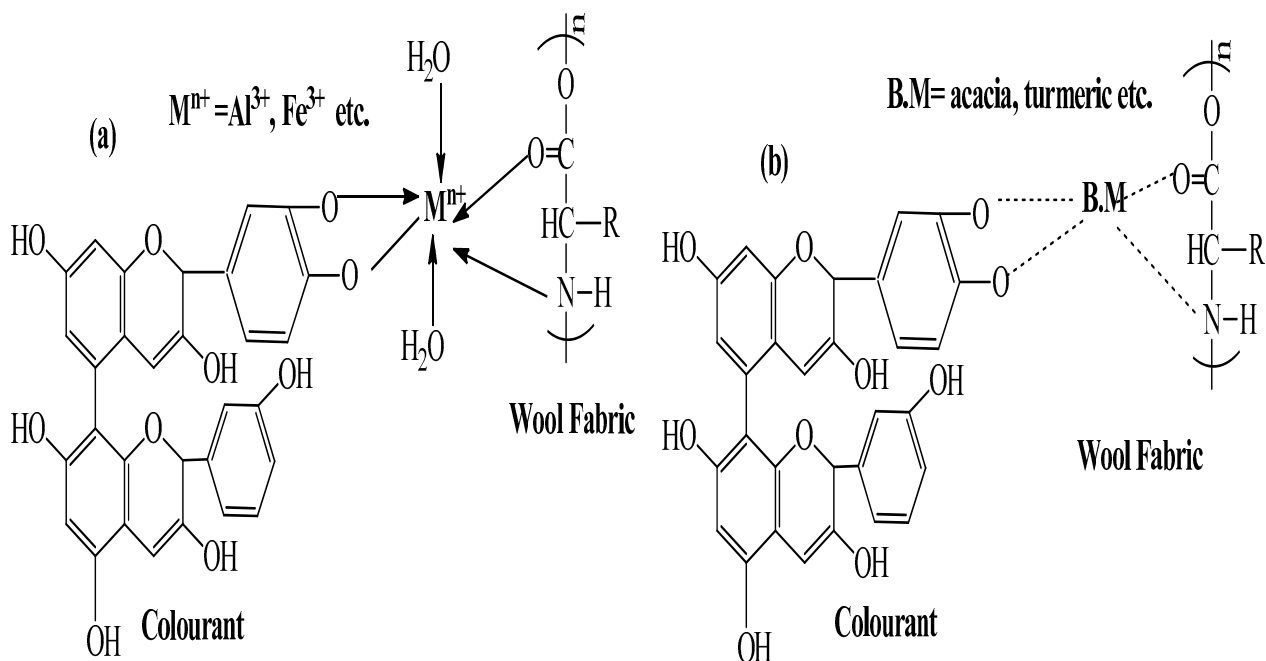


Figure 2. Optimization of dyeing variables such as contact level (a), salt (b), volume (c), and powder (d) under the influence of microwave radiation for dyeing of wool with optimal extract of neem.

Table 3. Color characteristics of chemical and bio-mordanted wool fabrics dyed before, after, and during dyeing with microwave-treated neem bark extract.

