Journal of New Technology and Materials(JNTM)
J. New Technol. Mater.
Vol. 07, N°02 (2017)56-66

OEB Univ. Publish.Co.

Antibacterial effects and fastness properties of dyed silk fabric with curcumin in presence of organic and bio mordants

Reyhaneh Azarmi* and Ali Ashjaran
Young Researchers and Elite Club, Yadegar-e- Imam Khomeini (RAH) -Shahre-Rey Branch, Islamic Azad University, Tehran, Iran,post code 1815163111
***Corresponding authors, E-mail: reyhaneh.azarmi@yahoo.com**

Received date: Sep 14, 2017; accepted date: Dec 16, 2017

Abstract
Curcumin, a common natural dye used for fabric and food colorations, was used as an antimicrobial finish due to its bactericidal properties on dyed textiles.
In thiswork, silk fabrics were treated with curcumin andmordant with inorganic salt such as ferric sulfate, aluminum sulfate, potassium dichromate and copper sulfate and biological mordant such as chitosan. The strength color and color fastness to rubbing and washing of the dyed silk fabrics in presense of organic and bio mordantswere evaluated.Also antibacterial properties of treated silk fabrics against two kinds of bacteria: staphylococcus aurous and Escherichia coli were investigated.All mordant fabrics have good color strength compered to unmordantonces.The treated silk samples with chitosan in 0.6% concentrations were found to have good antibacterial potential.
Key words: Natural dye, Mordants, colour fastness, silk, Curcumin.

1.Introduction

Dyeing is one of the oldest techniques of human civilization. People have dyed the textile materials since thousands of years and most of the times the dyes have come from nature [1]. The use of natural dyes has increased after the development of weaving technique for dyeing textile materials [2]. Curcumin or diferuloylmethane with chemical formula of (1,7-bis(4-hydroxy-3-methoxyphenyl)-1,6-heptadiene-3,5- dione) (Figure 1) and other curcuminoids constitute the main phytochemicals of Curcuma longa L. (Zingiberaceaefamily) rhizome with the common name of turmeric [3]. Curcuma longa rhizome has been traditionally used asantimicrobial agent as well as an insect repellant [4]. Several studies have reported the broad-spectrum antimicrobial activity for curcumin including antibacterial, antiviral, antifungal, and antimalarial activities.

Silk fiber can be used in many products such as precious fabrics, parachutes, tire lining materials,
artificial blood vessels, and surgical sutures. properties of silk fabric such as wash and wear properties, dye-ability, and color fastness are weak and they should be improved [5]. For this purpose, Modification of silk by some physical and chemical techniques such as mordanting and surface modification have been developed.

Curcumin (diferuloylmethane) is a bright yellow chemical produced by some plants and its polyphenolic compound curcumin have been subjected to a variety of antimicrobial investigations due to extensive traditional uses
and low side effects[5-7].

The most common applications are as a dietary supplement, incosmetics, as a food coloring and as flavoring for foods such as turmeric-flavored beverages[8].Chemically, aromatic ring systems, which are phenols, are connected by two α,β-unsaturated carbonyl groups [9].

The diketones form stable enols and are readily deprotonated to form enolates; the α,β-unsaturated carbonyl group is a good Michael acceptor and undergoes nucleophilic addition.Itis a tautomeric compound existing in enolic form in organic solvents and as a keto form in water [10-14].

Figure 1 shows the picture of curcumin dye and Figure 2 shows chemical structure of this dyes. Antimicrobial activities for curcumin with tpye of mordants against different bacteria have been reported[15-17].However, natural colors on the fabric have a low fastness. Features including the ability to form complexes with metal ions in natural dyes such as chromium ions, iron, copper, aluminum[18]. Other researches describe an encapsulation process that improves this property using a polyrhodanine shell material synthesized by oxidative polymerization. Unlike the previous methods of oxidativepolymerization we used the Fenton reaction to introduce a $FeCl_3/H_2O_2$ combination system. And the other study effect of curcumin on the production of hydroxyel radicals throught the Fenoton reactions has been studied. This study shows that curcumin can reduce ferric ion to ferrous and this ferrous ion can generate a hydroxyl radical in the presence of hydrogen peroxide through the Fenton reaction. Thus by keeping in view of above, the present study has been focused on the dyeing and antibacterial properties and surface modification of

the silk fabrics treated and mordant with inorganic salt and bio mordant which dyed with curcumin as a natural dye[19].

Figure 1. Curcumin dye

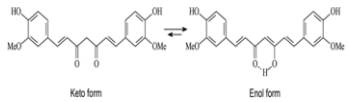


Figure 2. Chemical structure of curcumin

2. Material

Scoured silk fabric (100%silk, 30 denier, Warp and Weft Respectively 71 and 67) was purchased from Simin Company. Curcumin seeds were selected as a natural dye source to color silk fabric. Turmeric i.e. Curcuma Longla L. is used to obtain the dyestuff of curcumin, which is yellow in color. Turmeric is collected from the local market of Bangladesh. The followingmordants are used: Aluminum sulfate ($Al_2(so_4).18\ H_20$), ferric sulfate ($Fe\ so_4 .7\ H_2o$),potassium dichromate ($k_2cr_2o_7$) and copper sulfate ($Cu\ (So_4).5\ H_2o$) and bio mordant such as chitosan. They were pure grade chemicals used for laboratory purpose and Collected from Carl Roth GmbH, Germany.

3. Methods

In table 1 silks with different concentrations chitosan and mordant have been reported with symptoms.

