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The Effects of Various Dyes on the Antimicrobial Activities of Microbial Cellulose Nonwoven Fabrics Treated by Rodalon.

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ABSTARCT

The present study was undertaken to determine the effects of the presence of the various dyes such as reactive, direct and vat on the antimicrobial activity of Microbial cellulose nonwoven fabric treated by Rodalon (Z) as a quaternary ammonium compound. In addition, the effects of plasma on the antimicrobial properties of dyed fabrics were investigated. The effect of simultaneous dyeing and finishing on some physical properties such as mechanicals properties and color strength was assessed. The results of the experiments demonstrated a different antibacterial activity of the dyed fabrics in the presence of two types of bacteria (E.coli AATCC 8099 and S.aureus AATCC 6538). Furthermore, Rodalon and finishing conditions didn't have an impression on some physical properties of the dyed fabrics.

Keyword: Microbial Cellulose, Nano Fiber, Antimicrobial, Plasma, Rodalon (Z).

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INTRODUCTION

Many synthetic dyes are used in the textile industry. Some synthetic dyes have been specifically made to perform an antimicrobial activity. But another approach to achieve simultaneous dyeing and antimicrobial finishing is to covalently attach a biocide to a dye via a linker. Most of the synthetic dyes do not have an antimicrobial activity nor attach a linker part such as many vat, direct, reactive and disperse dyes. Recently, many researchers have explored dying and functional finishing of textiles. Many of the reported studies concern dying and durable press finishing. Thus, a compromise of dyeing and finishing conditions is usually required and the properties of the resultant fabrics are usually sacrificed. Humans need for personal health and hygienic clothing has led to creation of a great number of antimicrobial textile products [1]. The biodeterioration of textile fibers caused by microorganisms including bacteria, fungi, and algae present in the air, water and soil can cause multiple problems for textile products. Biodeterioration can lead to an unpleasant smell and colored stains. Changes in the textile colors and in the textile mechanical properties such as the breaking strength, elongation and elasticity negatively affect the applicability value of textiles from a hygienic and aesthetic point of view [2]. In the last few decades, prevention of microbial attacks on textile and wearers of textile materials has become increasingly important to consumers and textile producers. For instance, a market study in Germany revealed a steady increase in the demand for antimicrobial fabrics [3]. Therefore, with a rising interest in personal health and hygiene, textiles with antimicrobial properties are becoming an increasingly desirable aim of textile manufactures [4].

Microbial cellulose as a nano fibers and biological nonwoven fabric possesses high crystallinity, high tensile strength, and extreme insolubility in most of the solvents, mold ability and high degree of polymerization. The thickness of cellulose fibrils is generally 0.1-10 mm, one hundred times thinner than that of cellulose fibrils obtained from plants with good shape retention. Its water holding capacity is over 100 times (by mass) higher. Microbial cellulose is far stronger than plant cellulose. Macroscopic morphology of cellulose strictly depends on the culture conditions, which can easily be tailored for the physicochemical properties Microbial cellulose as a bio-nonwoven fabric has been used in production of high-quality paper, special separation membranes, nonwoven cloth, wound, medical pads, artificial skin, films, material coating, speaker diagrams, and many high performances and other things. Microbial cellulose products can be dyed with several dyes same as plan cellulose such as cotton [5-7].

