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Assessment of Antibacterial, Antimicrobial, and Colorimetric Properties of Cotton and Woolen Yarns Dyed with Some Plants Extracts

Zahra AHMADI[1*], Fatemeh GHOLAMI HOUJEGHAN[2]

[1]Department of Carpet, Faculty of Applied Arts, Art University, Tehran, Iran

[2]Department of Advance Studies of Art, Faculty of Visual Arts, University of Tehran, Tehran, Iran

*ahmadi@art.ac.ir

Article

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ABSTRACT

Nowadays according to more attention to natural products, the development of research on this issue seems necessary. Many plants are known around the world to have antibacterial, antimicrobial, and antifungal properties. By using those plant compounds, antibacterial, antimicrobial, and fungal properties can be created in textiles. Textiles are one of the unavoidable requirements that all human beings use abundantly in their lives. In the natural dyeing process, the use of some medicinal plant compounds resulted in antibacterial, antimicrobial, and antifungal properties. In this study, medicinal plants such as thyme, clove, marjoram, lavender, wormwood, and espand were studied for antibacterial and antimicrobial properties on textiles. The antibacterial property of mordanted and dyed wool & cotton yarns were tested against Escherichia coli (Gram-negative), Staphylococcus aureus (Gram-positive), and Candida albicans microbe for cotton. The range of colour developed on dyed materials was evaluated in terms of (L, a*, b*) CIE LAB coordinates and the dye absorption concentration on the yarns was studied by using K/S values. Also, fastness tests on dyed samples for light and washing fastness were carried out. The experimental results showed that the examined plants, in addition to creating antimicrobial and antibacterial properties on wool and cotton yarns, can be used as a dye to produce a durable yellow shade. The dyeing fastness of applied dyes was acceptable. Aloe vera, which has a polyphenolic structure, was used to increase dyeing fastness and durability of antibacterial activity against gram-negative and gram-positive bacteria. The property was created to maintain its durability for several washed cycles.*

KEYWORDS

herbal dye, antibacterial, antimicrobial, textile, natural dyeing

INTRODUCTION

The textile industry pollutants spread to the environment. Much research has been done on the use of natural materials for different purposes. Natural materials are biocompatible and more compatible with the human body. There is a tendency to use natural compounds [1]. Bacteria and germs are present in people's lives for many reasons requiring the use of antibacterial and antimicrobial agents. Such places are hospital environments or places with a high density of people such as hotels, religious centers, or humid environments, especially textiles, where infectious diseases are common and the growth of bacteria and germs progresses rapidly. To avoid or reduce the growth of bacteria, the use of antibacterial, antimicrobial, and antifungal substances is

necessary. The large surface area of the textiles and their wetness oxygen nutrients helps the multiplication and the growth of bacteria, which causes problems for the textiles and the user, such as bad odour, stains, paleness or colourless in the fabric, also reduced strength and increased contamination dermal infection [2,3]. But in recent decades, due to increasing environmental pollution and its destructive and harmful effects, such as indestructibility and toxicity, allergy problems and allergies to chemicals have led to the tendency of using natural dyes and compounds to create antibacterial and antimicrobial properties. Many plants have medicinal properties and depending on the molecules in their chemical structure can have antibacterial or microbial properties. Many researchers have studied the antibacterial and antimicrobial properties of plants [4-7]. Antibacterial growth was investigated in woollen fabric with natural biopolymeric material (chitosan) and natural dyeing (turmeric and madder). The results confirmed that the combination of chitosan and natural dye enhanced the antibacterial activity of woollen fiber against *Escherichia coli* and *Staphylococcus aureus* [8]. A study of the effects of artemisia lavender, lavender, and thyme on the bacteria *Pseudomonas aeruginosa*, *S. aureus*, and *Klebsiella pneumoniae* found that all three essential oils have an inhibitory effect on pathogenic bacteria, but plant essential oils have a better effect in decreasing bacteria growth than the essential oils of other plants [9,10]. *Thyme vulgaris* has extensive antibacterial effects against gram-positive and negative bacteria and is effective in inhibiting the growth of *S. aureus*, *Shigella toxic* and *E. coli* bacteria that can be used as food industry preservatives [11]. Also, the thyme plant can be used as an environmentally friendly dye in the dyeing process to give yellow colour to wool fibres, the results of the study indicated good light and washing fastness with this plant [12]. Aloe vera is one of the useful medicinal plants that seem unknown really but has very good properties in the natural dyeing process. This plant is consumed both as an extract and the green plant itself. Its important characteristic is antimicrobial property and improvement of the natural dyeing fastness as a bio-mordant. Also, aloe vera extract contains antibacterial agents that are effective against human pathogens, even those that are resistant to common antibiotics. The results obtained in the study indicated the effect of high concentrations of alcoholic extract of aloe vera effects on bacteria and antifungals that can be more widely used in the future [13-15]. Lavender is one of the most fragrant medicinal plants in the world, which grows in different parts of Iran, India, China, England, and Canada. This plant is used as a medicinal plant with many properties in the traditional medicine of Iran, China, and India [16,17]. Lavender essential oil (in high concentration) has the same effect as antibiotics, and *E. coli* has the most sensitivity to lavender. The alcoholic extract of this plant did not affect the growth of bacteria. This plant has the highest efficiency in controlling gram-negative bacteria and can be used as a herbal drug in the treatment of diseases. Textile has been dyed with lavender lesions and pulps to prevent environmental contamination [10,17]. Peganum contains