Mordant Concentration	K/S	L*	a*	b*	Mordant Concentration	K/S	L*	a*	b*
Al 5% (Pre)	4.0798	61.81	12.24	27.21	T. A 1% (Pre)	2.9228	58.86	5.35	17.47
Al 7% (Post)	1.9339	68.03	6.52	21.02	T. A 5% (Post)	3.7146	52.79	7.00	17.47
Al 5% (Meta)	1.8577	73.18	10.67	28.03	T. A 5% (Meta)	3.7450	50.01	6.58	14.07
Fe 5% (Pre)	4.1340	62.51	8.06	27.04	Acacia 5% (Pre)	3.6193	56.11	13.99	20.24
Fe 5% (Post)	3.8795	69.23	8.89	33.98	Acacia 5% (Post)	3.2125	57.08	14.24	19.97
Fe 3% (Meta)	2.5213	66.19	5.32	21.52	Acacia 3% (Meta)	3.8085	56.89	15.24	22.91
Co 5% (Pre)	3.7763	63.70	11.52	27.81	Pomegranate 7% (Pre)	4.6298	67.59	8.21	25.79
Co 3% (Post)	2.5504	65.54	9.69	22.08	Pomegranate 5% (Post)	5.6319	60.26	10.56	25.08
Co 5% (Meta)	3.1093	60.75	9.99	21.29	Pomegranate 3% (Meta)	7.0519	59.37	10.88	27.55
Sn 3% (Pre)	2.0064	56.32	7.76	12.20	Henna 7% (Pre)	6.2985	55.38	11.12	27.46
Sn 3% (Post)	1.6422	62.14	9.73	15.22	Henna 5% (Post)	4.2884	61.60	11.41	27.85
Sn 5% (Meta)	0.4971	84.86	2.48	10.41	Henna 7% (Meta)	10.427	46.63	6.05	22.01
Cu 7% (Pre)	0.6167	80.49	0.46	7.88	Turmeric 7% (Pre)	6.0265	59.25	9.25	28.02
Cu 3% (Post)	0.5680	80.83	0.21	7.64	Turmeric 5% (Post)	13.285	57.52	11.30	47.29
Cu 3% (Meta)	1.2408	69.96	5.56	12.69	Turmeric 3%(Meta)	15.567	56.52	9.53	50.41

Poor fastness ratings are a big issue in using natural dyes for all fields. To overcome this issue, sustainable chemicals and bio-extracts have been used [24]. The rating given in Table 4 reveals that using the selected amount of mordants before and after dyeing has given a colorfast gamut. The proposed mechanism in Figure 3 shows that the stable metal dye complex formed on surface-modified wool and extract H-bonding formed using biomolecules have developed shades and are resistant to light, washing, crocking, cleaning, and perspiration [31,32]. Hence, the problem of poor fastness can be overcome by using optimal amounts of chemicals and bio-mordants in the surface tuning of fabrics and using pollution-free tools. The variation in shades described in Figure 4 shows that tannin isolated from neem bark is effective for wool when used under MWT before and after mordanting.

**Figure 3.** Proposed interaction of metal (a) and mordants (b) with functional group of dye and binding site of wool fabric.

