Available online www.jocpr.com

Journal of Chemical and Pharmaceutical Research, 2015, 7(10):181-193

Research Article

ISSN : 0975-7384
CODEN(USA) : JCPRC5

Effect of treatment durability and coloration of coated cotton fabrics on antibacterial, UV-blocking, healing and anti-inflammatory properties

[1]Abeer A. A. Salama, [2]Rehab M. Kotb and [2]Rania N. Shaker

[1]Pharmacological Department, National Research Centre, Cairo, Egypt
[2]Textile and Clothing Department, Faculty of Women for Arts, Science, and Education, Ain Shams University, Cairo, Egypt

---

ABSTRACT

In the last few decades, there is a rising interest in personal health and hygiene textiles with protective properties are becoming an increasingly desirable aim of textile researchers, manufactures, and consumers. In this research modified cotton fabric surface using chitosan and/or alginate biopolymers, as well as titanium dioxide, zinc oxide, and their mixtures to impart vital and protective characteristics. The coated cotton fabrics were undergoing durability evaluation to repeated washing of antibacterial properties against Escherichia coli (E. coli) (Gram negative bacterium) and Staphylococcus aureus (S. aureus) (Gram positive bacterium) and the results revealed that excellent reduction% sustained up 30 washes. As well as very good and excellent UV protection category were maintained even after 30 washes. Healing and anti-inflammatory properties showed sustainability up to 10 washes but decreased after 30 washes. Furthermore, application of post-dyeing with reactive dye on the coated fabrics was done and the pre-treatment step to assess their dye-ability; showed a significant effect on the color strength (K/S) values. Hence, it could be proposed that the coated cotton fabrics maintained their protective properties as well as suitable for color coloration, which in turn make these finished and dyed cotton fabrics available for different textile and apparel applications.

Keywords: Antibacterial, UV-blocking, anti-inflammatory, chitosan, alginate, metal oxides, dyeing, textile applications.

INTRODUCTION

In recent years, considerable attention has been paid to functional finishing for textile materials, such as antibacterial activity, ultraviolet (UV) protection, self-cleaning and wrinkle-free properties. Among these properties, the interest for antibacterial activity has been rapidly increasing, because all kinds of textile products, especially those made of natural fibers are sensitive to contamination and growth of pathogenic microorganisms [1]. The consumers are demanding textile products with higher performances, even in the traditional clothing and home textiles areas as the world market of textiles is becoming highly competitive [2].

Antibacterial finishing of textiles protects users from pathogenic or odor-generating microorganisms, which can cause medical and hygienic problems, and protects textiles from undesirable aesthetic changes or damage caused by rotting, which can result in reduced functionality [3]. Textiles have long been recognized as media to support the growth of microorganisms such as bacteria and fungi. These microorganisms are found almost everywhere in the environment and can multiply quickly when basic requirements, such as moisture, nutrients and temperature are met .Consumers' demand for hygienic clothing and active wear has created a substantial market for antimicrobial textile products. Several antimicrobial agents have been used in the textile industry, chitosan, is the most non-toxic, biodegradable and biocompatible one [4]. Chitosan is a deacetylated derivate of chitin, which is a natural polysaccharide mainly derived from the shells of shrimps and other sea crustaceans. Chemically, it can be designated as poly-β-(1→4)-D-glucosamine or poly-(1,4)2-amido-deoxy-β-D-glucoses [3]. Chitosan is a polycationic biopolymer, which has a wide spectrum of biological activity against bacteria, fungi; as well as it has haemostatic properties and can help in process of wound healing [5].

Alginate has been used in wound dressing, as it is high absorbent material; therefore it is very appropriate for highly exuding wounds [6]. The alginate as acidic linear polysaccharide composed of cell wall and intercellular cementing matrix algae can be converted into hydrophilic gel. This material provides a moist wound environment which promotes healing and epidermal regeneration. When using chitosan with alginate, they compose polyelectrolyte complexes (PEC) of oppositely charged polymers which have advantages when applied as coating materials and controlled release delivery carriers [7].