antimicrobial substances such as flavonoids and alkaloids, which are found in many parts of the plant. Studies also showed that pecan (peganum) has antibacterial and anticoagulant effects and also has a good effect on antibiotic-resistant bacteria [17,18]. The beta-carboline alkaloids in pecan are the most important plant contents that have the most medicinal effects. In the dying process of woollen yarns with *Peganum harmala*, a comparison of antibacterial effects of natural plant dyes to synthetic materials showed that mordants increase colour absorption but reduce the anti-bacterial properties at the first stage and then the antifungal properties of the wool fibres improved. The researchers suggest using natural dyes with unstable antibacterial properties for baby clothes, sports, and hospitals [19]. Wormwood and marjoram were introduced to preserve the environment and produce healthy colours [20]. Also, the essential oil of wormwood has the most antibacterial properties against *E. coli* and has a small antimicrobial activity against *S. aureus*. It seems that oily extracts due to their hydrophobic properties exhibit more antibacterial properties, and gram-negative bacteria do not allow them to penetrate the medicine plant, due to pod produce polysaccharide membranes and limit the penetration of the essential oil to their layer. The ajwain/ajowan has a strong antibacterial effect. The suggestion was that phenolic compounds of plants interfere with the action of the bacterial cytoplasmic membrane and cause lethal toxicity to them and cause [21]. There is a suggestion to use an alcoholic extract of ajwain/ajowan (in high concentration) instead of some inactive or low-acting antimicrobials. Also, scientists in their research have introduced the use of antimicrobial substances such as nano silver, triclosan, biological plants, chitosan, antimicrobial polymers such as poly biguanides) and N-halamines [19,22-24]. These substances showed considerable antimicrobial activity. The results showed that textiles dyed with extracts of pomegranate peel, kamala and especially tannin, henna, walnut and turmeric peel and nano-silver particles, red seed, eggplant and rhubarb have good colour stability and excellent antibacterial properties against *E. coli* and *S. aureus*. The antimicrobial activity of the samples has been durable for up to ten washing cycles and could be used for hospital and hotel clothes and other silk, wool, and cotton consumer goods. Finally, the results of Yusuf et al. provided evidence for other researchers to introduce espand as a safe and effective source of treatment in the future [25]. The main challenge in using natural products for textile application is that most of these biomaterials are complex and a mixture of several compounds and also plant constitutions are not constant in different species of a particular plant. Depending on the geographical location, age, and method of extraction they change. Durability and antimicrobial efficiency against synthetic agents are other issues that should be considered [26-30]. However, due to their environmentally friendly nature and non-toxic properties, they could be used for applications in medicine and safe and healthy textiles. As mentioned earlier, some plants with antibacterial properties have auxochrome and are mostly used as a dye, such as henna or walnut, and some of them do not have pigments such as chitosan or

aloe vera. It was thought worthwhile to study the antimicrobial and antibacterial activity of natural medicine plants on the textile substrate. Then in the present study, medicine plants with antibacterial and antimicrobial properties with yellow colour were examined to create antibacterial and antimicrobial properties as well as good dyeing properties and appropriate fastness on wool and cotton products. The end use of these textiles is suggested for handwoven carpets or other traditional textiles.

EXPERIMENTAL

Materials and Methods

Materials

Pre-scoured wool yarn with a fineness of 20 metric, pre-scoured cotton yarn with a fineness of 10 Metric, Alum mordant ($KAl(SO_4)_2 \cdot 12H_2O$), and acetic acid were laboratory reagent grades from Merck Germany. Seven natural medicine plants: thymes, ajwain/ajowan, clove, lavender, espand, wormwood, and marjoram (were grinded to obtain a fine powder), and a fresh leaf of aloe vera was used throughout this work. All plants were bought from a herbal pharmacology store in Tehran. The chemical structure of plants has been shown in Table 1.

Dyeing & Mordanting

Mordanting of wool and cotton yarn: Mordanting of wool was carried out with Alum mordant at concentrations on the weight of goods 4% at boiling temperature for 1 h. with material to liquor ratio as 1:40. The mordanted yarn was squeezed and then applied for dyeing.

Grinded natural plants were directly added to the water in the beaker, they were heated to boiling for 1 h. after that the dye bath cooled to 60 °C. The mordanted yarn was added to a cooled dye bath at 60 °C. The dyeing was carried out at 90 °C for 1 h. The pH of the dye bath was adjusted to 4.5 using acetic acid. This pH was adopted from previous studies [31]. The constitution of wool and cotton dyeing bath has been explained in Table 2 and 3.

Table 1. Specifications, chemical structure, formulation of some ingredients of selected natural medicine plants (dyes) [29]

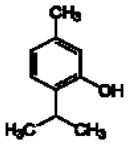
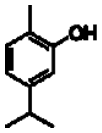
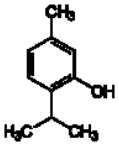
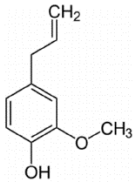
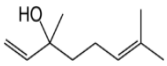
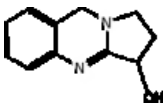
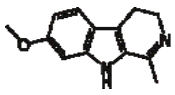
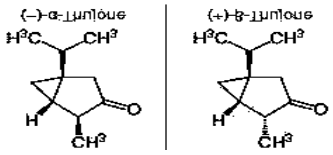
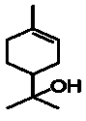
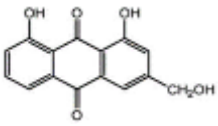
Name	Latin name	Used parts	Structure	Formula
Thymes	Thymus vulgaris L.	Twigs and leaves	 Thymol  Carvacrol	Thymol: $C_{10}H_{14}O$ Carvacrol: $C_{10}H_{14}O$
Ajwain/ Ajowan	Trachyspermum ammi	Fruits	 Thymol	Thymol: $C_{10}H_{14}O$
Clove	Syzygium aromaticum L.	Flower buds	 Eugenol	Eugenol: $C_{10}H_{12}O_2$
Lavender	Lavandula spica L.	Flowers without base	 Linalool  Eucalyptol (Cineole)	Linalool: $C_{10}H_{18}O$ Cineole: $C_{10}H_{18}O$
Espand/ Syrian rue	Peganum harmala L.	Seeds	 Vasicine (pegamine)  Harmaline	Vasicine: $C_{11}H_{12}N_2O$ Harmaline: $C_{13}H_{14}N_2O$
Wormwood	Artemisia absinthium L.	Flowers and aerial parts	 Thujone	Thujone: $C_{10}H_{16}O$
Marjoram	Origanum majorana L.	Flowered branches	 Terpineol	Terpineol: $C_{10}H_{18}O$
Aloe vera	Asparagales	Leaf		Anthraquinone $C_{15}H_{10}O_5$