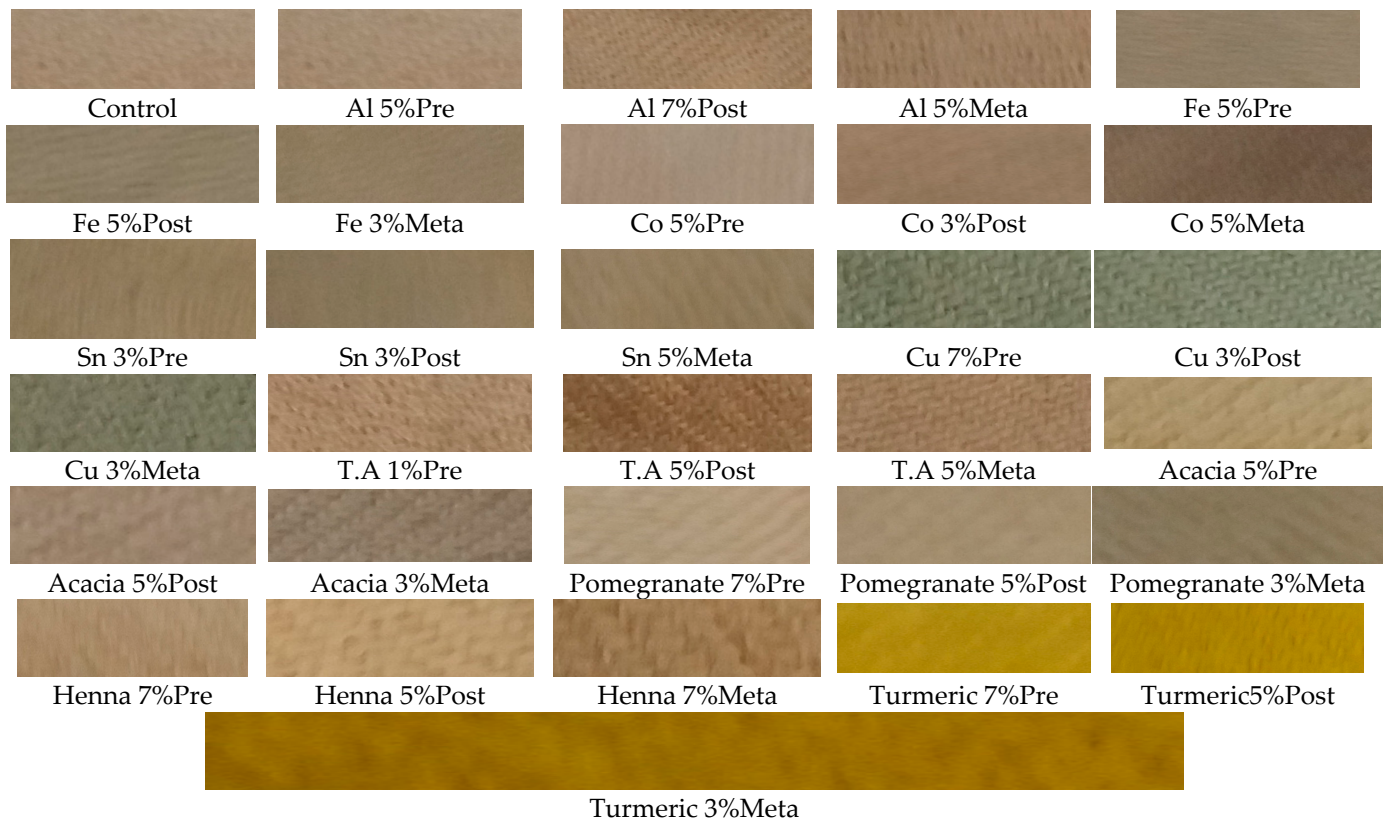


Figure 4. Tonal variation of microwave-treated dyed and mordanted wool fabrics using neem bark extract.

Table 4. Colorfastness rating of mordanted and dyed microwave-treated wool fabric using neem bark extract.

Mordant Concentration	LF	WF		RF		DCF	PF	
		c.s	c.c	DRF	WRF		Acidic	Alkaline
Control	3/4	3	3/4	3/4	3/4	3/4	3/4	3/4
Al 5% (Pre)	5	4/5	4/5	5	4	5	5	5
Al 7% (Post)	5	4/5	4/5	5	4/5	4/5	5	5
Al 5% (Meta)	5	4/5	4/5	5	4/5	4/5	5	5
Fe 5% (Pre)	5	4/5	4/5	5	4/5	5	4/5	5
Fe 5% (Post)	5	4/5	4/5	5	4/5	5	4/5	5
Fe 3% (Meta)	5	4/5	4/5	5	4/5	5	4/5	5
Co 5% (Pre)	4/5	4	4	4/5	4	4/5	4/5	5
Co 3% (Post)	4/5	4	4	4/5	4	4/5	5	4/5
Co 5% (Meta)	4/5	4	4	4/5	4	4/5	5	4/5
Sn 3% (Pre)	4/5	4	4	4/5	4	5	5	4/5
Sn 3% (Post)	4/5	4	4	4/5	4	5	5	4/5
Sn 5% (Meta)	5	4	4	4/5	4	5	5	4/5
Cu 7% (Pre)	4/5	4	4	4/5	4	4/5	5	5
Cu 3% (Post)	5	4	4	4/5	4	4/5	5	5
Cu 3% (Meta)	5	4	4	4/5	4	4/5	4/5	4/5
T. A 1% (Pre)	5	4/5	4/5	5	4/5	5	4/5	4/5
T. A 5% (post)	5	4/5	4/5	5	4/5	5	4/5	4/5
T. A 5% (Meta)	5	4/5	4/5	5	4/5	5	5	5
Acacia 5% (Pre)	5	4/5	4/5	5	4/5	4/5	5	5
Acacia 5% (Post)	5	4/5	4/5	5	4/5	4/5	5	5
Acacia 3% (Meta)	4/5	4/5	4/5	5	4/5	4/5	5	5

Table 4. Cont.

