BTCA/Nano TiO_2 Synergism on Cotton: Enhanced Antibacterial Features Optimized by Statistical Models

Ali Nazari[1], Majid Montazer[2], Mohammad Bameni Moghadam[3], Mohammad Esmail Yazdanshenas[4]

[1]Department of Art & Architectural, Yazd Branch, Islamic Azad University, Yazd, IRAN

[2]Department of Textile Engineering, Amirkabir University of Technology (Tehran Polytechnic), Tehran, IRAN

[3]Department of Statistics, Allameh Tabatabai University, Tehran, IRAN

[4]Department of Textile, Yazd Branch, Islamic Azad University, Yazd, IRAN

Correspondence to:
Ali Nazari email: a.nazari@iauyazd.ac.ir

ABSTRACT
The application of Butane tetra carboxylic acid/nano titanium dioxide [(BTCA)/Nano TiO_2 (NTO)] on cotton fabric has been used to cross-linked cellulose, and yield higher NTO absorption and enhanced self-cleaning properties. In this research, the synergistic effect of BTCA and NTO on antibacterial properties of the cotton fabric was investigated. The NTO particles were stabilized on the cotton surface using BTCA. Then, different curing conditions (UV, Temp, and UV-Temp) were examined. The treated fabrics indicated an antibacterial activity against both *Staphylococcus aureus* and *Escherichia coli* bacteria. The role of both BTCA and NTO concentrations on antibacterial properties of the cotton fabric were investigated using response surface methodology (RSM). Also, X-Ray Diffraction (XRD) and Scanning Electron Microscopy (SEM) were employed to confirm the NTO particles on the cotton surface. Cotton fabric treated with optimum concentration of NTO and BTCA obtained enhanced antibacterial properties.

Keywords: antibacterial; butane tetra carboxylic acid; central composite design; cotton; nano TiO_2

INTRODUCTION

The growth of microbes on clothing while in use and in storage has an impact on both the wearer and the clothing. Pathogenic microbes are among the most common impurities in water and air. The outbreak of these impurities motivates scholars to work on health approaches in order to make human lives easier [1]. Various types of antimicrobial agents have been employed to protect textiles against pathogenic microorganisms [2-4].

TiO_2 has been attended more than other kind of nano particles, because of its chemical durability, reasonable cost, availability, non-poisoning, and optical properties [5-10]. When UV rays ($\lambda <$ 388 nm), irradiate to TiO_2, an electron from the valence band exited to the conduction band, creating pairs of negative electrons (e^-) and positive holes (h^+) [11-13]. The construction of efficient species such as OH° has much activity for materials oxidation, organic pollutions and microorganism deactivation [14].

The obtained properties of textiles utilizing nano-technology include water repellency, soil resistance, anti-microbial [15-20], anti-static and UV-protection [21], wrinkle resistance [22], flame retardancy, preventing photo yellowing [23] and self-cleaning on different textiles [24-29] fibres.

The multifunctional properties of poly carboxylic acids such as high accessibility and environment-friendly lead to their application on the textile surfaces [30-31]. The cross-linking [32], antibacterial activity [33], and the finishing of anti-microbial and cross-linking [34] at the same time are among these uses. BTCA showed excellent multifunctional characteristics as enhancing the adsorption and stability of NTO particles on the surface [35-36]. This caused the effective increase of NTO particles efficiency on characteristics such as hydrophilicity, self-cleaning and UV protection [37].

In order to optimize the variables in various processing conditions, the use of central composite design (CCD) attracted the attention of many researchers in their recent scientific studies. These conditions include the optimization of wool dyeing with madder and liposome [38], modification of wool surface by liposome for dyeing with weld [39], photo-catalytic decolorization of reactive Black 5 [40], extraction process of polysaccharides from poria cocos [41], and dried longan pulp [42], electrochemical treatment [43], protease production [44], and more recently self-cleaning of conventional and cationized cotton [45].

The effects of NTO particles on the concurrent cross-linking and anti-microbial finishing of various cotton fabrics are presented in our recent reports [46]. Later, in 2010, the optimization of dry crease recovery angle (DCRA) of bleached cotton with poly carboxylic acids/SHP/NTO using CCD approach demonstrated our work [47].

The photo induced silver on NTO/CA to improve antimicrobial properties on wool was reported by M. Montazer and his colleagues [48] in 2011. Their results revealed that the increase of CA, improved Ag/TiO_2 adsorption on the wool fabric surface and enhanced antibacterial activity.

The enhanced antibacterial activity of cotton fabric explored in the presence of BTCA/SHP/NTO under different curing conditions (UV, Temp, and UV-Temp) is reported in the present study. Antibacterial activity was measured with calculation of bacteria reduction against Gram-positive (*Staphylococcus aureus)* and Gram-negative (*Escherichia coli*) bacteria. The role of the concentration of cross-linking agent (BTCA) and NTO particles over the results was investigated using response surface methodology (RSM). The best treatment conditions for optimizing the antibacterial characteristics of the cotton samples were demonstrated.