Table 2. Dyeing conditions for woolen yarns

	Dye	Sample code	Goods	Id Auxiliary material 5%	Mordant 5%	Dye Co.%	L:R	Time (min.)
Meta Mordant	Thymes	T1	wool	Acetic acid	KAl $(SO_4)_2 \cdot 12H_2O$	50	1:40	60
	Ajwain	A1	wool	Acetic acid	KAl $(SO_4)_2 \cdot 12H_2O$	50	1:40	60
	Clove	C1	wool	Acetic acid	KAl $(SO_4)_2 \cdot 12H_2O$	50	1:40	60
	Lavender	L1	wool	Acetic acid	KAl $(SO_4)_2 \cdot 12H_2O$	50	1:40	60
	Esfand	P1	wool	Acetic acid	KAl $(SO_4)_2 \cdot 12H_2O$	50	1:40	60
	Wormwood	W1	wool	Acetic acid	KAl $(SO_4)_2 \cdot 12H_2O$	50	1:40	60
	Marjoram	M1	wool	Acetic acid	KAl $(SO_4)_2 \cdot 12H_2O$	50	1:40	60

Table 3. Dyeing conditions for cotton yarns

	Dye	Sample code	Goods	Auxiliary material 5%	Mordant5%	Dye Co.%	L:R	Time (min)
Meta Mordant	Thymes	T2	cotton	Na_2SO_4	KAl $(SO_4)_2 \cdot 12H_2O$	50	1:40	60
	Ajwain	A2	cotton	Na_2SO_4	KAl $(SO_4)_2 \cdot 12H_2O$	50	1:40	60
	Clove	C2	cotton	Na_2SO_4	KAl $(SO_4)_2 \cdot 12H_2O$	50	1:40	60
	Lavender	L2	cotton	Na_2SO_4	KAl $(SO_4)_2 \cdot 12H_2O$	50	1:40	60
	Esfand	P2	cotton	Na_2SO_4	KAl $(SO_4)_2 \cdot 12H_2O$	50	1:40	60
	Wormwood	W2	cotton	Na_2SO_4	KAl $(SO_4)_2 \cdot 12H_2O$	50	1:40	60
	Marjoram	M2	cotton	Na_2SO_4	KAl $(SO_4)_2 \cdot 12H_2O$	50	1:40	60

Colour Strength measurement

The reflectance and colour quality of dyed samples was measured using a Colour-eye 7000A spectrophotometer (X-rite, USA) under illuminant D65 and 10 o standard observer. Colour values (L *, a*, b *, K/S, C*, h o, λ max) and Relative colour strengths (K/S values) were determined by using the Kubelk and Munk equation (1) [31]:

$$K/S = \frac{(1-R)^2}{2R} \quad (1)$$

where, R is the decimal fraction of the reflectance of dyed woollen yarn, K is the absorption coefficient, and S is the scattering coefficient. Approximately K/S is directly related to the concentration of each dye in the samples.

Culture of Staphylococcus aureus, Escherichia coli and Candida albicans on textiles

The antibacterial property of mordanted and dyed wool & cotton yarns according to previous work was quantitatively evaluated according to AATCC 100-2004 and were tested in the Microbiology Laboratory of the University of Tehran [31].

After dyeing each sample wa sterilized under standard conditions before bacterial and microbial culture can be performed on woollen and cotton yarns or medicine plants. Sterilization has been done with C-type UV irradiation for cultures of bacteria or germs. The bacterial species used were: *E. coli* (Gram-negative), *S. aureus* (Gram-positive), and *C. albicans* microbe for cotton.

Antibacterial durability measurements

To determine the effectiveness and durability of the plants used to create antibacterial properties on wool and cotton products, the dyed yarns were subjected to standard washing conditions one and three times. The reason for choosing 1 and 3 cycles of washing is due to the common duration of hand-woven carpet washing cycles. In previous research, cotton as home textiles have been washed for up to 30 cycles, often for hospital sheets, but in handwoven carpets, the duration of washing cycles is longer [13,32]. In addition, to increase and have durable antibacterial properties, the cotton and wool yarn dyeing process was followed by a 30% weight of aloe vera plant. In the previous research aloe vera showed an improvement effect on the dyeing fastness in the natural dyeing process [34]. The conditions of the antibacterial durability measurements have been reported in Table 4.