Table 1. Symptoms of samples

Coding of samples	Name of samples
$M_{Fe}.D$	Silk mordant with ferric sulfate
$M_{Cu}.D$	Silk mordant with copper sulfate
$M_{Al}.D$	Silk mordant with aluminum sulfate
$M_{Cr}.D$	Silk mordant potassium di chromate
$M_{Fe}.C_{0/1\%}.D$	Silk mordant with ferric sulfate and then finished with 0.1% chitosan
$M_{Cu}.C_{0/1\%}.D$	Silk mordant with copper sulfate and then finished with 0.1% chitosan
$M_{Al}.C_{0/1\%}.D$	Silk mordant with aluminum sulfate and then finished with 0.1% chitosan
$M_{Cr}.C_{0/1\%}.D$	Silk mordant with potassium di chromate and then finished with 0.1% chitosan
$M_{Fe}.C_{0/6\%}.D$	Silk mordant with ferric sulfate and then finished with 0.6% chitosan
$M_{Cu}.C_{0/6\%}.D$	Silk mordant with copper sulfate and then finished with 0.6% chitosan
$M_{Al}.C_{0/6\%}.D$	Silk mordant with aluminum sulfate and then finished with 0.6% chitosan
$M_{Cr}.C_{0/6\%}.D$	Silk mordant with potassium di chromate and then finished with 0.6% chitosan
$C_{0/1\%}.\ M_{Fe}.D$	Silk finished with 0.1% chitosan and then mordant with ferric sulfate
$C_{0/1\%}.\ M_{Cu}.D$	Silk finished with 0.1% chitosan and then mordant with copper sulfate
$C_{0/1\%}.\ M_{Al}.D$	Silk finished with 0.1% chitosan and then mordant with aluminum sulfate
$C_{0/1\%}\ .M_{Cr}.D$	Silk finished with 0.1% chitosan and then mordant with potassium di chromate
$C_{0/6\%}.M_{Fe}.\ D$	Silk finished with 0.6% chitosan and then mordant with ferric sulfate
$C_{0/6\%}.M_{Cu}.D$	Silk finished with 0.6% chitosan and then mordant with copper sulfate
$C_{0/6\%}.M_{Al}.D$	Silk finished with 0.6% chitosan and then mordant with aluminum sulfate
$C_{0/6\%}.M_{Cr}.D$	Silk finished with 0.6% chitosan and then mordant with potassium di chromate
R_f	Sample of row fabric

3.1. Mordanting

Fabric was mordanted prior to dyeing by treating with aluminum sulfate, ferricsulfate, potassium dichromate and copper sulfatemordant at boil for 45 minutes. And some fabrics finished with 0.1 and 0.6 % oconcentration with chitosan. Theliquor ratio is 1:40 and mordants concentration were 5% on weight of the fabric. Aftermordanting allfabrics were squeezed.

3.2. Dyeing

Finished and unfinished fabrics were dyed with curcumi dye.The dye baths were prepared by adding dye (25 ml), to distilled water (15ml) at temperature 40° C.Wet fabricswere added to dye-baths and then temperature was raised to boiling temperature
at 2.5°C /min, and dyeing was continued at boiling temperature for 15 minutes and thenadded acetic acid and again continued at boiling temperature for 15 minutes.

3.3. Determination of Color Strength

Reflectance values of the treated and dyedfabrics were measured using reflectancespectrophotometer(X-rite, color Eye 1000 A,America).
Relative colour strengths (K/S values) were determined using the Kubelka–Munk equation.

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

Where, K and S are the absorption and scattering co-efficient of the sample. R is the absolute reflectance.

3.4. Assesment of color fastness

Rubbing and washing fastness of treated and dyed fabrics were measured by using ISO 15 X12 1993 and 15 c.1998to deetermine the chang and stain in fabrics after test.

3.5. Antibacterial test

Antibacterial activity against Gram-positive bacteria (S. aureus) and Gram-negative bacteria (E. coli) was tested quantitatively by AATCCTest Method 100-147. The number of viable bacterial colonies on the agar plate before and after dyeing was counted and the results reported as percentages of bacteria reduction according to:

$$(R = \frac{A-B}{B} \times 100) \quad (2)$$

where R notes the percentage of reduction of microbial population; B is the absorbance of the media inoculated with microbes and un-dyed fabric and A shows the absorbance of the media inoculated with microbes and dyed fabric.

3.6. Scanning Electron Microscopy (SEM) and Transmission Electron Microscopy.

For SEM observation, a thinlm of cells was smeared on a silver stub. samples were gold-covered by curcumin. Morphology was observed on a scanning electronic microscope (ZEISSEVO 50). SEM observation was done under the followinganalytical condition: EHT = 20.00 kV, WD = 9.5 mm, andSignal A = SE1.

4. Results and Discussion

4.1 Effect of mordanting on color strength of silk fabrics dyed with curcumin

The effects of mordant on relative color strength for the premordanting method are shown in figure3-7. It is observed that
relative color strength values are higher for silk fabrics dyed
with curcumin and $Fe_2 SO_4$. After this sample respectively Cu_2SO_4, $k_2 cr_2O_7$
And $Al_2 SO_4$,have most to less color strength. It is also seen that different mordantsare different color strength of silk fabric that are not same effective.