Mordant Concentration	LF	WF		RF		DCF	PF	
		c.s	c.c	DRF	WRF		Acidic	Alkaline
Pomegranate 7% (Pre)	4/5	4/5	4/5	5	4/5	4/5	5	4/5
Pomegranate 5%(Post)	5	4/5	4/5	5	4/5	5	5	4/5
Pomegranate 3%(Meta)	5	4/5	4/5	5	4/5	5	5	4/5
Henna 7% (Pre)	4/5	4/5	4/5	5	4/5	5	5	5
Henna 5% (Post)	5	4/5	4/5	5	4/5	5	5	5
Henna 7% (Meta)	5	4/5	4/5	5	4/5	4/5	5	5
Turmeric 7% (Pre)	4/5	4/5	4/5	4/5	4	4/5	4/5	4/5
Turmeric 5% (Post)	4/5	4/5	4/5	4/5	4/5	4/5	4/5	4/5
Turmeric 3%(Meta)	4/5	4/5	4/5	4/5	4/5	4/5	4/5	4/5

LF = light fastness, WF = wash fastness, c.s = color stain, c.c = color change, RF = rub fastness, DRF = dry rub fastness, WRF = wet rub fastness, DCF = dry clean fastness, PF = perspiration fastness.

4. Conclusions

The awareness spread by research organizations has compelled the use of pollution-free green technologies in textile processing. Sustainable products such as natural dyes for all fields are now catching the eye of the global market due to their versatile health benefits such as antiviral, antioxidant, antibacterial, and antiallergic actions. Neem bark is a natural blessing having an excellent effect for almost all diseases. MW treatment as a green pollution-free heating source has helped to isolate colorants in high yields. The inclusion of plant extracts has provided a benefit in the development of a colorfast shade. These results were statistically analyzed to reveal that this fast and consistent tool is cost-, energy-, and time-effective for textiles. It is concluded that this technology should be used to explore new sources of bio-colorants and their applications under mild conditions. Additionally, herbal plant extracts as alternatives to toxic chemicals should be used to overcome the issue of poor fastness.

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References

1. Khobrani, S.H.A. Synthesis and characteristics of Some organic dyes. *Adv. Appl. Sci. Res.* **2023**, *14*, 109.
2. Al-Etaibi, A.M.; El-Asasery, M.A. Microwave-Assisted synthesis of azo disperse dyes for dyeing polyester fabrics: Our contributions over the past decade. *Polymers* **2022**, *14*, 1703. [[CrossRef](#)]
3. Hassan, E.A.; Mashaly, H.M.; Hashem, Z.M.; Zayed, S.E.; Abo-Bakr, A.M. Eco-friendly synthesis of new polyfunctional azo dyes using shrimp chitin as a catalyst: Application on polyester fabrics and their biological activities. *Fibers Polym.* **2022**, *23*, 2373–2383. [[CrossRef](#)]
4. Talib, A.; Adeel, S.; Ali, A.; Ahmad, T.; Hussaan, M.; Qayyum, M.A. Sustainable isolation and application of plant extract-based natural dye for bio-dyeing of silk fabric. *Coatings* **2023**, *13*, 112. [[CrossRef](#)]
5. Mohamed, A.L.; Shaarawy, S.; Elshemy, N.; Hebeish, A.; Hassabo, A.G. Treatment of cotton fabrics using polyamines for improved coloration with natural dyes extracted from plant and insect sources. *Egypt. J. Chem.* **2023**, *66*, 1–19. [[CrossRef](#)]
6. Özomay, M. Sustainable and environmental dyeing with maut method comparative selection of the dyeing recipe. *Sustainability* **2023**, *15*, 2738. [[CrossRef](#)]