Table 4. Conditions of antibacterial durability measurements

Sample / Code	1	2	3	4	5	6
T	Wool meta mordanting	Cotton meta mordanting	1 Washing Cycle	3 Washing Cycle	Code1+ 30% Aloe Vera	Code2+ 30% Aloe Vera
C	Wool meta mordanting	Cotton meta mordanting	1 Washing Cycle	3 Washing Cycle	Code1+ 30% Aloe Vera	Code2+ 30% Aloe Vera
M	Wool meta mordanting	Cotton meta mordanting	1 Washing Cycle	3 Washing Cycle	Code1+ 30% Aloe Vera	Code2+ 30% Aloe Vera
W	Wool meta mordanting	Cotton meta mordanting	1 Washing Cycle	3 Washing Cycle	Code1+ 30% Aloe Vera	Code2+ 30% Aloe Vera

Colour Fastness measurements

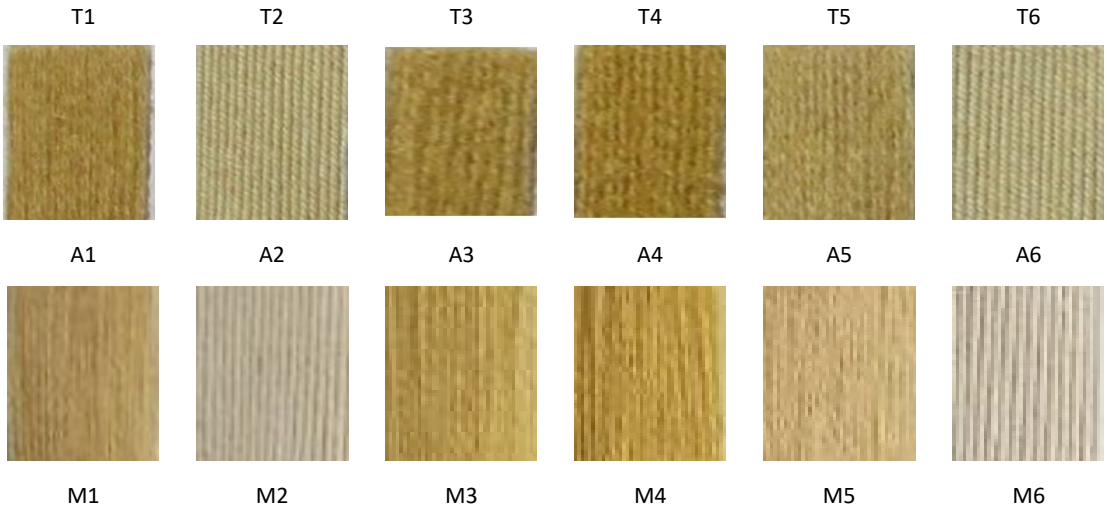
The dyed woollen and cotton yarns were tested according to ISO standard methods. The specific tests were as follows: ISO 105-C04 (2010) for measuring colour fastness to washing the extent of the staining samples was expressed in the appropriate staining grey scale and ISO 105- B02 for measuring colour fastness to light (xenon arc). Naturally dyed samples were exposed to sunlight for a maximum of 40 hours (8 hours period). After a time of light exposure, the samples were graded for colour fastness.

RESULTS

Dyeing properties

Figure 1 illustrates the dyeing results for the colour of woollen and cotton yarn dyed by selected medicinal plants. Wool yarns show a better affinity to dyes compared to cotton yarns. The colour coordinates of dyed wool were measured and colour properties were evaluated.

The colourimetric parameters such as L*, a*, b*, h°, C*, and colour strength (K/S) of the dyed wool before and after the washing process have been shown in Table 5. A high level of b* value is related to the deep yellow colour of samples; marjoram shows the highest yellowness. Also, the saturation of dye in this sample is more than in others. The washing process causes to lightness be reduced and yellowness level increased. This trend was observed in all samples. The standard amount of h° for yellow colour is equal to 90, the results of Table 5 show deep yellow colour for samples dyed with marjoram. The Colour reflectance of dyed wool yarn has been shown in Figure 2. The washing process has been changed on the reflectance percentage of samples. All four samples in Figures 2a and 2b, show a decrease in reflectance. It is in the agreement with the saturation value of these samples after dyeing.



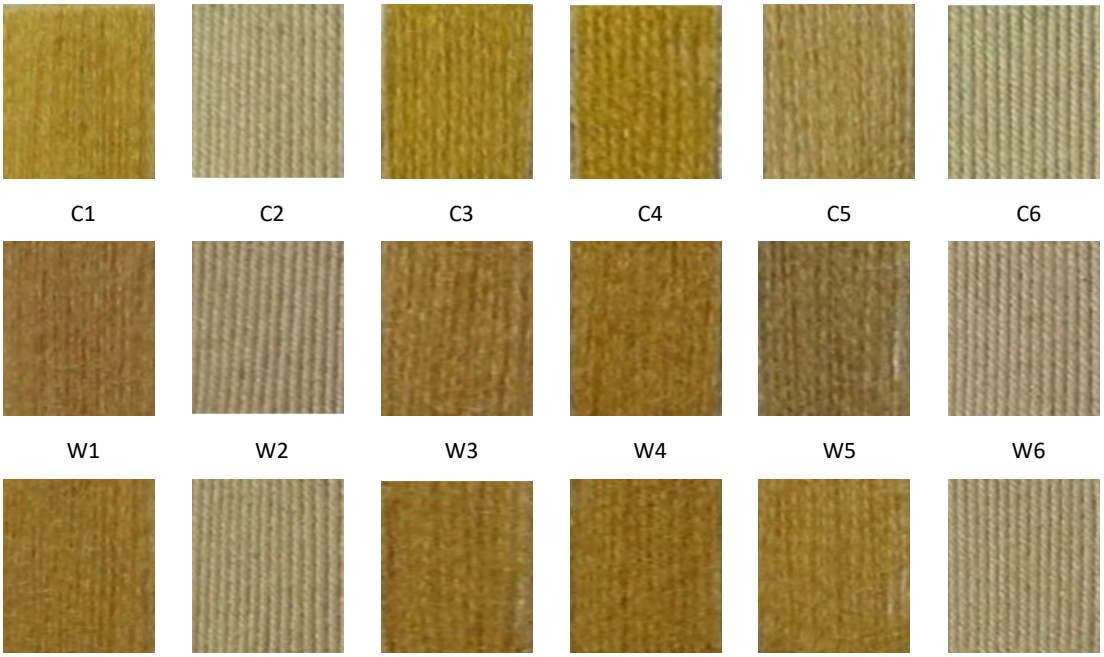
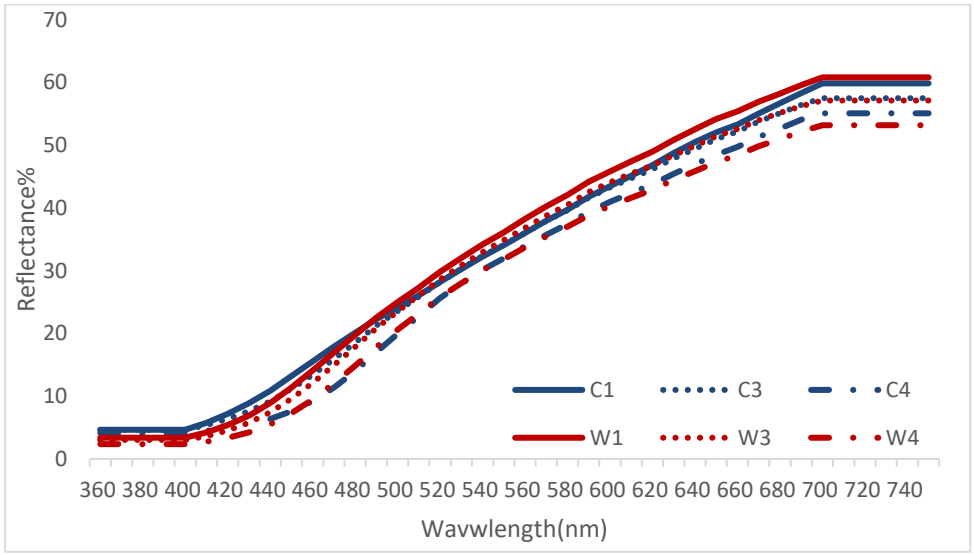