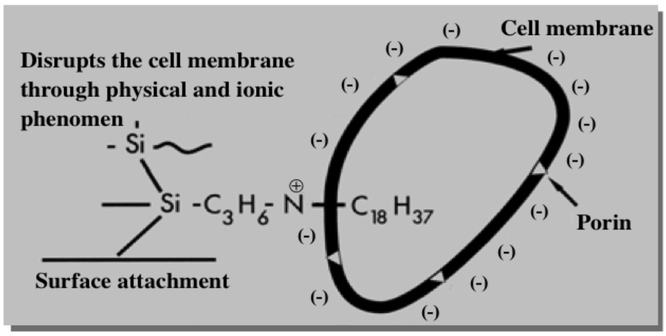


Figure 1: Chemical bonded mechanism

Several different types of antimicrobial agents such as oxidizing agents, coagulants, diphenyl ether (bis – phenyl) derivatives, heavy Metals and metallic compounds, chitosan and quaternary ammonium compounds (QACs) are used in the textile industry to confer antimicrobial properties. Among them, QACs, chains of 12-18 carbon atoms, have been widely used as cationic disinfectants or biocide coating to prevent the growth of microorganisms on the surface of materials including fibers, paints filters and packing films. These compounds carry a positive charge at the N atom in solution and inflict a variety of detrimental effects on microbes they also damage cell membranes, denaturant proteins and disrupt the cell structure [4]. A significantly different and much more unique antimicrobial technology used in the nonwovens and building construction industries does not leach but instead remains permanently affixed to the surface on which it is applied. Applied in a single stage of the wet finish process, the attachment of this technology to the surfaces involves two means. First and the most important is a very rapid process which coats the substrate (fabric, fiber, etc.) with the cationic species (physisorption) one molecule deep. This is an ion exchange process by which the action of the silane QACs replaces protons from water or chemicals on the surface. The second mechanism is unique to certain materials such as silane QACs. In this case, the silanol allows covalent bonding to receptive surfaces to occur (chemisorption). This bonding to the substrate is then rendered even more durable by the silanol functionality, which enables them to homopolymerize. After they have coated the surface in this manner, they become virtually irremovable, even on the surfaces with which they cannot react covalently (Figure 1) [8].

In this research, Rodalon which is a poly siloxan based cationic QAC is used for retentive antimicrobial and anti-odor finish on Microbial cellulose substrates. Microbial cellulose nonwoven fabrics are used in this study because they are frequently used fabric for many medical and common new textiles [9]. For dyeing Microbial cellulose fabrics, Vat, reactive, or direct dyes are used. Existence of these dyes in fabric structure can be effective on other finishing that has been done on the fabric. Therefore, the target of this study is examination of antibacterial properties of Rodalon in the presence of different kinds of dyes in diverse situations such as plasma treated or untreated, washing procedure and also investigation of every impression of this substance that causes a change in the physical properties such as mechanical properties [10].

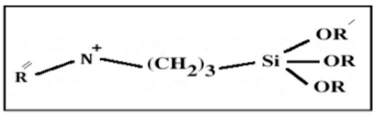
MATERIAL

Microbial cellulose Preparation

Accetobacter Xylinum ATCC 23768 was used for Microbial cellulose production. The microbe was provided from the medical sciences faculty tarbialmodares university, Tehran, Iran. The bacterium was grown in SH medium at 28ºC under static culture conditions. SH medium was composed of 2% (W/V) glucose, 0.5% (W/V) yeast extract, 0.5% peptone, 0.27% (W/V) Na2HPO4 and 0.115% (W/V) citric acid. Preinoculum for all experiments was prepared by transferring a single colony grown on SH agar medium into a 50 mL Erlenmeyer flask filled with liquid SH medium. After 7 days of cultivation at 28°C, the cellulose pellicle formed on the surface of the culture broth. Ten milliliters of the cell suspension was introduced into a 500 mL Erlenmeyer flask containing 100 mL of a fresh SH medium. The culture was carried out statically for 72 h and the cell suspension derived from the synthesized cellulose pellicle was used as the inoculums for further cultures [5-7].The cellulose sheets were removed after cultivation and rinsed with distilled water. They were cut into 0.05 gr and used for dye biosorbent.

Antimicrobial agent

The antimicrobial agent, Rodalon was obtained from Exiresharghe Mashhad ltd, the representative of Zydex Co. in Iran. (It is based on 2-Hydroxyethoxydimethoxysilyl, shown in figure 2. It shows in paper Z for Nomenclature.



(2-Hydroxyethoxydimethoxysilyl)
Octadecyldimethyl Ammonium Cholorid

Figure 2: The chemical structure of Rodalon (Z)

Dyes

The characteristics and chemical structures of the dyes are shown in Table 1 and figure 3(a-c):

Table 1: Name, color index and the manufacture of the dyes

Commercial name	C.I	Company
BASF 58	24400 Direct Blue 15	T.H.C
NovaticbluBc	69825 Vat Blue 6	Atic industrial
Remazole Brilliant blue special	61200 Reactive 19	HOE AG

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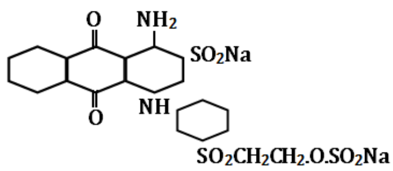
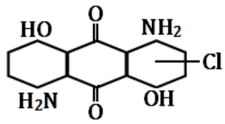
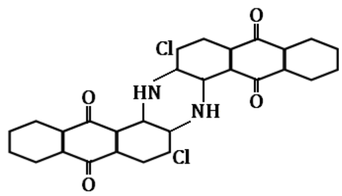