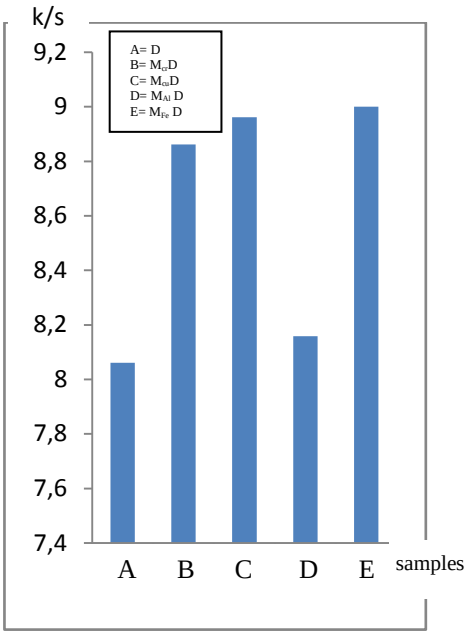


Figure 3. Comparison of k/sof$M_{(Fe,Cu,Al,Cr)}$.D,D

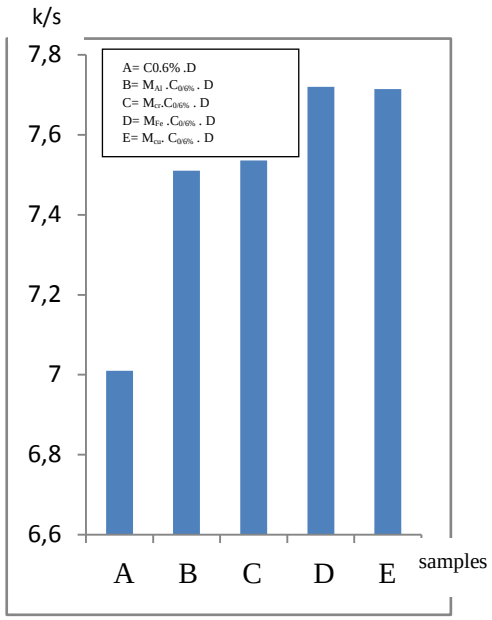


Figure 4. Comparison of k/sof $M_{(Fe,Cu,Al,Cr)}$. $C_{0.6}$D, $C_{0.6}$D

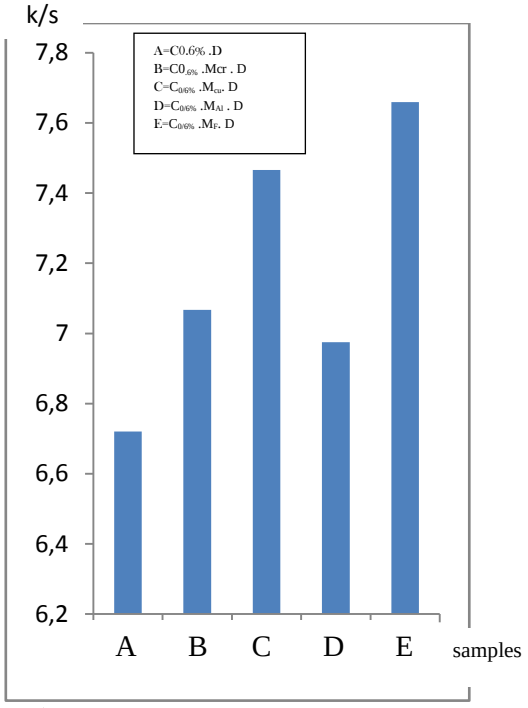


Figure 5. Comparison of k/sof $C_{0.6}M_{(Fe,Cu,Al,Cr)}$. D, $C_{0.6}$D

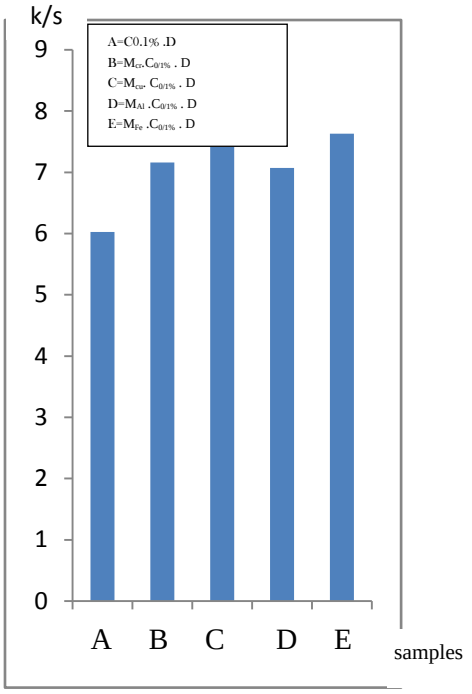


Figure 6. Comparison of k/S of$M_{(Fe,Cu,Al,Cr)}$.$C_{0.1}$D, $C_{0.1}$D

k/s

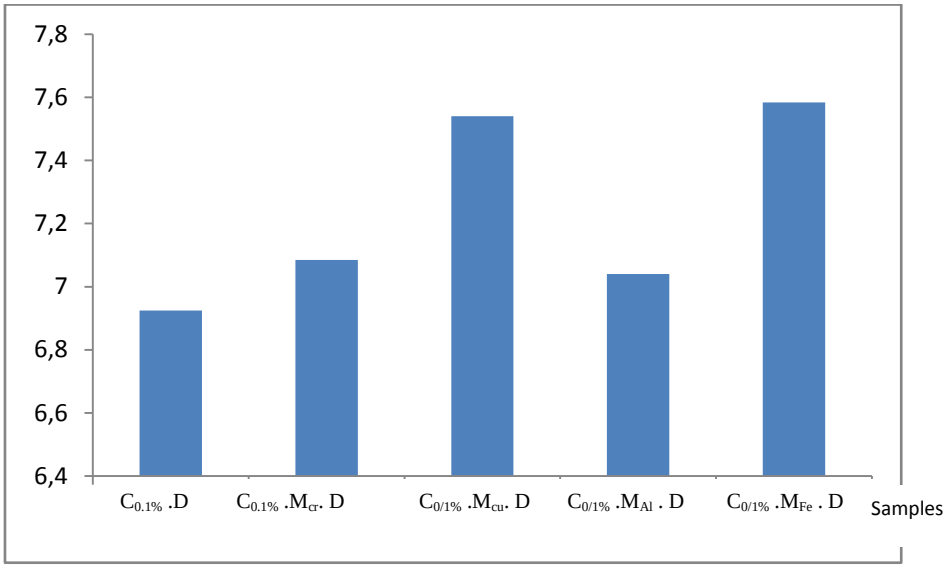


Figure 7. Comparison of k/s of $C_{0.1}M_{(Fe,Cu,Al,Cr)}$. D, $C_{0.1}$D

4.2. Result of fastness properties

Wash fastness of the samples dyed under the optimized conditions is tested according to ISO 105-CO_3 method. The wash fastness rating of silk fabrics dyed with curcumin dye with or without mordants and chitosan at the
concentration of 1% and 6% (owf) is presented in table 2. The results show that washing fastness of the model is not good without using the mordant. Rubbing fastness of the dyed model with curcumin improved after using the mordant. Also, according to the obtained results from rubbing fastness, we can find out that the mordants plus the dyes have the best rubbing fastness. Because, the mordant sticks the fiber directly and mordants are able to constitute metal complex (dative bond) with some active groups of fiber and curcumin dye groups which results in the increase of stability characteristics. In chitosan link models, a part of color might stick the chitosan. As a result, due to lack of dative bond, it quickly separates the fiber and is connected to a silky fabric. Iron sulfate mordant has the best rubbing and washing stability.

Table 2. fastness properties

Samples	Rubbing fatness		Washing fatness
	Dry	wet	
M_{Fe}.D	5	5	5
M_{Fe}.$C_{0/1\%}$.D	4-5	4-5	5
$C_{0/1\%}$. M_{Fe}.D	4-5	4-5	4-5