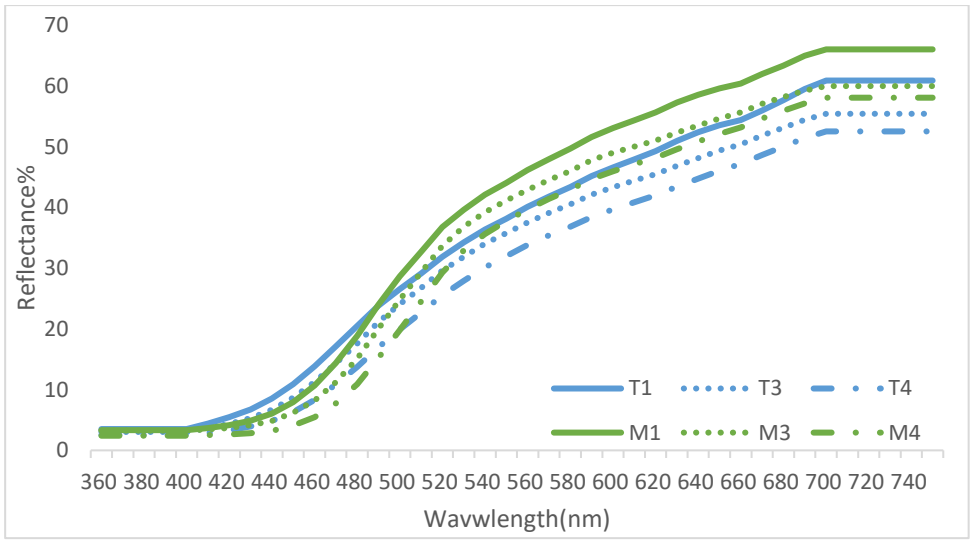
Figure 1. Colour of wool and cotton yarn dyed by plants

Table 5. Measurement of colour parameters for dyed wool yarns with thyme, marjoram, clove and wormwood

Sample	L*	a*	b*	C*	h°	k/s
T1	67.39	3.76	43.86	44.02	85.10	6.86
T3	65.3	3.25	46.33	46.45	85.99	7.43
T4	62.24	4.84	49.29	49.53	84.39	7.36
W1	66.35	5.91	41.94	42.35	81.98	6.98
W3	65.17	5.40	44.35	44.68	83.06	7.05
W4	62.56	5.39	46.97	47.28	83.45	7.23
M1	70.87	2.66	55.94	56.01	87.27	6.91
M3	68.43	2.70	58.12	58.18	87.34	7.55
M4	65.77	4.93	63.10	63.29	85.53	7.36
C1	65.03	6.84	36.07	36.72	79.27	6.88
C3	64.79	6.01	39.87	40.32	81.43	7.45
C4	62.68	6.73	46.27	46.75	81.72	7.13



(a)



(b)

Figure 2. Colour Reflectance of Woolen yarn dyed by medicine plants: (a) clove and wormwood; (b) thyme and marjoram

Antimicrobial activity of natural dyes

Since some natural medicine showed good antimicrobial activity against selected microbes in solution, the antimicrobial efficacy of a compound will vary when it is present in solution and when it is held intimately by a textile substrate. Therefore, the antibacterial activity of the origin plants was determined. The antibacterial effect of origin plants has been shown in Table 6. The powder of plants was used for the culture of both bacteria. The results prove that the selected plants could be effective against both gram-negative and gram-positive bacteria and prohibit their growth. It seems, in the future these plants will be used to produce various value-added products.