MATERIALS AND METHODS

Material

BTCA, sodium hydroxide, sodium hypophosphite (SHP) and tryptic soy agar culture medium prepared from Merck Chemical Co., Germany. Non-ionic detergent (Rucogen DEN) composed of fatty alcohol ethoxylate purchased from Rudolf Chemie Co., (Tehran, Iran). Nano titanium dioxide was employed as the photo catalyst with the anatase crystalline structure and average particle size of 21 nm from Degussa Chemie Co., Duisburg, Germany. *Escherichia coli* (ATCC 11303) and *Staphylococcus aureus* (ATCC 1112) were adapted as pathogenic bacteria. The desized, scoured and bleached plain weave 100% cotton fabric was used with wrap density 32 yarn/cm, weft density 30yarn/cm and fabric weight of 118 g/m^2.

Instrument

Finishing compounds were prepared and dispersed using an ultrasonic bath (200V, 50W, 40 KHz). Thermal oven was used to dry and cure the samples. Some treated samples were exposed to the UV-A irradiation of an HPA 400S lamp (400 W, Philips, Belgium). UV-C irradiation (30W, Philips, Holland) was used to increase the efficiency of antimicrobial activity. Scanning electron microscopic (SEM) observations on specimens of treated fabrics were carried out using an LEO 440i electron microscope (UK). An X-ray diffractometer type 3003 PTS, SEIFERT, Germany (λ=1.54060A°, at 40 kV and 30mA) with Cu Kα irradiation was used to identify the crystalline phase and also crystal size, using the Scherrer method.

Bleaching Pre-treatment

The samples of cotton fabric were prepared in 14×5 cm^2 swatches. Bleaching treatment of cotton fiber was performed using 3.5% hydrogen peroxide and 2% sodium hydroxide (based on weight of fabric: o.w.f) at a liquor ratio of 8:1 under the boiling condition for 90 min.

NTO/BTCA Suspension Preparation

The dispersion of aqueous finishing was prepared with mixture of BTCA, SHP (60% of BTCA) and NTO with required distilled water in ultrasound bath for 30 min.

NTO/ BTCA Suspension Treatment

The cotton fabrics were padded with 95% wet pick-up using freshly prepared aqueous solutions and dried at 65°C for 3 min followed by curing with different conditions: 15 min under UVA (UV), 180°C for 2 min (Temp) and UV-Temp. Then, the finished samples were washed at 75°C for 20 min with 1.5 g/L Na_2CO_3 and 1 g/L non-ionic detergent (Rucogen DEN), and finally dried at room temperature.

Antibacterial Performance

Two pathogenic microorganisms including *Escherichia coli* (*E.coli)* as Gram-negative and *Staphylococcus aureus (S.aureus)* as Gram-positive bacteria were tested using the AATCC 100-2004 test method. In this method, treated samples were placed adjacent to the bacteria suspension and exposed to (UV-C) irradiation for 30 min and then incubated for 24 h. The number of viable bacteria colonies on the agar plate before and after NTO/BTCA treatment was counted and the results reported as percentages of bacteria reduction according to Eq. (1).

$$R(\%) = 100(A - B)/A \qquad (1)$$

where (A) and (B) are the numbers of bacteria colonies recovered from the untreated and the treated cotton samples respectively after inoculation, and (R) is the reduction percentage of bacteria colonies [49].

Evaluation of UV-C Irradiation Time

Each bacterium was prepared and exposed to UV-C radiation for 0, 15, 30 and 45 min to evaluate the influence of the UV-C irradiation. Then, the percentage of bacteria reduction was determined.

The Experimental Design

The central composite design with four variables was used as the experimental plan. These variables were: BTCA (50.96-99.04 g/L), NTO (0.12-5.02%) and curing conditions (UV, Temp and UV-Temp). Details of the treatment design with BTCA/NTO are presented in Table I.

RESULTS AND DISCUSSIONS

Effect of UV-C Irradiation Time

Antibacterial properties of UV-C irradiation were accomplished based on a quantitative standard method. It is clear that the UV irradiation was able to kill bacteria as shown in *Figure 1*. However, the rate of reduction was low even after 45 min UV-C irradiation, and the highest decrease was 10.61% for *S. aureus* and 12.54% for *E. coli*.

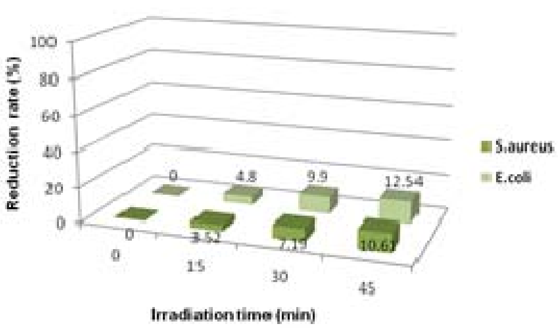


FIGURE 1. Effect of UV-C irradiation times on the reduction rate.

Antibacterial Properties

The designed experiments in *Table I* show the reduction percentages of *E.coli* and *S.aureus* bacteria of the treated cotton fabrics with various BTCA, NTO concentrations and different curing methods UV, Temp, and UV-Temp. Increasing BTCA concentration decreased the bacteria growth considerably on both types of the bacteria. This can be due to the presence of more carboxylic groups of BTCA that leads to the more absorption of NTO particles on the fabric surface via electrostatic attraction. Also increasing NTO concentration caused increasing antibacterial property, however, with higher NTO, a small decrease was observed in antibacterial activity. This can be referred to the NTO particles aggregation in the finishing bath or on the cotton fabric surface which consequently decreased antibacterial efficiency of the nano particles. Also the samples cured with UV-Temp demonstrated the highest antibacterial efficiency as compared with other curing methods. The consequence of the influence of curing method on the antibacterial efficiency was as follows: UV<Temp<UV-Temp. It has indicated that the UV-Temp method was more effective than the UV or Temp method in our previous findings [50]. This was explained by the different cross-linking mechanisms of cotton fabric based on UV and Temp while both of them occurred in the UV–Temp curing method.