7. Al-Tohamy, R.; Ali, S.S.; Li, F.; Okasha, K.M.; Mahmoud, Y.A.G.; Elsamahy, T.; Sun, J. A critical review on the treatment of dye-containing wastewater: Ecotoxicological and health concerns of textile dyes and possible remediation approaches for environmental safety. *Ecotoxicol. Environ. Saf.* **2022**, *231*, 113160. [[CrossRef](#)]
8. Rahman, M.M.; Kim, M.; Youm, K.; Kumar, S.; Koh, J.; Hong, K.H. Sustainable one-bath natural dyeing of cotton fabric using turmeric root extract and chitosan bio-mordant. *J. Clean. Prod.* **2023**, *382*, 135303. [[CrossRef](#)]
9. Sharma, N.; Sheikh, Z.N.; Alamri, S.; Singh, B.; Kesawat, M.S.; Guleria, S. Chemical Composition, Antibacterial and Combinatorial Effects of the Essential Oils from *Cymbopogon* spp. and *Mentha arvensis* with Conventional Antibiotics. *Agronomy* **2023**, *13*, 1091. [[CrossRef](#)]
10. Thakker, A.M.; Sun, D. Sustainable application of novel herbs on cotton fabrics as bio-mordants and colourants. *Environ. Sci. Pollut. Res.* **2022**, *29*, 47598–47616. [[CrossRef](#)]
11. Otaviano, B.T.H.; Sannomiya, M.; de Queiroz, R.S.; Sánchez, A.A.C.; Freeman, H.S.; Mendoza, L.E.R.; da Costa, S.M. Natural dye extracted from pomegranate peel: Physicochemical characterization, dyeing of cotton fabric, color fastness, and photoprotective properties. *Fibers Polym.* **2023**, *24*, 1321–1332. [[CrossRef](#)]
12. Shaban, S.; Vats, A.K.; Pandey, S.S. Bifacial dye-sensitized solar cells utilizing visible and nir dyes: Implications of dye adsorption behaviour. *Molecules* **2023**, *28*, 2784. [[CrossRef](#)]
13. Ali, N.F.; El-Khatib, E.M.; Bassyouni, F.A. Utilization and characterization of natural products pretreatment and dyeing wool fabric by natural dyes with economical methods. *J. Text. Eng. Fash. Technol.* **2022**, *8*, 178–183. [[CrossRef](#)]
14. Geetha, P.; Dhanya, M.; Sankaralingam, S.; Sugapriya, M.P.; Vasthi, G.R.; Mahendran, S.; Balasundaram, H. Extraction and applications of natural dye from the flower of *Tagetes erecta*. L in different fabrics and focus on antimicrobial activity. *Res. J. Pharm. Technol.* **2022**, *15*, 1287.
15. Sutlović, A.; Brlek, I.; Ljubić, V.; Glogar, M.I. Optimization of dyeing process of cotton fabric with cochineal dye. *Fibers Polym.* **2020**, *21*, 555–563. [[CrossRef](#)]
16. Naik, N.M.; Krishnaveni, M.; Mahadevswamy, M.; Bheemanna, M.; Nidoni, U.; Kumar, V.; Tejashri, K. Characterization of phyto-components with antimicrobial traits in supercritical carbon dioxide and soxhlet *Prosopis juliflora* leaves extract using GC-MS. *Sci. Rep.* **2023**, *13*, 4064. [[CrossRef](#)]
17. Asghar, A.; Tajammal, A.; Ashiq, S.; Hussain, S.; Ahmad, M.; Sian, K.; Bodlah, M.A. Eco-friendly dye of olive fruit peel and its color fastness applications on wool/silk fabrics. *Pol. J. Environ. Stud.* **2023**, *32*, 1995–1999. [[CrossRef](#)]
18. Kabish, A.K.; Abate, M.T.; Alemar, Z.A.; Girmay, S. The importance of natural indigo dye and its revitalization and ethiopian potential for indigo growing. *Adv. Mater. Sci. Eng.* **2023**, *2023*, 2135014. [[CrossRef](#)]