Table 6. Antibacterial properties of origin plants

Group	*Staphylococcus aureus*	*Escherichia coli*
A	6 × 10-2	> 6 × 10-2
ALV	> 6 × 10-2	> 6 × 10-2
C	> 6 × 10-2	> 6 × 10-2
L	6 × 10-2	6 × 10-2
M	> 6 × 10-2	> 6 × 10-2
P	6 × 10-2	6 × 10-2
T	> 6 × 10-2	> 6 × 10-2
W	6 × 10-2	> 6 × 10-2

An accurate quantitative evaluation of antimicrobial activity was possible on the yarn by colony counting and spectroscopic assessment. Table 7 and 8 show the number of two bacteria after 24 hours of incubation on the surface of undyed & mordanted and dyed yarns. Control samples show a nearly equal number of *E. coli* and *S. aureus* but after dyeing the antibacterial properties of yarns are lower than origin plants. Table 7 shows the antibacterial properties of dyed woollen yarns against both *S. aureus* (G+) and *E. coli* (G-) bacteria. It is obvious that with all dyes, the dyed wool has excellent antibacterial activity against *E. coli* bacteria used in this study. This activity is about ten times more than control samples in the case of *E. coli* bacteria. The antibacterial activity against *S. aureus* (G +) was found to be greater than that of *E. coli* (G-) which can be attributed to the differences in the chemical structure between the two types of bacteria.

Table 7. Results of antibacterial activity of dyed woollen yarns

Groups	*Escherichia Coli*	*Staphylococcus aureus*
Control	19×10^3	17×10^3
A_1	$< 10^2$	5×10^3
C_1	$< 10^2$	1×10^3
T_1	$< 10^2$	2×10^3
P_1	$< 10^2$	3×10^3
L_1	$< 10^2$	2×10^3
M_1	$< 10^2$	4×10^3
W_1	$< 10^2$	6×10^3

Control sample: undyed wool yarn

Samples dyed with clove, thymes, and lavender show the highest antibacterial activity level. In the cotton yarn as the reported result in Table 8 shows, marjoram and wormwood and lavender have better antibacterial activity than others. Naturally, cotton is more stable in *E. coli* growth than wool.

Results show better anti-bacterial properties in the woollen dyed yarn than in cotton as shown in Table 8. The samples treated with 7 natural dyes, wool and cotton, show very high activity with reduction of bacteria against *E. coli.*

Table 8. Results of antibacterial activity of dyed cotton yarns

Groups	*Candida albicans*	*Escherichia Coli*	*Staphylococcus aureus*
Control	23×10^3	6×10^3	9×10^3
A_2	7×10^3	$< 10^2$	8×10^3
C_2	8×10^3	$< 10^2$	6×10^3
T_2	11×10^3	$< 10^2$	4×10^3
P_2	16×10^3	$< 10^2$	6×10^3
L_2	14×10^3	$< 10^2$	5×10^3
M_2	8×10^3	$< 10^2$	2×10^3
W_2	14×10^3	$< 10^2$	2×10^3

Control sample-undyed cotton yarn

The durability of antibacterial and antimicrobial activity

According to a previous study, aloe vera as a bio-mordant was applied for increasing antibacterial and antimicrobial activity. A fresh leaf of aloe vera was used. The gel was gotten from the leaves into a clean container and was added to the dyeing bath directly. In other words, 30% (owf) of aloe vera was added to the dyeing bath of clove, thyme, marjoram, and wormwood. Also, the dyed samples were washed in different cycles to measure the durability of antibacterial and antimicrobial activity. The antimicrobial and antibacterial activity of the dyed samples and their potency were quantitatively assessed by the number of activity growth. Table 9 shows the durability of the antibacterial activity of the treated woollen yarns after 1 and 3 washing cycles.

Table 9. Durability of antibacterial activity dyed wool yarns after 1 and 3 washing cycles

Groups	*Escherichia coli*		*Staphylococcus aureus*	
Control	19×10^3		17×10^3	
	3	4	3	4
C	10×10^3	9×10^3	11×10^3	9×10^3
T	10×10^3	5×10^3	8×10^3	7×10^3
M	4×10^3	3×10^3	8×10^3	4×10^3
W	$< 10^2$	$< 10^2$	6×10^3	2×10^3

According to Table 4, code 3 refers to dyed wool yarns after 1-time washing, code 4 refers to dyed wool yarns after 3 times of washing

The efficacy of aloe vera against bacteria and microbe growth has been shown in Table 10 and 11. The ability of aloe vera to form true covalent bonds with wool yarn leads to an increase the antibacterial properties. Its ability on positive gram *S. aureus* is more than negative gram. In the case of wormwood and marjoram, the best antibacterial properties resulted.

Table 10. Bacteria growth reduction of dyed wool in the presence of aloe vera

Sample	*Escherichia Coli*		*Staphylococcus aureus*	
control	6×10^3		23×10^3	
C_5	5×10^3	17%↓	12×10^3	51%↓
T_5	3×10^3	50%↓	8×10^3	65%↓
M_5	$< 10^2$	>100%↓	4×10^3	83%↓
W_5	$< 10^2$	>100%↓	$< 10^2$	>100%↓

According to Table 4, code 5 refers to 30% aloe vera in the dyeing bath of wool yarns

Table 11. Bacteria and microbe growth reduction of dyed cotton in the presence of aloe vera

Sample	*Candida Albicans*		*Escherichia Coli*		*Staphylococcus aureus*	
control	9×10^3		6×10^3		23×10^3	
C_6	8×10^3	12%↓	4×10^3	67%↓	6×10^3	74%↓
T_6	11×10^3	50%↑	2×10^3	33%↓	4×10^3	83%↓
M_6	8×10^3	12%↓	$<10^2$	>100%↓	2×10^3	91.3%↓
W_6	14×10^3	16%↑	$<10^2$	>100%↓	2×10^3	91.3%↓

According to Table 4, code 6 refers to 30% aloe vera in the dyeing bath of cotton yarns

Dyeing Fastness

The fastness properties of the dyed woollen and cotton yarn by medicine plants are reported in Table 12. As the results show, cotton and wool yarns show good and excellent general fastness. Even, the three cycles of the washing process have not changed their fastness much. The presence of metal mordant and multiplicity of hydroxyl groups on the medicine plants and their cyclic structure may have led to the formation of more hydrogen bonds as well as covalent and van der Waals bonds with the wool and cotton yarns. The result of light fastness in the wool is due to a larger amount of dye being adsorbed onto the yarn. Using hand-woven carpets in high-traffic places such as religious centers, hotels, etc. accelerated the growth of bacteria and microbes. Woollen pile yarn of hand-woven carpet is dyed with natural dyes commonly, then it will be valuable that natural medicine plants used as dyes with anti-bacterial and anti-microbe properties.