The highest bacteria reduction was 99.79% for *E.coli* (*Figure 2a*) and 96.76% for *S.aureus* (*Figure 2b*) of sample 12 (UV-Temp, NTO 4.30%, 92.00 g/L). Thus *S.aureus* exhibited a little more resistance than *E.coli*. The main difference of these two bacteria relates to their cellular wall and peptidoglycan thickness as it is more for Gram-positive bacteria which showed a relative higher resistance against antibacterial agents. Killing bacteria by antibacterial agents can be obtained by the demolition of bacteria membrane, the spatial deformation, deformation of bacteria enzyme, and the chromosome damage [51]. BTCA accompanying with NTO particles had a complementary effect and showed a noticeable reduction in *E.coli* Gram-negative (urine, infirmary, and blood infection source) and *S.aueus* Gram-positive (eye, skin, bone and joins infection source) bacteria.

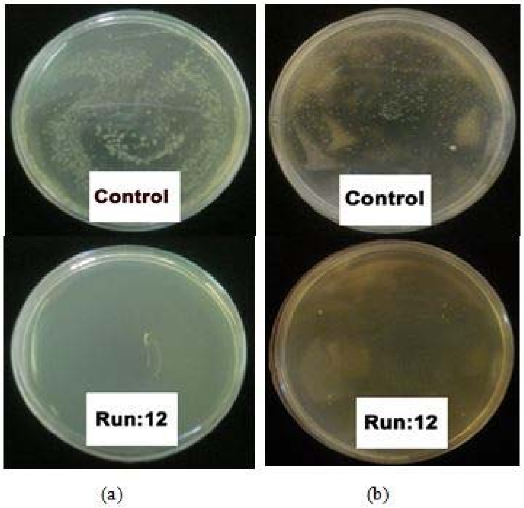


FIGURE 2. Antibacterial activity of the treated cotton fabrics against *E. coli* (left) (a) blank cotton sample and sample Run: 12, and *S. aureus* (right) (b) blank cotton sample and sample Run: 12, based on quantitative method: (Run: 12, treated with 92.00 g/L BTCA, 55.20 g/L SHP, and 4.3% NTO and UV–Temp curing method).

Statistical Analysis

This study was conducted according to response surface methodology (RSM) and central composite design (CCD). In total, 78 experimental CCD designed runs were conducted according to Table I. In this model, the impacts of independent variables including NTO, BTCA and curing methods on response surfaces were assessed. For each of them optimum condition obtained (*Table I*), through which the variation of response surface was discussed. These explained antibacterial activity of treated samples against *S. aureus* and *E. coli* bacteria. Based on the results of relations between surface responses and independent factors, several mathematical models were obtained, whose related responses were a function of its independent variables.

Mathematical models related to the antibacterial activity of the treated fabrics against *E. coli* and *S. aureus* are presented in Eq. (2-4) and Eq. (5-7) respectively:

$$Y_{1\,(E.coli,\ UV)} = +30.47 + 0.087\times BTCA + 20.79\times NTO - 0.095\times BTCA \times NTO + 2.78E\text{-}003\times BTCA^2 - 2.10\times NTO^2 \quad (2)$$

$$Y_{1\,(E.coli,\ Temp)} = +34.03 + 0.15\times BTCA + 21.28\times NTO - 0.095\times BTCA \times NTO + 2.78E\text{-}003\times BTCA^2 - 2.10\times NTO^2 \quad (3)$$

$$Y_{1\,(E.coli,\ UV\text{-}Temp)} = +39.41 + 0.19\times BTCA + 21.60\times NTO - 0.095\times BTCA \times NTO + 2.78E\text{-}003\times BTCA^2 - 2.10\times NTO^2 \quad (4)$$

$$Y_{2\,(S.aureus,\ UV)} = +27.86 + 0.07\times BTCA + 20.73\times NTO - 0.09\times BT\,CA \times NTO + 2.84E\text{-}003\times BTCA^2 - 2.10\times NTO^2 \quad (5)$$

$$Y_{2\,(S.aureus,\ Temp)} = +31.54 + 0.14\times BTCA + 21.22\times NTO - 0.09\times BT\,CA \times NTO + 2.85\,E\text{-}003\times BTCA^2 - 2.10\times NTO^2 \quad (6)$$

$$Y_{2\,(S.aureus,\ UV\text{-}Temp)} = +36.74 + 0.18\times BTCA + 21.57\times NTO - 0.09\times BT\,CA \times NTO + 2.85\,E\text{-}003\times BTCA^2 - 2.10\times NTO^2 \quad (7)$$

The response surfaces of cotton samples against *E. coli* and *S. aureus* were constructed and are shown in *Figure 3. Figure 3* (a-c) and (d-f) show the response surfaces of the model for treated fabrics against *E. coli* and *S. aureus*, respectively.