(a). 61200 C.I. Reactive Blue 19 (*Bright blue)*



(b). 63285   C.I. Disperse Blue 56 (*Blue*)



(c). 69825 C.I Vat blue (Bright blue)

Figure 3 (a-c): Chemical structures of dyes by C.I number

Microorganism

The bacterium was provided by the microbial bank of Tarbiat modarres university of Tehran: gram – negative (Escherichia coli AATCC8099) and gram – positive (staphylococcus aureus AATCC6538).

Nomenclature

The explanations of signs which are used in the present study are to be found in the following description:

Z, Rodalon treated; Z+10w, Rodalon treated then 10 times washed; P+Z, plasma then Rodalon treated; P+Z+10w, first plasma then Rodalon treated and at after 10 times washed; D, R & V, dyed fabrics with direct, reactive and vat dyes.

METHODS

Dyeing

Microbial cellulose fabrics dyed with direct, reactive and vat dyes.

Plasma treatment

The fabric samples with dimensions of 5cm*5cm were produced using an injection molding machine. The samples were laid on stainless steel substrates and inserted into the plasma immersion ion implanter. The O_2 plasma treatment was performed in the optimal conditions based on many experiments: bias voltage = -12 Kv, voltage pulse width = 20 µs, pulsing frequency = 30 Hz, gas flow= 35 seem, RF power = 1000w and treatment time = 30 min [11].

Antibacterial treatment

In order to finish antibacterial property as per gram weight of goods, 1.5g water and as 3% ratio of weight of goods, Rodalon were added to the bath, then specimen was entered into the bath and immersed for 10 minutes and finally it was dried for 5 minutes in the oven with a temperature of 90^{oC}.

Washing procedure

The treated samples were washed repetitively 10 times according to AATCC test method 61-1994. The treated samples were then evaluated for antimicrobial activity before and after washing [12, 13].

Antibacterial activity evaluation

The antimicrobial activity of the treated and untreated samples was quantitatively evaluated against both gram–positive (S.aureus–AATCC6538) and gram–negative (E.coli–AATCC 8099) bacteria according to the AATCC test method 100-2004. The percentage of the reduction of bacteria was calculated by following formulas:

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

Where, R = % reduction, A = number of bacteria recovered from the treated specimen after 24 hr, B = number of bacteria recovered from the untreated specimen after 24 hr [14-16]

Color strength

The color strength (K/S) of dyes fabric was assessed using Kubeka-Munk equation:

$$K/S = (1 – R)^2 / 2R$$

Where K, and R are the absorption coefficient, scattering coefficient and reflectance, respectively [1].

Physical tests

Some mechanical properties such as tenacity, elongation at rupture, force at rupture and initial modulus of the untreated and treated fabrics were measured according to ASTM D5030 by Testometric M500-25 CT [18,19].

RESULTS AND DISCUSSION

The effect of different kinds of dyes on antibacterial properties

To investigate the effect of different dyes on the antibacterial activities of Rodalon first Microbiall cellulose fabric samples corresponding to the description recipe (in method) were dyed then treated with Rodalon according to the prescription in method. Finally, their antibacterial activity was evaluated against two kinds of bacteria E.coli and S.aureus according to AATCC test method 100–2004 and shown in figure 4 (according to the antibacterial performance percentage). The results indicate that in fabrics several dyeing may have different effects on the antibacterial property of a complementary material. Direct dyeing increases antibacterial property against two types of bacteria called E.coli and S.aureus; meanwhile, vat dyeing increases this property against S.aureus bacteria while decreasing it against E.coli bacteria. On the other hand, reactive dyeing decreases the total antibacterial property.