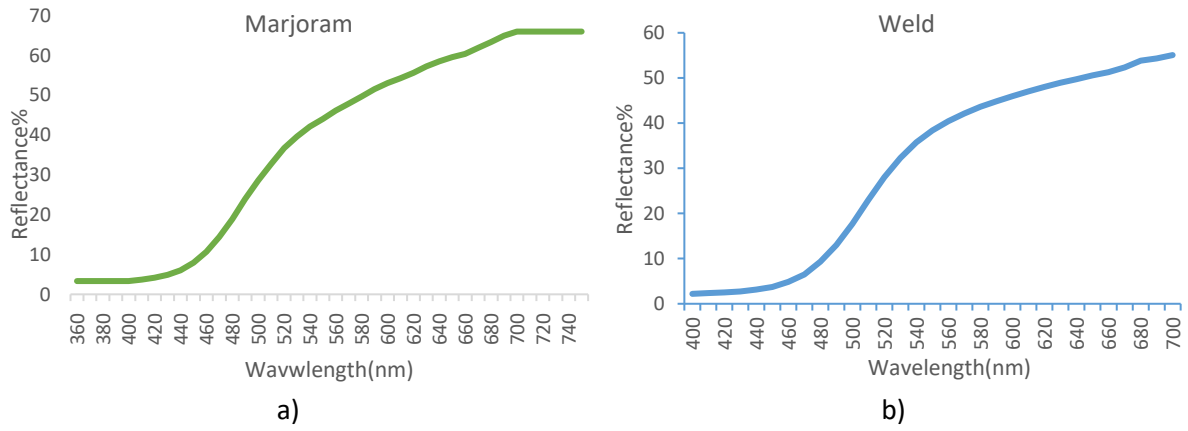
Table 12. Fastness properties of dyed wool & cotton yarn

Wool					Cotton				
Sample	Light fastness	Washing fastness	Stain on wool	Stain on cotton	Sample	Light fastness	Washing fastness	Stain on wool	Stain on cotton
A_1	7	5	5	5	A_2	6	4	5	5
C_1	7	4	5	5	C_2	6	5	5	4
T_1	7	4	5	5	T_2	6-7	5	5	5
P_1	7	5	5	5	P_2	6	4	5	5
L_1	7	5	5	5	L_2	6	5	5	5
M_1	7	5	5	5	M_2	6	4	5	4
W_1	7	5	5	5	W_2	6	4	5	5

DISCUSSION

Dyeing properties

It is understood from the colour of dyed yarn and colourimetric measurements, the dyeing properties of selected dyes are similar to usual natural dyes with yellow colour. The deep and brilliant yellow colour of these plants is similar to those one dyed with weld [34]. Reflectance measurements of marjoram have been compared to weld in Figure 3. As it is shown the spectra are nearly the same. All studied medicine plants produce good yellow colour on the woollen yarn. Washing caused the saturation level of dyes to increase in all samples. It seems that detergents in the washing process change the chemical structure of dyes. The Bathochromic effect causes the OH group in the dyes with anthraquinone chemical structure (thyme) to shift from α position to β position, then the darkness of the hue causes a reduction in lightness and variation of yellowness [35].



a) b)

Figure 3. Colour Reflectance of Woollen yarn dyed by marjoram (a) and weld (b)

Also, they showed good fastness according to the results in Table 12. Then it seems, that the selected medicine plants can dye textiles and are introduced as a natural source for dyeing.

Antibacterial properties

As the result in Table 7 showed lavender, thymes and clove had the highest antibacterial activity. It is obvious lavender, clove, and thymes due to their heterocyclic structure and position of OH groups linked to polypeptide bondage as shown in Figure 1, induced better antibacterial properties than others. This finding is in agreement with the results of references [36,10]. Carcavol and terpineol chemical structure has a simple structure that shares the OH group easily in the reaction with the cellulosic structure to inhibit bacteria growth. In the study, *Toddalia asiatica* extract showed the highest antimicrobial activity with inhibition zones ranging from 14 to 16 mm against both gram-positive and gram-negative organisms, which is lower when compared with *Araucaria columnaris* dye extract [37]. Methanolic extract bark showed maximum antibacterial activity compared to other extracts. It was reported that the methanolic extract manifested the highest antibacterial activity and ethyl acetate showed some degree of antibacterial activity. It is known that methanol is a better solvent for when compared with raw dye extract, dyed fabric showed low antibacterial activity; it may be due to the diffusion range or amount of dye used. As mentioned before, in the present study origin plants showed better antibacterial properties in the case of both *E. coli* and *S. aureus* than raw plants powder too. In the dying process, some parts of plants are present on the textile so, the lower efficacy of dyed goods against bacteria and microbe is related to the lack of those substances. Woollen-dyed yarn showed good activity against bacterial growth. There is no evidence of bacteria growth in the dish culture (Figure 4).