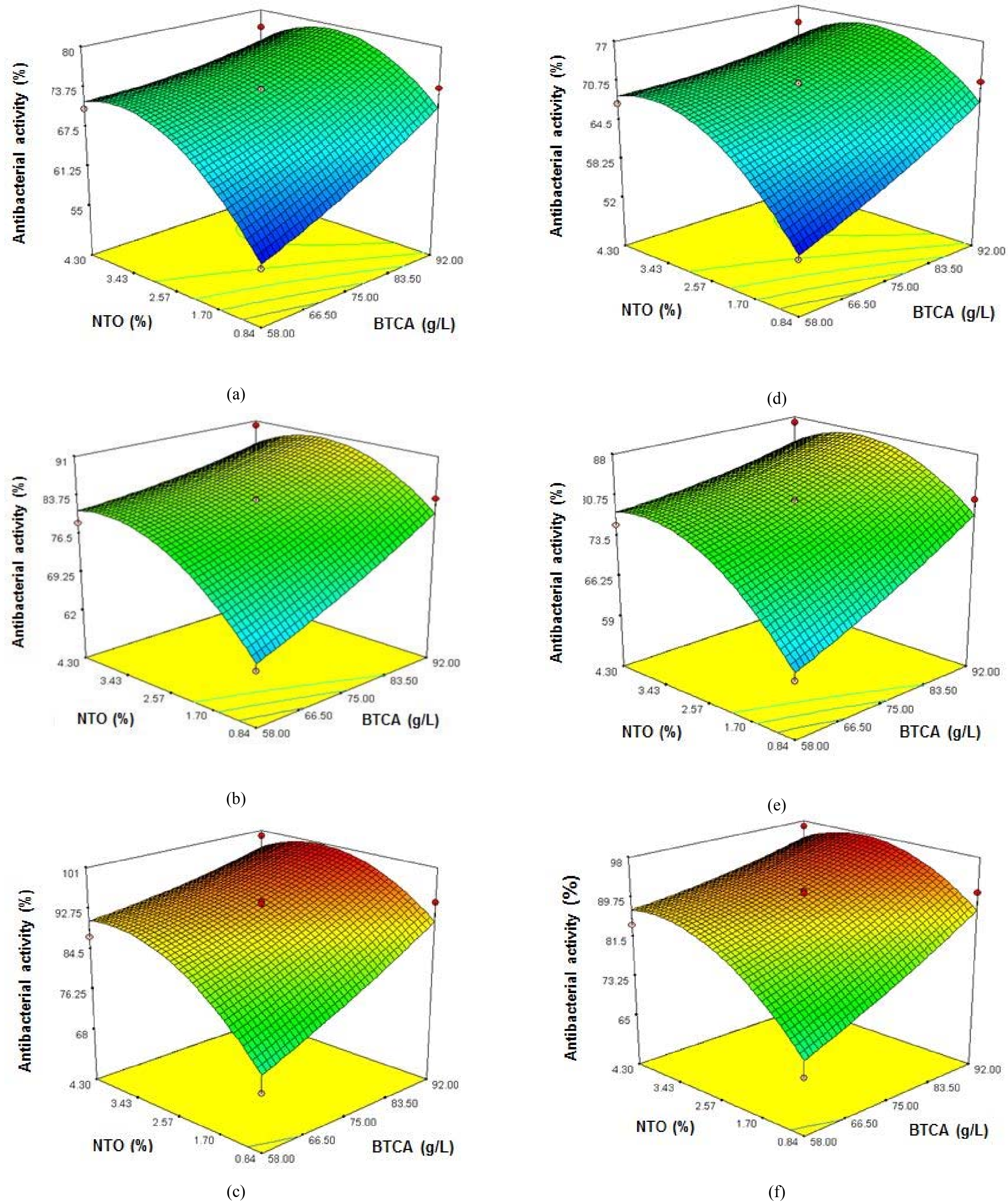


FIGURE 3. Response surfaces for antibacterial activity as a function of NTO and BTCA for treated fabrics against *E. coli*: (a) UV, (b) Temp, (c) UV-Temp and against *S. aureus*: (d) UV, (e) Temp, and (f) UV-Temp.

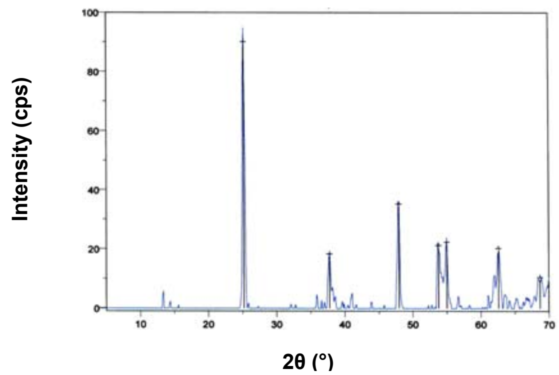
By using the Design of Expert software, the optimum condition of *E. coli* reduction (Y_1 = 99.92 %) with desirability of 1.00 was found to be for the cotton fabric containing 90.31 (g/L) BTCA and 3.00 (%) NTO cured with UV–Temp method. The optimum point of *S. aureus* reduction (Y_2=97.54 %) with desirability of 1.00 was found to be for the cotton fabric including 91.98 (g/L) BTCA, 3.00% NTO, cured with UV–Temp.