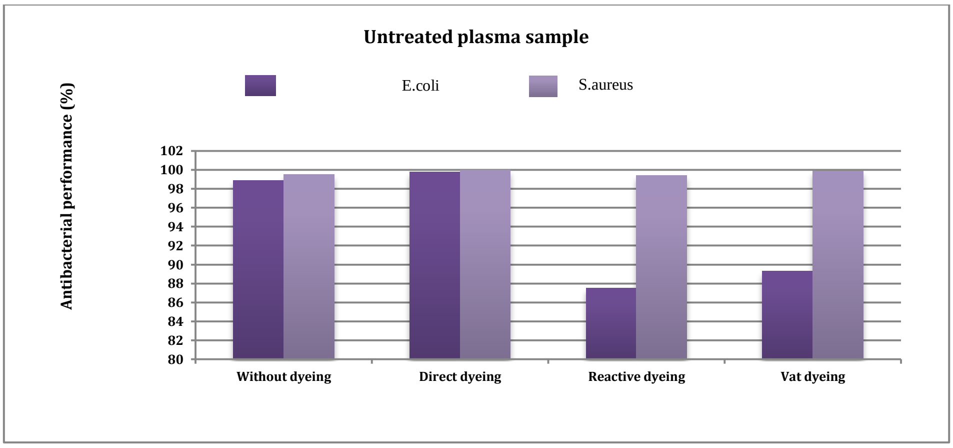


Figure 4: Effect of different kinds of dyes on antibacterial activity of Rodalon against two types of bacteria E.coli & S.aureus

The Effect of Plasma Treatment

The plasma treatment effects on antibacterial activity of raw and dyed fabrics are shown in figures (5,6) according to the antibacterial performance percentage. According to the obtained data it is observed that existence of plasma increases antibacterial property of specimens both in the presence or absence of different dyeing methods. In addition, provided that the specimens were washed for 10 times, the antibacterial property of specimens that were treated with plasma is higher in comparison to the other specimens that were not treated with plasma.

The Effect of Washing Procedure

The biocide can be gradually lost during the use and washing of the textile [20]. For the sake of inspection of repetitious washing procedures effects on antibacterial activity of Rodalon, all of the treated samples in different conditions were washed repetitively 10 times then their antibacterial activities were calculated. According to the obtained data, it is observed that the repeated washing procedure has a negative effect on Rodalon antibacterial material especially if different dyeing materials are used. Moreover, it is observed that there is a larger decrease in the antibacterial property after 10 times washing in comparison to the conditions where the specimen does not undergo dyeing. However, provided that the specimens have been treated with plasma, the antibacterial property has a smaller decrease as a result of the repeated washings. Washing procedure effects on antimicrobial activity of dyed fabrics are shown in (5,6) .

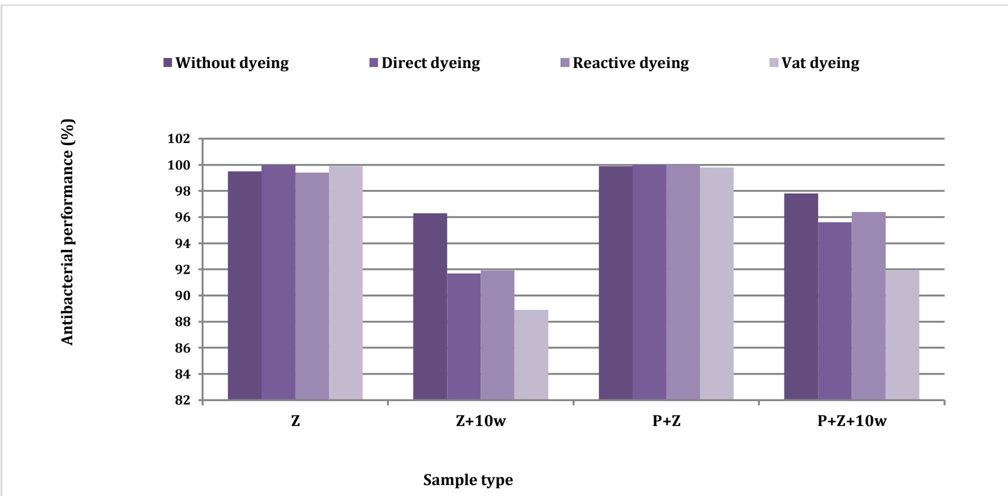


Figure 5: Antibacterial activity of simultaneous dyed and treated fabrics against Gram-positive bacteria (S.aureus AATCC 6538)