M_{Fe}.$C_{0/6\%}$.D	4-5	4-5	4-5
$C_{0/6\%}$.M_{Fe}. D	4-5	4-5	4-5
M_{Cu}.D	4-5	4	4
M_{Cu}.$C_{0/1\%}$.D	4-5	4	4
$C_{0/1\%}$. M_{Cu}.D	4	4	3-4
M_{Cu}.$C_{0/6\%}$.D	4	4	3-4
$C_{0/6\%}$.M_{Cu}.D	4	4	3-4
M_{Cr}.D	4	4	4
M_{Cr}.$C_{0/1\%}$.D	4	4	4

$C_{0/1\%}$ .M_{Cr}.D	4	4	4
M_{Cr}.$C_{0/6\%}$.D	3-4	3-4	3-4
$C_{0/6\%}$.M_{Cr}.D	3-4	3-4	3-4
M_{Al}.D	3-4	3-4	3-4
M_{Al}.$C_{0/1\%}$.D	3-4	3-4	3
$C_{0/1\%}$. M_{Al}.D	3	3	3
M_{Al}.$C_{0/6\%}$.D	3	3	3
$C_{0/6\%}$.M_{Al}.D	3	3	3

4.3. Antibacterial activity

Antimicrobial evaluation of silky fabrics in the presence and absence of chitosan and different mordants have been mentioned in the following table. The results of antimicrobial quantitative experiment based on AATCC100 standard showed that respectively, M.$C_{0.6\%}$.D, $C_{0.6\%}$.M.D , M $C_{0.1\%}$D, , $C_{0.1\%}$.M.D models had antimicrobial trait from more to less. Also, the results show that D fabrics and also M D fabric don't show acceptable antimicrobial effect from themselves. Also, M $_{(Fe,\ Cu,Al,Cr)}$.$C_{0.6\%}$.D has more antimicrobial property as compared to the other models.

Table 3. Decrease of bacteria in samples of $M_{(Fe,Cu,Al,Cr)}$.$C_{0/6\%}$D, $C_{0/6\%}M_{(Fe,Cu,Al,Cr)}$.D

$M_{(Fe,Cu,Al,Cr)}$.$C_{0/1\%}$D,.$C_{0/1\%}M_{(Fe,Cu,Al,Cr)}$. against bacteria Escherichia coli

Sample names	Pre-washing antibacterial percent	After washing antibacterial percen
M_{cu}.$C_{0/6\%}$D	100	99/9
M_{fe}.$C_{0/6\%}$D	100	99
M_{Al}.$C_{0/6\%}$D	98	94.5
M_{cr}.$C_{0/6\%}$D	100	93.5
$C_{0/6\%}$ M_{cu}.D	99	99.3
$C_{0/6\%}$ M_{Fe} .D	99/99	98
$C_{0/6\%}$ M_{Al} .D	95	89.25
$C_{0/6\%}$ Mcr .D	99.9	87
M_{cu}.$C_{0/1\%}$D	98	71
M_{Fe}.$C_{0/1\%}$D	93/9	70
M_{Al}.$C_{0/1\%}$D	88	68
M_{cr}.$C_{0/1\%}$D	86.1	67
$C_{0/1\%}$ M_{cu} .D	92.5	58.4
$C_{0/1\%}$ M_{Fe} .D	90	55
$C_{0/1\%}$ M_{Al} .D	87	38.8
$C_{0/1\%}$ M_{cr}.D	84	36
U	0	0