Analysis of variance (ANOVA) was used to analyze the data to obtain the interaction between the process independent variables and the responses. The results were then analyzed by ANOVA to assess the ''goodness of fit'' (*Tables II and III*).

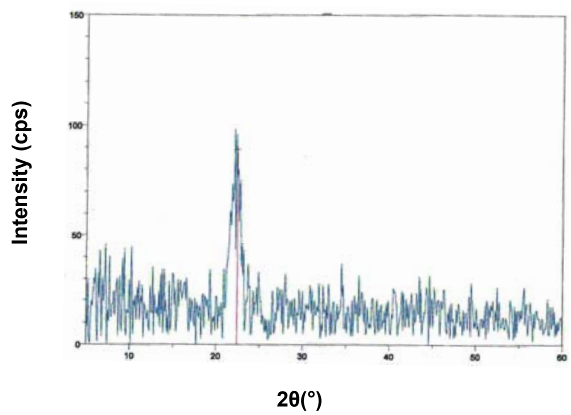
It was observed that the designed model for antibacterial activity of treated samples against *E. c*oli was statistically significant at an F value of 59.42 and values of prob > F (<0.0001) (*Table II*). Similarly, antibacterial activity against *S. aureus* was significant at an F value of 59.59 and values of prob > F (<0.0001) (*Table III*). These models indicate an effectiveness of antibacterial activity against both *S. aureus* and *E. coli* bacteria significant at a high confidence level (99.99%). The values of prob>F which was less than 0.0500, indicates the significance of models terms. Therefore, the independent variables of NTO, BTCA, and curing methods (UV, Temp, and UV-Temp) with prob>F (<0.0001) were statistically significant in the models.

X-Ray Diffraction (XRD) Analysis

The XRD patterns of pure NTO (*Figure 4a*), sample Run: 12 (*Figure 4b*) are presented in *Figure 4*. The major peak of both spectrums were anatase (2θ=25.2°), but the peak related to rutile phase (2θ=27.5°) was not observed in XRD spectrums [52]. Thus, the treated fabrics with NTO particles had anatase crystallite phase and efficiently acted under UV irradiation. From full width at half maximum (FWHM) of the peak at 25.2° (0.3971) and using Scherrer's equation, the average crystal sizes of about 38.55 nm was estimated.



(a)



(b)

FIGURE 4. XRD pattern of pure NTO (a) and sample Run: 12 (b) treated with 92.00 g/L BTCA, 55.20 g/L SHP, 4.3% NTO, and UV–Temp curing method

TABLE I. Central composite design for antibacterial activity against *E. coli* and *S. aureus* of cotton samples treated with BTCA, SHP and NTO cured with different methods.

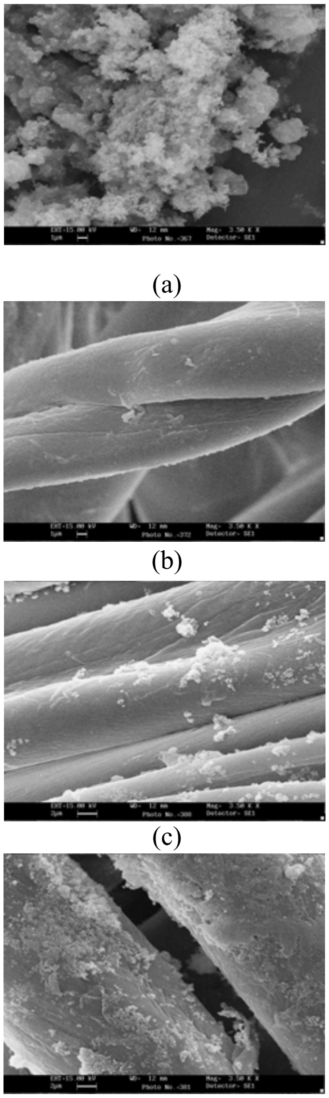
Run Numbers	BTCA (g/L)	SHP (g/L)	NTO (%)	Reduction of *E. coli* (UV)	Reduction of *E. coli* (Temp)	Reduction of *E. coli* (UV-Temp)	Reduction of *S. aureus* (UV)	Reduction of *S. aureus* (Temp)	Reduction of *S. aureus* (UV-Temp)
Control	0	0	0	0	0	0	0	0	0
Blank 1	0	0	2.57	42.65	44.17	46.86	36.69	40.81	43.19
Blank 2	75	45	0	50.12	54.38	58.19	44.18	48.90	54.26
1	58.00	34.80	4.30	70.42	87.72	87.23	67.24	75.61	84.19
2	75.00	45.00	2.57	73.57	82.98	94.02	70.42	79.94	91.05
3	99.04	59.42	2.57	76.81	89.87	99.57	73.68	86.79	96.54
4	75.00	45.00	2.57	73.23	82.89	93.91	70.39	79.73	90.99
5	75.00	45.00	5.02	70.34	78.64	87.15	67.23	75.48	84.03
6	75.00	45.00	0.12	55.49	62.38	68.59	52.39	59.24	65.47
7	92.00	55.20	0.84	73.62	83.19	94.04	70.61	80.02	90.85
8	75.00	45.00	2.57	73.40	83.15	94.08	70.47	79.97	90.74
9	50.96	30.58	2.57	69.36	81.28	90.89	66.35	78.22	87.73
10	58.00	34.80	0.84	55.53	62.55	68.72	52.40	59.50	65.63
11	75.00	45.00	2.57	73.32	82.81	93.83	70.13	79.64	90.87
12	92.00	55.20	4.30	77.02	90.01	99.79	74.01	86.89	96.76
13	75.00	45.00	2.57	73.49	83.06	94.17	70.17	79.97	90.69

TABLE II. ANOVA results of antibacterial activity against *E. coli* bacteria for the treated fabrics.