19. Bhouri, N.; Ltaief, S.; Bhouri, N.; Ben Abdesslem, S. Optimization of a green dyeing process using natural dyes extracted from *corchorus olitorius* leaves. *J. Nat. Fibers.* **2023**, *20*, 2170946. [[CrossRef](#)]
20. Castillo-Suárez, L.A.; Sierra-Sánchez, A.G.; Linares-Hernández, I.; Martínez-Miranda, V.; Teutli-Sequeira, E.A. A critical review of textile industry wastewater: Green technologies for the removal of indigo dyes. *Int. J. Environ. Sci. Technol.* **2023**, *20*, 10553–10590. [[CrossRef](#)] [[PubMed](#)]
21. Periyasamy, A.P.; Periyasami, S. Critical review on sustainability in denim: A step toward sustainable production and consumption of denim. *ACS Omega* **2023**, *8*, 4472–4490. [[CrossRef](#)]
22. Yadav, S.; Tiwari, K.S.; Gupta, C.; Tiwari, M.K.; Khan, A.; Sonkar, S.P. A brief review on natural dyes, pigments: Recent advances and future perspectives. *Results Chem.* **2023**, *5*, 100733. [[CrossRef](#)]
23. Khajeh Mehrizi, M.; Jokar, M.; Shahi, Z. The coloration of polyester fabric with Prangos ferulacea natural dye: New approaches toward a cleaner production. *Pigm. Resin Technol.* **2023**, *52*, 377–382. [[CrossRef](#)]
24. El Sayed, N.A.; El-Bendary, M.A.; Ahmed, O.K. A sustainable approach for linen dyeing and finishing with natural lac dye through chitosan bio-mordanting and microwave heating. *J. Eng. Fibers Fabr.* **2023**, *18*, 15589250231155882. [[CrossRef](#)]
25. Mahboob, M.; Adeel, S.; Bakraat, S.; Ahamd, T.; Ahamd, T.; Ozomay, M.; Mia, R.; Mirnezhad, S.; Zuber, M. Assisted santalin extraction from *Pterocarpus santalinus* for mordanted woolen yarn dyeing. *Sustain. Chem. Pharm.* **2023**, *35*, 101224. [[CrossRef](#)]
26. Syafaatullah, A.Q.; Mahfud, M. Optimization extraction of *Indigofera tinctoria* L. using microwave-assisted extraction. *Conf. Ser. Mater. Sci. Eng.* **2021**, *1053*, 012131. [[CrossRef](#)]
27. Ji, X.; Zhao, Z.; Ren, Y.; Xu, F.; Liu, J. Dyeing properties, color gamut, and color evaluation of cotton fabrics dyed with *Phellodendron amurense* rupr.(amur cork tree bark). *Molecules* **2023**, *28*, 2220. [[CrossRef](#)]
28. Phan, P.T.; Hong, J.; Tran, N.; Le, T.H. The properties of microwave-assisted synthesis of metal-organic frameworks and their applications. *Nanomaterials* **2023**, *13*, 352. [[CrossRef](#)]
29. Jabar, J.M.; Ogunsade, A.F.; Odusote, Y.A.; Yilmaz, M. Utilization of nigerian mango (*Mangifera indica* L) leaves dye extract for silk fabric coloration: Influence of extraction technique, mordant and mordanting type on the fabric color attributes. *Indus. Crop. Prod.* **2023**, *193*, 116235. [[CrossRef](#)]
30. Haji, A.; Shahmoradi Ghaheh, F.; Indrie, L. Pomegranate fallen leaves as a source of natural dye for mordant-free dyeing of wool. *Color. Technol.* **2022**, *139*, 165–170. [[CrossRef](#)]
31. Hosseinneshad, M.; Gharanjig, K.; Iamin, H.; Rouhani, S.; Adeel, S. Environmentally dyeing of wool yarns using of combination of myrobalan and walnut husk as bio-mordants. *Prog. Color Color. Coat.* **2023**, *16*, 197–205.
32. Phromphen, P. Optimization of marigold flower dye using banana peel as a biomordant. *J. Nat. Fibers* **2023**, *20*, 2153193. [[CrossRef](#)]