Table 4. Decrease of bacteria in samples $M_{(Fe,Cu,Al,Cr)}.C_{0/6\%}.D$, $C_{0/6\%}.M_{(Fe,Cu,Al,Cr)}.D$,$M_{(Fe,Cu,Al,Cr)}.C_{0/1\%}.D$,.$C_{0/1\%}.M_{(Fe,Cu,Al,Cr)}.D$ against bacteria Staphylococcus aureus

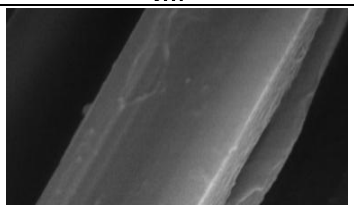
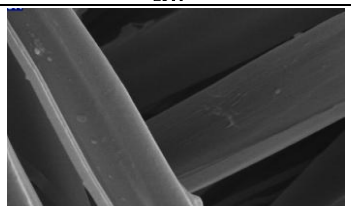
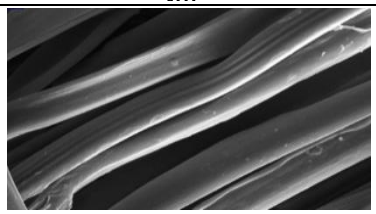
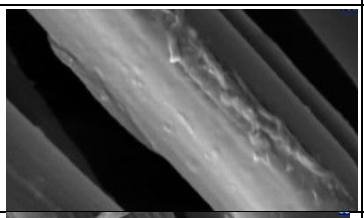
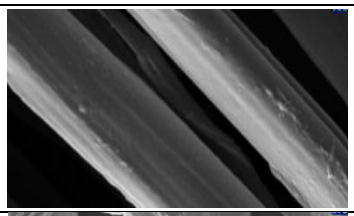
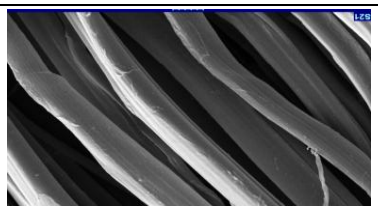
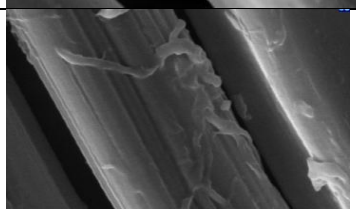
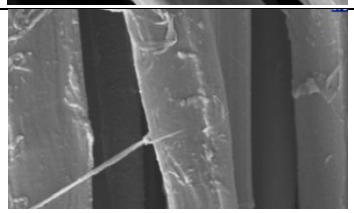
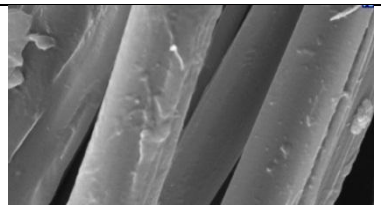
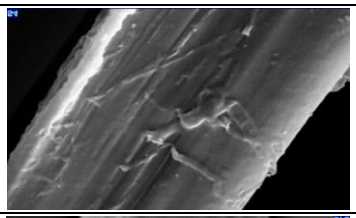
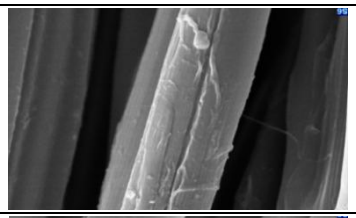
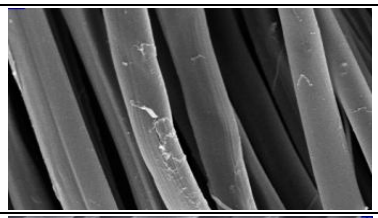
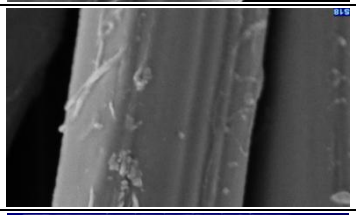
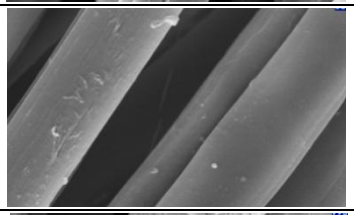
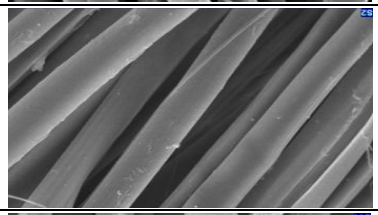
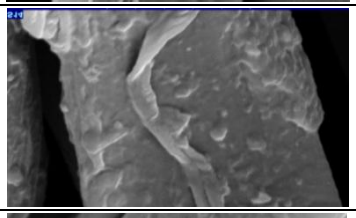
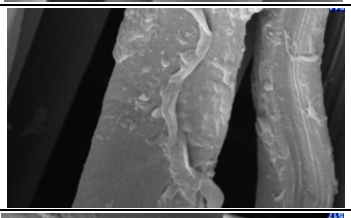
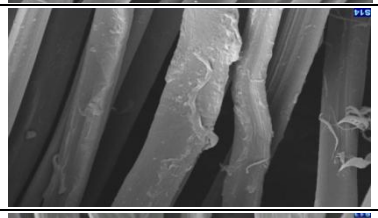
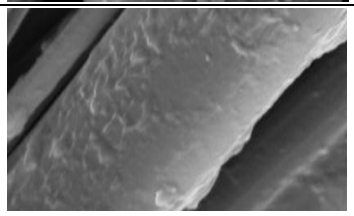
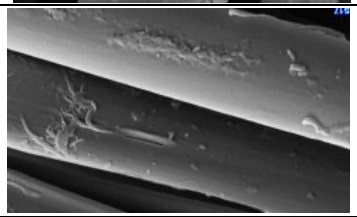
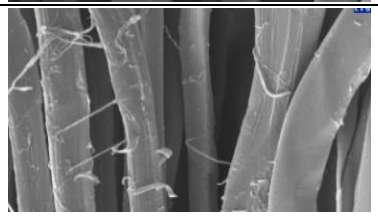
Sample names	Pre-washing antibacterial percent	After washing antibacterial percent
$M_{cu}.C_{0/6\%}.D$	99.9	90
$M_{fe}.C_{0/6\%}.D$	99	86.9
$M_{Al}.C_{0/6\%}.D$	98.9	69
$M_{cr}.C_{0/6\%}.D$	96.4	71.8
$C_{0/6\%}.M_{cu}.D$	99	89
$C_{0/6\%}.M_{Fe}.D$	96.4	80.9
$C_{0/6\%}.M_{Al}.D$	93	68
$C_{0/6\%}.M_{cr}.D$	89.4	71.4
$M_{cu}.C_{0/1\%}.D$	63	0
$M_{Fe}.C_{0/1\%}.D$	61	0
$M_{Al}.C_{0/1\%}.D$	56.8	0
$M_{cr}.C_{0/1\%}.D$	55.3	0
$C_{0/1\%}.M_{cu}.D$	50.2	0
$C_{0/1\%}.M_{Fe}.D$	45.3	0
$C_{0/1\%}.M_{Al}.D$	39.6	0
$C_{0/1\%}.M_{cr}.D$	0	0
U	0	0