Source	Sum of squares	Df	Mean Square	F Value	p-value Prob>F
Model	4877.41	11	443.40	59.42	<0.0001 signifcant
A [BTCA]	698.32	1	698.32	93.58	<0.0001
B [NTO]	768.22	1	768.22	102.95	<0.0001
C [Curing]	2411.38	2	1205.69	161.57	<0.0001
AB	94.08	1	94.08	12.61	0.0014
AC	14.09	2	7.05	0.94	0.4015
BC	7.96	2	3.98	0.53	0.5926
A^2	13.45	1	13.45	1.80	0.1906
B^2	827.36	1	827.36	110.87	<0.0001
Residual	201.48	27	7.46		
Lack of fit	201.26	15	13.42	741.16	<0.0001 signifcant
Pure error	0.22	12	0.018		
Cor Total	5078.89	38			

TABLE III. ANOVA results of antibacterial activity against *S.areus* bacteria for the treated fabrics.

Source	Sum of squares	Df	Mean Square	F Value	p-value Prob>F
Model	4873.58	11	443.05	59.59	<0.0001 signifcant
A [BTCA]	698.92	1	698.92	94.00	<0.0001
B [NTO]	769.44	1	769.44	103.48	<0.0001
C [Curing]	2404.82	2	1202.41	161.72	<0.0001
AB	92.57	1	92.57	12.45	0.0015
AC	13.99	2	6.99	0.94	0.4028
BC	8.33	2	4.17	0.56	0.5775
A^2	14.15	1	14.15	1.90	0.1790
B^2	828.05	1	828.05	111.37	<0.0001
Residual	200.75	27	7.44		
Lack of fit	200.47	15	13.36	558.01	<0.0001 signifcant
Pure error	0.29	12	0.024		
Cor Total	5074.34	38			



(a)

(b)

(c)

(d)

FIGURE 5. SEM images of (a) NTO, (b) control cotton sample, (c) treated cotton with 50.96 g/L BTCA, 30.58 g/L SHP, and 2.57% NTO (Run: 9), (d) treated cotton with 92.00 g/L BTCA, 55.20 g/L SHP, and 4.3% NTO (magnification=3500×) and UV–Temp curing (Run: 12)

SEM Images

Figure 5 (a-d) are the SEM images of NTO powder (a), control cotton sample (b), sample Run: 9 (c), and sample Run: 12 (d) (magnification=3500×). Fig. 5b shows the morphology of untreated cotton fabric. The surface alterations of cotton fibres treated with different concentrations of BTCA/SHP/NTO presented after padding and UV-Temp curing are shown in *Figure 5* (c-d). These pictures show the higher amount of NTO loading on the surface of sample Run: 12 as compared to sample Run: 9. This can be attributed to the higher concentrations of NTO particles and carboxylic acid groups on the surface of the treated sample, Run: 12. These images show the NTO particles embedded on cotton fabric by the conventional padding process, and their distribution on the cotton surface is not quite uniform; possibly due to the aggregation of some NTO particles on the surfaces.

CONCLUSION

This study introduced an effective nano composition in order to produce an ideal antibacterial cotton fabric. The NTO particles were successfully applied on the cotton surface and established through BTCA as cross-linking agent. The BTCA cross-linking agent also assisted to enhance the NTO particles absorption and stabilized them on the cotton fabric surface. XRD patterns and SEM images confirmed the presence of NTO particles on the surface of the treated cotton fabrics. RSM analysis illustrated that optimized conditions reduced more than 97% and 99% of Gram positive (*S. aureus*) and Gram negative (*E. coli*) bacteria, respectively. However, BTCA along with NTO under UV-Temp curing condition had complementary effects with enhanced antibacterial activities.

ACKNOWLEDGMENT

The authors wish to thank Yazd Branch, Islamic Azad University, for its support of the research project under the title of optimizing of antimicrobial property of cotton fabric with nano TiO_2: the statistical approach of central composite design.

REFERENCES

[1] Gao Y., Cranston R., "Recent advances in antimicrobial treatments of textiles", *Text. Res. J.,* 78 (1), 2008, 60–72.

[2] Tayel A.A., et al., "Antimicrobial textile treated with chitosan from Aspergillus niger mycelial waste", *Int. J. Biol. Macromol.,* 49, 2011, 241–245.

[3] Rajendran R., et al., "Synthesis and characterization of neem chitosan nanocomposites for development of antimicrobial cotton textiles", *Journal of Engineered Fibers and Fabrics,* 7, 1, 2012, 136-141.

[4] Fras L., Ristić T., Tkavc T.,"Adsorption and antibacterial activity of soluble and precipitated chitosan on cellulose viscose fibers", *Journal of Engineered Fibers and Fabrics,* 7, 1, 2012, 50-57.

[5] Mellott N.P., et al., "Commercial and laboratory prepared titanium dioxide thin films for self-cleaning glasses: photocatalytic performance and chemical durability", *Thin Solid Films,* 502, 2006, 112–120.