33. Nazir, F.; Siddique, A.; Nazir, A.; Javed, S.; Hussain, T.; Abid, S. Eco-friendly dyeing of cotton using waste-derived natural dyes and mordants. *Color. Technol.* **2022**, *138*, 684–692. [[CrossRef](#)]
34. Shahmoradi Ghaheh, F.; Moghaddam, M.K.; Tehrani, M. Comparison of the effect of metal mordants and bio-mordants on the colorimetric and antibacterial properties of natural dyes on cotton fabric. *Color. Technol.* **2021**, *137*, 689–698. [[CrossRef](#)]
35. Baaka, N.; Khiari, R.; Haji, A. Ecofriendly dyeing of textile materials with natural colorants from date palm fiber Fibrillium. *Sustainability* **2023**, *15*, 1688. [[CrossRef](#)]
36. Islam, M.R.; Khan, A.N.N.; Mahmud, R.U.; Haque, S.M.N.; Khan, M.M.I. Sustainable dyeing of jute–cotton union fabrics with onion skin (*Allium cepa*) dye using banana peel (*Musa*) and guava leaves (*Psidium guajava*) extract as biomordants. *Pigm. Resin Technol.* **2022**. [[CrossRef](#)]
37. El-Khatib, E.M.; Ali, N.F.; El-Mohamedy, R.S.R. Influence of Neem oil pretreatment on the dyeing and antimicrobial properties of wool and silk fibers with some natural dyes. *Arab. J. Chem.* **2020**, *13*, 1094–1104. [[CrossRef](#)]
38. Wylie, M.R.; Scott Merrell, D. The antimicrobial potential of the neem tree *Azadirachta indica*. *Front. Pharmacol.* **2022**, *13*, 891535. [[CrossRef](#)]
39. Baby, A.R.; Freire, T.B.; Marques, G.D.A.; Rijo, P.; Lima, F.V.; Carvalho, J.C.M.D.; Morocho-Jácume, A.L. *Azadirachta indica* (Neem) as a Potential natural active for dermocosmetic and topical products: A narrative review. *Cosmetics* **2022**, *9*, 58. [[CrossRef](#)]
40. Altayb, H.N.; Yassin, N.F.; Hosawi, S.; Kazmi, I. In-vitro and in-silico antibacterial activity of *Azadirachta indica* (Neem), methanolic extract, and identification of Beta. d-Mannofuranoside as a promising antibacterial agent. *BMC Plant Bio.* **2022**, *22*, 1–14.
41. Chrysargyris, A.; Goumenos, C.; Tzortzakis, N. Use of medicinal and aromatic plant residues for partial peat substitution in growing media for *Sonchus oleraceus* production. *Agronomy* **2023**, *13*, 1074. [[CrossRef](#)]
42. Gomaa, S.K.; Zaki, R.A.; Wahba, M.I.; Taleb, M.A.; El-Refai, H.A.; El-Fiky, A.F.; El-Sayed, H. Green method for improving performance attributes of wool fibres using immobilized proteolytic thermozyyme. *3 Biotech.* **2022**, *12*, 254. [[CrossRef](#)] [[PubMed](#)]
43. Sanchez Ramirez, D.O.; Vineis, C.; Cruz-Maya, I.; Tonetti, C.; Guarino, V.; Varesano, A. Wool keratin nanofibers for bioinspired and sustainable use in biomedical field. *J. Funct. Biomater.* **2022**, *14*, 5. [[CrossRef](#)] [[PubMed](#)]
44. Aazou, N.; Nadi, A.; Cherkaoui, O.; Gmouh, S. Study and characterization of Moroccan wool fibers for the development of non-woven products. *Conf. Ser. Mater. Sci. Eng.* **2023**, *1266*, 012016. [[CrossRef](#)]
45. Adeel, S.; Anjum, F.; Zuber, M.; Hussaan, M.; Amin, N.; Ozomay, M. Sustainable extraction of colourant from harmful seeds (*Peganum harmala*) for dyeing of bio-mordanted wool fabric. *Sustainability* **2023**, *14*, 12226. [[CrossRef](#)]