Also, the results showed that copper sulfate mordant had the most antimicrobial property. After this mordant; respectively, iron sulfate, Aluminum sulfate and potassium dichromate and will have the most to least antimicrobial effect.

4.4.Morphologies of silk fabrics

The SEM of untreated, dyeing fabrics, chitosan (0.1, 0.6 conce) treated and simultaneous chitosan (0.1, 0.6 conce) with mordant treated fabrics followed by a dyeing process using curcumin dye are shown in table1. Fabrics with out treated chitosan showed smooth and clean surface, while treated fabrics with chitosan in concentration 0.1% and 0.6% have got the polymeric chains that relatives chitosan concentrations

Table 5. Morphologies of silk fabrics with chitosan and mordants

5000*	2500*	1000*	sample
			R_f
			$C_{0.1\%}$
			$C_{0/6\%}$
			$M.C_{0/1\%}.D$
			$C_{0/1\%}.M\ D$
			$M.C_{0/6\%}.D$
			$C_{0/6\%}.M.D$

According to the table 4, $M.C_{0.6\%}.D$ and $C_{0.6\%}.M.D$ have more polymeric chains on the surface of fiber which have increased as a result of antimicrobial effect and by means of chitosan. Therefore; according to the tables, we generally can conclude that the samples without chitosan actions have smooth and almost monotonous surface; whereas in other chitosan link models, placement of polymeric chains and chitosan swell are observed. Chitosan has created rough surface on fiber surface.

4-5-Result of IR spectroscopy

Raw silk samples in the presence of 0.1 and 0.6 percent chitosan have been mentioned in the following charts. Evaluation of infrared spectrum is reinforced and NH_2 is compared. It is observed that messenger is related to $C_{0.6\%}$ and $C_{0.1\%}$ with R_f sample, when the model almost inthree OH models strengthens the silk threads. But,in NH2 which is from NH2 groupis different in raw with C0.6% sample,in raw and Co.1 samples absorption band in bending is around 3276but in $C_{\%0.6}$ sample this absorption band is in3278.37, it is observed that in this NH2 sample messenger, some amounts of this absorption band has been reinforced.