[6] Sunada K., Watanabe T., Hashimoto K., “Studies on photokilling of bacteria on TiO_2 thin film”, *J. Photochem. Photobiol, A: Chem.,* 156, 2003, 227-233.

[7] Nawi M.A., et al., “Photocatalytic-oxidation of solid state chitosan by immobilized bilayer assembly of TiO_2–chitosan under a compact household fluorescent lamp irradiation”, *Carbohydr. Polym.,* 83, 2011, 1146–1152.

[8] Klaus P.K., et al., “Disinfection of surfaces by photocatalytic oxidation with titanium dioxide and UVA light”, *Chemosphere*, 53, 2003, 71-77.

[9] Montazer M., Morshedi S., “Nano photo scouring and nano photo bleaching of raw cellulosic fabric using nano TiO_2”, *Int. J. Biol. Macromol.,* 50, 2012, 1018– 1025.

[10] Yao K.S., et al., “Photocatalytic bactericidal effect of TiO_2 thin film on plant pathogens”, *Surf. Coat. Tech.,* 201, 2007, 6886-6888.

[11] Onar N., et al., “Low-temperature, sol–gelsynthesized, silver doped titanium dioxide coating to improve ultraviolet blocking properties for cotton fabrics”, *J. Appl. Polym. Sci.*, 106, 2006, 514–525.

[12] Machida M., Norimoto K., Kimura T., “Antibacterial activity of photocatalytic titanium dioxide thin films with photodeposited silver on the surface of sanitary ware”, *J. Am. Ceram. Soc.,* 88, 2004, 95–100.

[13] Valentine Rupa A., et al., “Effect of deposition of Ag on TiO_2 nanoparticles on the photodegradation of reactive yellow-17”, *J. Hazard. Mater,* 147, 2007, 906–913.

[14] Rincon A.G., Pulgarin C., “Photocatalytical inactivation of E.coli: effect of (continuous-intermittent) light intensity and of (suspended-fixed) TiO_2 concentration”, *Appl. Catal. B.,* 44, 2003, 263-284.

[15] Selvam S., Sundrarajan M., Functionalization of cotton fabric with PVP/ZnO nanoparticles for improved reactive dyeability and antibacterial activity, *Carbohydr. Polym.,* 87, 2012, 1419– 1424.

[16] Sivakumar P.M., et al., “Effective antibacterial adhesive coating on cotton fabric using ZnO nanorods and chalcone”, *Carbohydr. Polym.,* 79, 2010, 717–723.

[17] El-Rafie M.H., et al., “Antimicrobial effect of silver nanoparticles produced by fungal process on cotton fabrics”, *Carbohydr. Polym.,* 80, 2010, 779–782.

[18] Hebeish A., et al., “Highly effective antibacterial textiles containing green synthesized silver nanoparticles”, *Carbohydr. Polym.,* 86, 2011, 936– 940.

[19] Son W.K., Youk J.H., Park W.H., “Antimicrobial cellulose acetate nanofibers containing silver nanoparticles”, *Carbohydr. Polym.,* 65, 2006, 430-434.

[20] Montazer M., et al., “In situ synthesis of nano silver on cotton using Tollens’ reagent”, *Carbohydr. Polym.,* 87, 2012, 1706– 1712.

[21] Shafei A.El., Abou-Okeil A., “ZnO/ carboxymethyl chitosan bionano-composite to impart antibacterial and UV protection for cotton fabric”, *Carbohydr. Polym.,* 83, 2011, 920–925.

[22] Yuen C.W.M., et al., “Improvement of wrinkle-resistant treatment by nanotechnology”, *J. Text. Inst.,* 100, 2009, 173–180.

[23] Montazer M., Pakdel E., “Reducing photoyellowing of wool using nano TiO_2”, *Photochem. Photobiol.,* 86, 2010, 255–260.

[24] Meilert K.T., Laub D., Kiwi J., “Photocatalytic self-cleaning of modified cotton textiles by TiO_2 clusters attached by chemical spacers”, *J. Mol. Catal. A: Chem.* 237, 2005, 101–108.

[25] Veronovski N., et al., Self-cleaning and handle properties of TiO_2-modified textiles”, *Fibers Polym.,* 10 (4), 2009, 551-556.

[26] Yuranova T., et al., “Self-cleaning cotton textiles surfaces modified by photoactive SiO_2/TiO_2 coating”, *J. Mol. Catal. A: Chem.,* 244, 2006, 160–167.

[27] Vero N., et al., “Homogeneous self-cleaning coatings on cellulose materials derived from TIP/TiO_2 P25”, *Fibers Polym.,* 10(5), 2009, 716-723.

[28] Tung W.S., Daoud W.A., “Photocatalytic formulations for protein fibers: experimental analysis of the effect of preparation on compatibility and photocatalytic activities”, *J. Colloid Interface Sci.*, 326, 2008, 283–288.

[29] Yuranova T., Laub D., Kiwi J., “Synthesis, activity and characterization of textiles showing self-cleaning activity under daylight irradiation”, *Catal. Today,* 122, 2007, 109–117.

[30] Kim Y.H., et al., “Durable antimicrobial treatment of cotton fabrics using N-(2-hydroxy) propyl-3-trimethylammonium chitosan chloride and polycarboxylic acids”, *J. Appl. Polym. Sci.*, 88, 2003, 1567–1572.