46. Wang, M.; Yi, N.; Fang, K.; Zhao, Z.; Xie, R.; Chen, W. Deep colorful antibacterial wool fabrics by high-efficiency pad dyeing with insoluble curcumin. *Chem. Eng. J.* **2023**, *452*, 139121. [[CrossRef](#)]
47. Průša, D.; Šuhajda, K.; Žajdlík, T.; Svobodová, K.; Šťastník, S.; Hobzova, K.; Venkrbec, V. Effect of microwave radiation on the compressive strength of solid ceramic brick. *Buildings* **2023**, *13*, 1018. [[CrossRef](#)]
48. Elmahaishi, M.F.; Ismail, I.; Muhammad, F.D. A review on electromagnetic microwave absorption properties: Their materials and performance. *J. Mater. Res. Technol.* **2022**, *20*, 2188–2220. [[CrossRef](#)]
49. Stanfield, L.D.; Powell, A.W.; Horsley, S.A.R.; Sambles, J.R.; Hibbins, A.P. Microwave demonstration of Purcell effect enhanced radiation efficiency. *Sci. Rep.* **2023**, *13*, 5065. [[CrossRef](#)]
50. Ibrahim, N.A.; Hala, A.; Mohamed, A.; Abdel-Aziz, S.; Basma, M.E. A green approach for modification and functionalization of wool fabric using bio- and nano-technologies. *Clean Technol. Environ. Policy* **2022**, *24*, 3287–3302. [[CrossRef](#)]
51. Xue, Z. Effect of microwave pretreatment on dyeing performance of wool fabric. *J. Text. Eng. Fashion Technol.* **2017**, *1*, 217–222. [[CrossRef](#)]
52. Norambuena-Contreras, J.; Garcia, A. Self-healing of asphalt mixture by microwave and induction heating. *Mater. Des.* **2016**, *106*, 404–414. [[CrossRef](#)]
53. Zhang, Y.; Rather, L.J.; Li, Q. Recent advances in the surface modification strategies to improve functional finishing of cotton with natural colourants—A review. *J. Clean. Prod.* **2022**, *335*, 130313. [[CrossRef](#)]
54. AL-Khateeb, D.S.M. Extraction dyes from two natural plants olive leaves and beta vulgaris and the uses in dyeing textile. *J. Phys. Conf. Ser.* **2019**, *1294*, 062109. [[CrossRef](#)]
55. Mulder, R.; Morshed, M.N.; Seipel, S.; Norén, U.; Niit, E.; Nierstrasz, V. Study on hydraulic spray atomizing system as a new resource-efficient dyeing-finishing method for wool fabric. *Sci. Rep.* **2022**, *12*, 21814. [[CrossRef](#)] [[PubMed](#)]
56. Rehman, A.; Irfan, M.; Hameed, A.; Saif, M.J.; Qayyum, M.A.; Farooq, T. Chemical-free dyeing of cotton with functional natural dye: A pollution-free and cleaner production approach. *Front. Environ. Sci.* **2022**, *115*, 848245. [[CrossRef](#)]
57. Adeel, S.; Habiba, M.; Kiran, S.; Iqbal, S.; Abrar, S.; Hassan, C.M. Utilization of colored extracts for the formulation of ecological friendly plant-based green products. *Sustainability* **2022**, *14*, 11758. [[CrossRef](#)]
58. Mesrar, F.E.; Tachallait, H.; Cherkaoui, O.; Bougrin, K.; Benhida, R. Green and sustainable dyeing of wool with madder plant of the Moroccan flora. *IOP Conf. Ser. Mater. Sci. Eng.* **2023**, *1266*, 012015. [[CrossRef](#)]
59. Vankar, P.S.; Gangwar, A. Natural dyeing mediated by atmospheric air pressure plasma treatment of polyester. *Pigm. Resin Technol.* **2023**. [[CrossRef](#)]

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