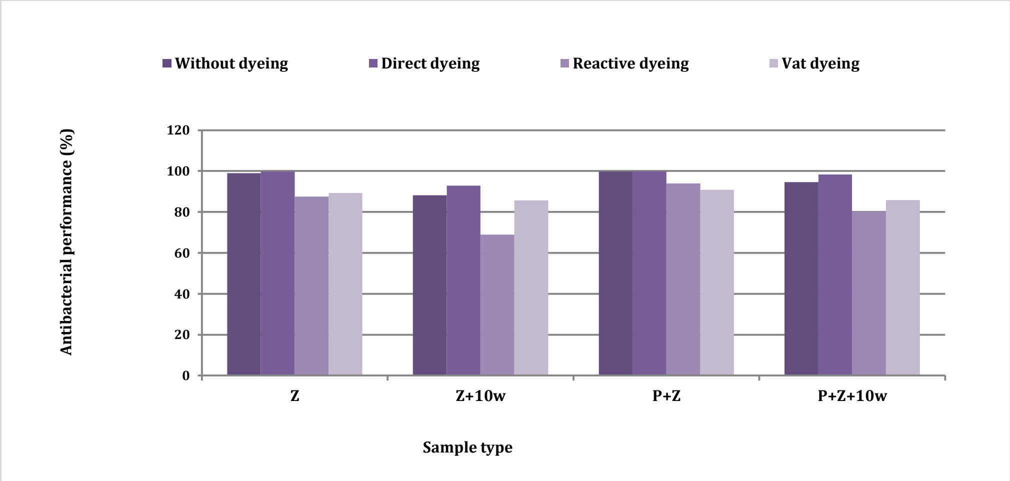


Figure 6: Antibacterial activity of simultaneous dyed and treated fabrics against Gram-negative bacteria (Escherichia coli AATCC8099)

The Effect of the treated fabrics on two Kinds of Bacteria

This section deals with studying antibacterial property of Rodalon on two types of positive gram and negative gram bacteria. According to the data, it is observed that the antibacterial property of Rodalon on Staphylococcus positive gram bacteria is higher in comparison to E.coli negative gram. The figures (5,6) show the antimicrobial activity of dyed and non-dyed fabrics.

Effect of Zycrobial on K/S values of fabrics

K/S values of dyed fabrics are calculated according to Kubeka-Munk formula in λ_{max}. Considering the obtained data in Table 2, it is found that Rodalon doesn't have any adverse effects on the color shades of fabrics tested and doesn't construct any alteration on it.

Table 2: K/S values of fabrics dyed in λ_{max}

Dyes	Without Zycrobial	Z	Z+10w	P+Z	P+Z+10 w
Direct	4.185	4.191	3.833	4.232	3.719
Reactive	1.862	1.818	1.753	1.780	1.654
Vat	1.892	1.944	1.849	2.018	1.850

The effect of Rodalon on mechanical properties of fabrics

The effect of 3% solution of Rodalon on some mechanical properties of the treated fabric in comparison with the untreated one is shown in Table 3. There is no significant deterioration effect on the studied mechanical properties (e.g. the significant level of α=0.05, about Table 3) [5].

Table 3: Mechanical properties of treated and untreated samples

The types of samples	Tenacity (g/dTex*10)	Initial Modulus (g/dTex)	Elongation @ rupture (mm)	Force @ rupture (Kgf*100)
Untreated	10.89	22.67	13.84	209.7
Treated	10.71	22.81	13.26	209.4

CONCLUSIONS

In this study, the dyed Bacterial cellulose fabrics were treated by Rodalon as an antimicrobial agent in various finishing conditions, and the dyed and treated fabrics were challenged with bacteria cultures. Enjoying an appropriate antibacterial effect, being less harmful to humans, easy to use and compatible with other agent materials are the main reasons for selection of Rodalon as an antiseptic in this study. All the treated fabrics exhibited antimicrobial efficacy against E.coli and S. aureus. A comparison of antibacterial effects of Rodalon on fabrics, which are dyed with different dyes, demonstrated the highest antibacterial effect appeared on those fabrics dyed with a direct dye. This is due to the large size of this molecule compared to other dyes and existence of numerous sites with negative charges that could create an affinity on Rodalon and the fiber, whereas reactive dyes have the least effective antibacterial property due to their small structure and resistance to establishing a link with Rodalon. Plasma finishing has a positive effect on the antibacterial activity of Rodalon and improves durability of the antibacterial effect in comparison with domestic washing procedures. The measurement of K/S values of same fabrics further confirmed that washing might cause a dye loss. Rodalon finishing on the fabric doesn't have incompatible effects on the mechanical properties and creates a soft surface and no changes on the shade of the colors of the dyed fabrics.

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