[31] Lam Y.L., Kan C.W., Yuen C.W.M., "Effect of concentration of titanium dioxide acting as catalyst or co-catalyst on the wrinkle-resistant finishing of cotton fabric", *Fibers Polym.*, 11(4), 2010, 551-558.

[32] Orhan M., Kut D., Gunesoglu C., "Improving the antibacterial activity of cotton fabrics finished with triclosan by the use of 1,2,3,4-butanetetracarboxylic acid and citric acid", *J. Appl. Polym. Sci.,* 111, 2009, 1344–1352.

[33] M. Montazer, M. Gorbanali Afjeh, Simultaneous x-linking and antimicrobial finishing of cotton fabric, J. Appl. Polym. Sci. 103 (2007) 178–185.

[34] M. Montazer, S. Seifollahzadeh, Pretreatment of wool/polyester blended fabrics to enhance titanium dioxide nanoparticle adsorption and self-cleaning properties, Color. Technol. 127 (2011) 322–327.

[35] M. Montazer, S. Seifollahzadeh, Enhanced self-cleaning, antibacterial and UV protection properties of nano TiO_2 treated textile through enzymatic pretreatment, Photochem. Photobiol. 87 (2011) 877–883.

[36] Montazer M., Pakdel E., Bameni Moghadam M., "The role of nano colloid of TiO2 and butane tetra carboxylic acid on the alkali solubility and hydrophilicity of proteinous fibers", *Colloids Surf. A.,* 375, 2011, 1–11.

[37] Montazer M., et al., "Optimization of dyeing of wool with madder and liposomes by central composite design". *J. Appl. Polym. Sci.*, 106, 2007, 1614–1621.

[38] Montazer M., et al., "Modification of wool surface by liposomes for dyeing with weld", *J. Liposome Res.,* 19(3), 2009, 173–179.

[39] Secula M.S., et al., "Response surface optimization of the photocatalytic decolorization of a simulated dyestuff effluent", *Chem. Eng. J.,* 141, 2008, 18–26.

[40] Yongjiang W., et al., "Optimization of ultrasonic-assisted extraction process of poria cocos polysaccharides by response surface methodology", *Carbohydr. Polym.,* 77, 2009, 713–717.

[41] Zhong K., Wang Q., "Optimization of ultrasonic extraction of polysaccharides from dried longan pulp using response surface methodology", *Carbohydr. Polym.,* 80, 2010, 19–25.

[42] Körbahti B.K., "Response surface optimization of electrochemical treatment of textile dye wastewater", *J. Hazard. Mater,* 145, 2007, 277–286.

[43] Dutta J.R., Dutta P.K., Banerjee R., "Optimization of culture parameters for extracellular protease production from a newly isolated Pseudomonas sp. using response surface and artificial neural network models", *Process Biochem.*, 39, 2004, 2193–2198.

[44] Nazari A., et al., "Self-cleaning properties of bleached and cationized cotton using nanoTiO_2: A statistical approach", *Carbohydr. Polym.*, 83, 2011, 1119–1127.

[45] Nazari A., Montazer M., Rahimi M.K., "Concurrent antimicrobial and cross-linking of bleached and cationic cotton using nano TiO_2 and BTCA", *Iran. J. Polym. Sci. Technol.,* 22 (1), 2009, 41–51.

[46] Nazari A., et al., "Optimization of cotton crosslinking with polycarboxylic acids and nano TiO_2 using central composite design", *J. Appl. Polym. Sci.,* 117, 2010, 2740–2748.

[47] Montazer M., et al., "Photo induced silver on nano titanium dioxide as an enhanced antimicrobial agent for wool", *J. Photochem. Photobiol. B: Biol.,* 103, 2011, 207-214.

[48] Bingshe X., et al., "The structural analysis of biomacromolecule wool fibre with Ag-loading SiO_2 nano-antibacterial agent by UV radiation", *J. Photochem. Photobiol. A: Chem.*, 188, 2007, 98–105.

[49] Nazari A., et al., "Nano TiO_2 photo-catalyst and sodium hypophosphite for cross-linking cotton with poly carboxylic acids under UV and high temperature", *Appl. Catal. A: Gen.*, 371, 2009, 10–16.

[50] Melnick J.L., Adelberg E.A., Medical Microbiology, 24e, McGraw-Hill, Australia, 2007.

[51] Jung K.Y., Park S.B., "Anatase-phase titania: preparation by embedding silica and photocatalytic activity for the decomposition of trichloroethylene, Photochem". *Photobiol. A: Chem.,* 127, 1999, 117-122.

AUTHORS' ADDRESSES
Ali Nazari, PhD
Department of Art & Architectural
Islamic Azad University
Yazd Branch
Hesabi BLV, Safaieh, YAZD, I.R.IRAN
Yazd, 89195-155
IRAN

Majid Montazer, PhD
Department of Textile Engineering
Amirkabir University of Technology
Hafez Avenue
Tehran,
IRAN

Mohammad Bameni Moghadam, PhD
Department of Statistics
Allameh Tabatabaí University
Tehran
IRAN

Mohammad Esmail Yazdanshenas, PhD
Department of Textile
Yazd Branch
Islamic Azad University
Yazd
IRAN