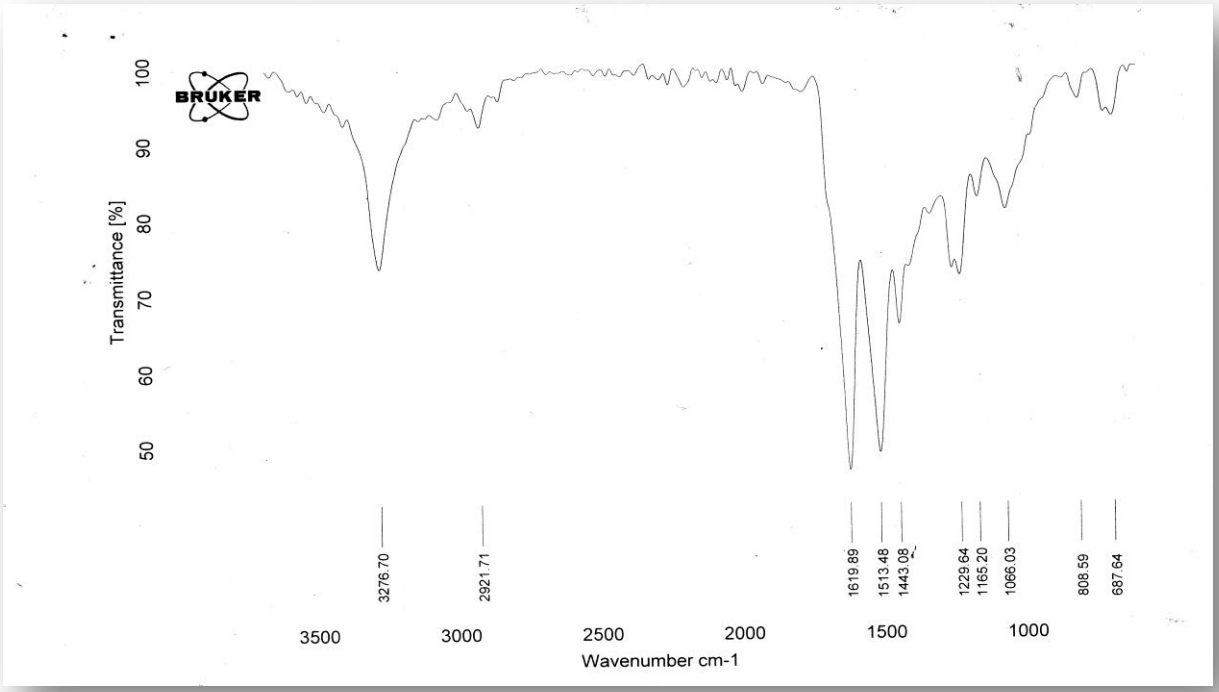


Figure 8. FTIR of raw sample

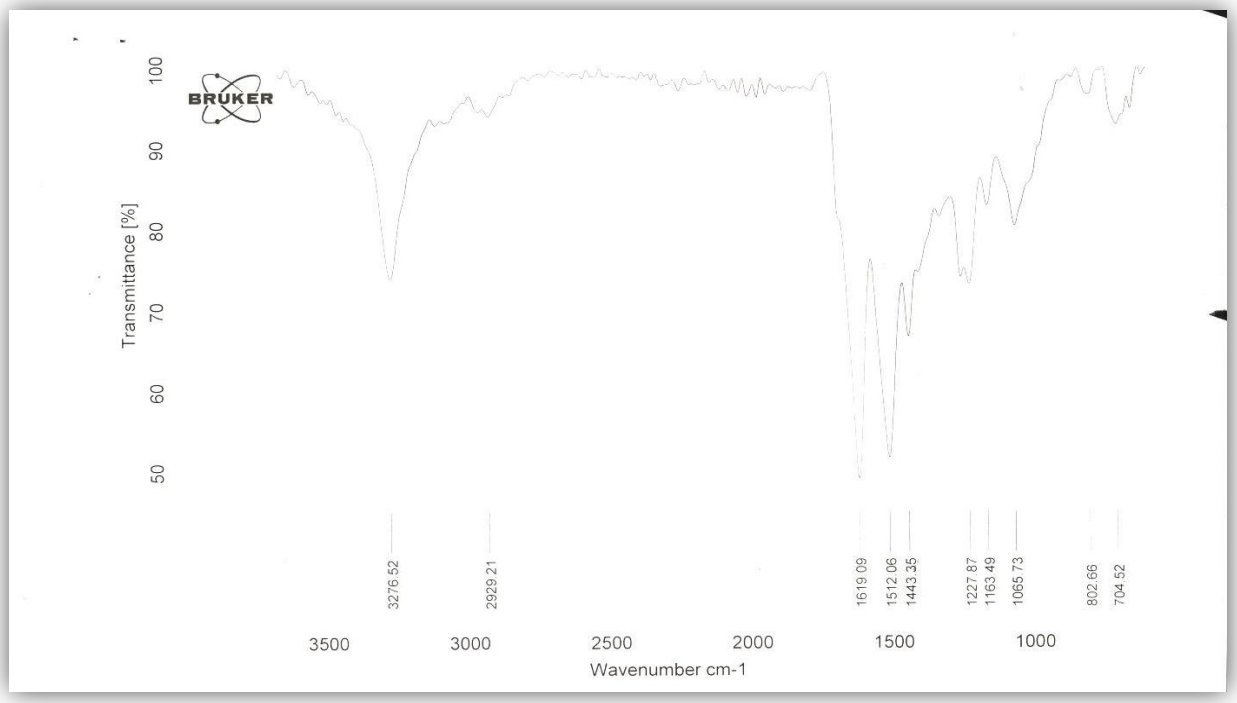


Figure 9. FTIR of samples $M_{(Fe,Cu,Al,Cr)}.C_{0/1\%}.D$ and $C_{0/1\%}.M_{(Fe,Cu,Al,Cr)}.D$