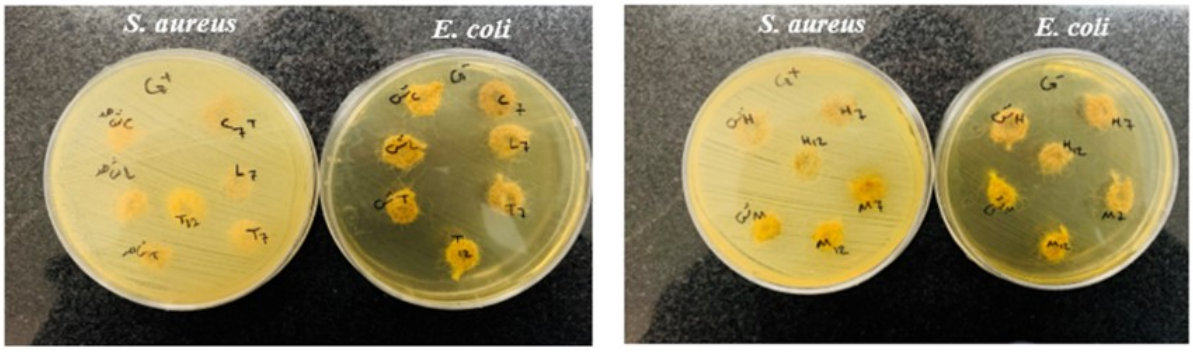


Figure 4. Antibacterial activity of the dyes on *Staphylococcus aureus* (G+) and *Escherichia coli* (G-) bacteria

The result in Table 9 and 10 shows that aloe vera improved the antibacterial activity in wool and cotton yarns. In the cotton samples, the synergism effect of aloe vera with wormwood and marjoram against both *E. coli* and *S. aureus*. The antimicrobial activity decreased in the presence of aloe vera gel. The

result of previous research proved our results, they observed the gel of aloe vera inhibited the growth of *Trichophyton mentagrophytes*, and the growth of *C. albicans* was also inhibited by aloe vera leaf but was not affected by the gel [38,39].

It can be seen from Table 9 that the antibacterial properties are not affected by repeated washing up to 3 washing cycles but in some cases, the antibacterial properties have been increased.

Antimicrobial properties are closely related to the dye structure, especially the presence of functional groups on it. Also, it can be seen that all dyes were completely effective against *E. coli*. This confirms that the chemistry of the dye and textile substrate has a significant influence on its antibacterial activity. Textile substrates change the antibacterial ability of plants. Cotton and wool, both, showed good properties against *E. coli* but in the case of *S. aureus*, each dye showed a different result. A schematic diagram of the interaction between aloe vera, metal mordant, wool, and medical dye (clove) has been shown in Figure 4. An increase in the number of hydrogen bonding, complex size, and coordination caused a positive effect of aloe vera in antibacterial activity. The metal ions can form coordinate complexes with amino groups of wool fibre. Some coordination sites remained unoccupied when they interacted with the fibre and can form complexes with dye molecules. This could be the consequence of the chelation of the dye by the metal salt. The only functional groups present in selected plants are hydroxy and it is well known that ortho hydroxy groups to the chromophore become involved in coordinate bond formation. It is therefore the formation of coordinate bonds that block the groupings responsible for antibacterial activity.

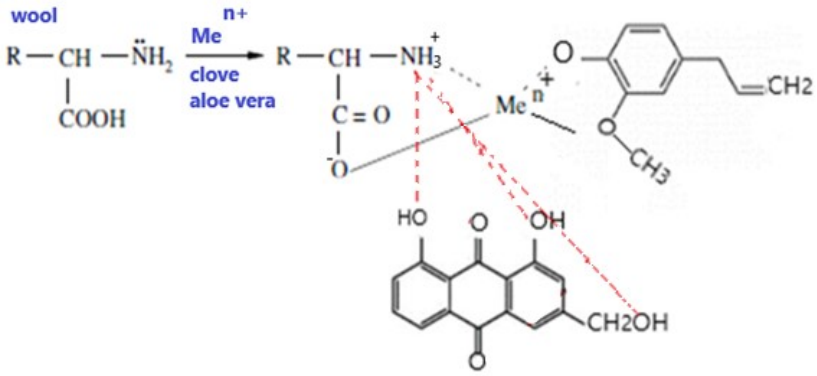


Figure 4. Probable interactions between a wool substrate, metal mordant, clove and aloe vera

CONCLUSION

The present study has revealed the importance of natural products to control microbe and bacteria growth on textiles. Herbs with medicinal properties that are widely used in Iran were tested for antibacterial and antimicrobial properties. Plants that have medicinal properties are often grown in most parts of Iran and are in hand. One of the important aims of this study was to use medicinal plant

compounds as a dye and investigation the antibacterial or antimicrobial activity of those plants on the textiles. Therefore, plants with this feature were used as natural dyes on both cotton and wool products. The results of the experiments showed that the studied plants can be used in dyeing wool to achieve a yellow colour. Besides the stable yellow hue, the washing fastness of the yarns was significant. Bacterial and microbial culture experiments on cotton and wool yarns dyed with medicine plants showed very good antimicrobial and antibacterial activities, the antibacterial activity remained unchanged until after 3 washing cycles too. The gel of aloe vera leaves increased the antibacterial activity but the antimicrobial properties of medicine plants decreased. Aloe vera, due to its polyphenolic structure by forming multiple bonds, improves antibacterial activity. Results showed that aloe vera could be exploited for new potent antibacterial agents and bio-mordant. Among the tested plants, marjoram and wormwood obtained the best results. The present study introduces the marjoram, clove, thymes, lavender, ajwain, and wormwood as natural dyes that could be effective against both *E. coli* and *S. aureus* bacteria on cotton and wool. Also, the waste of the dyeing process is biodegradable and can be utilized as excellent manure. These initial results have opened up an entirely new perspective of research, that medicinal plants whose properties have not been fully characterized should be paid attention to in developing green textiles. So, it seems in the future these plants will be developed to produce various value-added products.

Author Contributions

Both authors contributed to the article equally.

Conflicts of Interest

The authors declare no conflict of interest.

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