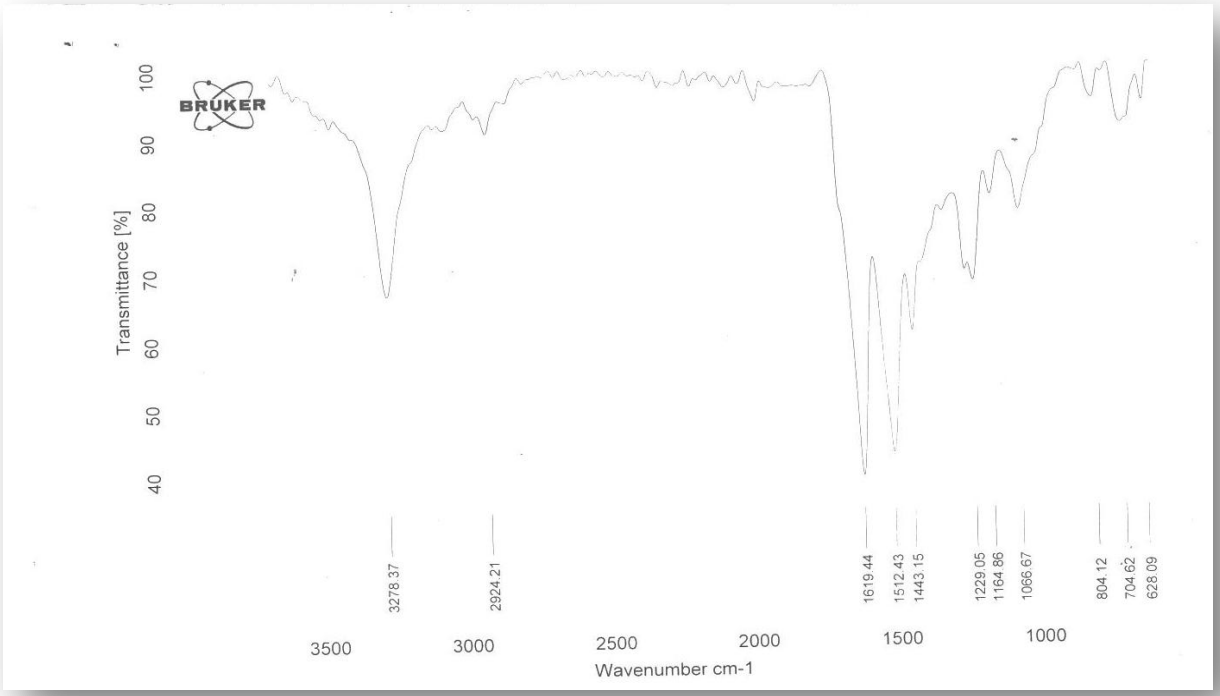


Figure10. FTIR of $M_{(Fe,Cu,Al,Cr)}.C_{0/6\%}.D$ and $C_{0/6\%}.M_{(Fe,Cu,Al,Cr)}.D$

5. Conclusion

This paper observed that relative color strength values are higher for silk fabrics dyed with curcumin and Fe_2SO_4. Also evaluates stability characteristics and antimicrobial effects of dyed silk fabric with curcumin in the presence and absence of mordants and chitosan. The results of antimicrobial tests show that $M.C_{0.6\%}.D$ has the best antimicrobial property. Because, the number of colonies in this model has decreased to a large amount before and after washing. The experiments related to the views of scanning electron microscope also showed that $C_{0.6\%}.M.D$ and $M.C_{0.6\%}.D$ had a large amount of chitosan on fiber surface, and as a result, their antimicrobial property increased. Also, according to washing and rubbing fastness experiments, we can find out that iron sulfate mordant has the best washing and rubbing fastness.FTIR analysis showed evidence that improved NH and NH_2 in $C_{0.6\%}$.

Acknowledgements

Many results presented here wer obtained in the frame supported by Young Researchers and Elite Club.

Refrences

[1] Z.Amar,B. Saber, A. Yasmine,G. Noureddine, J. New Technology and Materials. 01 (2014) 155-160
[2] H. Ouled-Haddar, M. Sifour, T. Idoui, H. Bouridane , S. Arid, J. New Technology and Materials. 01(2013) 37-42
[3] H. P. T. Ammon, M. A. Wahl, J. Planta Medica, Germany.12 (3) 2001, 1-7.
[4] S. K anhathaisong, S. R attanaphani, V. R attanaphani, T. Manyum , J. Sci. Technology. 13(2011) 159-165
[5]N. Babaii, S.h.Zamaninejad, J .Dent Mater Tech. 5(2016) 196-201
[6] O.A.K. Khalil, O. M. M. FariaOliveira, J. C. R. Vellosa,J. Food. Chemistry. Sci. Elsevier, Amsterdam. 133(2012) 1001-1005.
[7] K. Vimala, Y. M. Mohan, K. Varaprasad, J. Biomaterials andNanobiotechnology. 2(2011) 55-64.
[8] M. Karaman, F. Fırıncı, Z. ArıkanAyyıldız, I. H. Bahar, Mikrobiy.Bulteni,.J.Curr.Microbiol. 47(2013) 192-194.
[9] S. A.Marathe, S.Ray, D.Chakravortty, J.pone. 5(2010) e11511.
[10] L. Tomei, S. Altamura, G. Paonessa, R. De Francesco, G.Migliaccio, J. International Medical Press.16(2005) 225-245.
[11] M. Lemoine, S. Nayagam, M. Thursz, J. Future. Viro. 8(2013) 65-74.

[12] R. Singh, A. Jain, S. Panwar, D. Gupta, and S. K. Khare,J.DyesandPigment,ELSEVIER. 66(2005) 67-70
[13] F. C. Odds, J.Antimicrobial Chemotherapy.53(2003) 1.
[14] K. M. Moghaddam, M. Iranshahi, M. C. Yazdi, A. R. Shahverdi,J. International . Green Pharmacy.3(2009) 141-143.
[15] D. M. Parkin, F. Bray, J. Ferlay, and P. Pisani, J.A Cancer for Clinicians. 55 (2005) 74-108.
[16] G. Grynkiewicz, P. Slifirski, J. ctabiochimicaPolonica. 59(2012) 1-11.
[17] R. Singh, A. Jain, S. Panwar, D. Gupta, S. K. Khare,J.Fibre& Textile Research. 34 (2009)295-304.
[18] Kumar V & Bharti B V ,JIndian Text . 2 (1998) 18 .
[19] H. C. Tiwari, P. Singh, P. K .Mishra, P. Srivastava, J.Indian Fibre& Text Res. 35 (2010) 272.