In addition, the harmful effects of ultraviolet (UV) radiation led to a considerable need for a photo-protection [8].The most popular choices for protection from UV radiation are UV blocking textiles and sun blocking creams .There are two types of UV blockers, the organic blockers or UV absorber since they absorb the UV rays and inorganic blockers which are usually certain semiconductor oxides such as TiO_2, ZnO, SiO_2, and Al_2O_3. Inorganic blockers are the preferred according to their exclusive features, non-toxicity, and chemical stability under both high temperature and UV -ray exposure [9] .Consequently, several studies have been carried out to block the textile fabrics against UV radiation [10,11].

Over the past few decades, modification of natural fibers and functional finishing of textiles have attracted tremendous attention because of increasing awareness of human beings toward environmental protection and healthy, safe, and comfortable life [12]. Cotton fibers are the most important fibers in the apparel industry, since they can readily absorb moisture and cotton-made clothes are the most comfortable garments [13]. More recently, an awareness of general sanitation, contact disease transmission, and personal protection have led to the development of antibacterial fibers to protect wearers against the spread of bacteria and diseases rather than to protect the quality and durability of the textile material [14]. For this reason, consumer demand for hygienic products has dramatically increased the use of antimicrobial substances on textiles such as socks, underwear, medical textiles and sportswear [1] .Also For old people who are permanently staying in bed or spend most of the time in bed, the comfort properties are crucial, as the risk for bedsores is high [15].

Reactive dyes are popular dyestuff for cellulose dyeing, which is the most worldwide acceptable dye for the coloration of cotton goods due to their ease of applicability, cost, brilliancy of color, and high wet fastness properties [16,17]. Introduction of cationic sites within the cellulose is an effective way to increase the dye adsorption. Cationic sites can be introduced either by aminization or cationization. Treatment of cotton with chitosan is an aminization technique to introduce cationic site within the fiber polymer structure and increase the hydroxyl group in the fiber for dye absorption [18, 19].

As much as is needed to protect textile fabrics from surrounding environmental hazardous such as bacterial attack, UV radiation, injuring and inflammation, they still require durability of applied protective coating as well as dyeing treatment to give the fabric or garment the functional and attractive value. In the present study, an attempt was made to evaluate durability of antibacterial, UV-blocking, healing and anti-inflammatory properties of cotton fabric coated with different mixtures. Additionally, investigate dyeing properties and its effect on cotton fabric before and after dyeing on the aforementioned protective properties. Hence, application of these protective dyed textile fabrics for numerous end uses such as protective garments, medical textiles, sportswear, children apparels, uniforms, and even daily use outfits.

EXPERIMENTAL SECTION

2.1. Fabric material

Half bleached, (2/2) plain weave 100% cotton fabrics (140g/m^2) was purchased from SHATEX, Egypt.

2.2. Chemicals and Dyes

Chitosan (low molecular weight), sodium alginate, glacial acetic acid, citric acid, sodium hypophosphite, TiO_2, ZnO, peptone, beef extract, and agar, were of laboratory grade chemicals, a non ionic detergent Hostpal® CVL-EL, Miner binder SME2, non ionic dispersing agent of commercial grade, and Cibacron® Red LS-B (C.I. Reactive Red 270) dye was kindly supplied by Ciba-Geigy

2.3. Microorganisms

Escherichia coli (*E. coli*) (Gram negative bacterium) and *Staphylococcus aureus* (*S. aureus*) (Gram positive bacterium), were used for estimation of antibacterial activities.

2.4. Media

Nutrient broth/ agar medium: contains beef extract (3 g/l), peptone (5 g/l). For solid medium (15 g/l) agar was added. This medium was sterilized for 20 min at 121°C under pressure.

2.5. Pharmacological studies

2.5.1. Animals

Wiser male rats, weighing ranged from 125-150g, were used throughout the experiment for the study of the anti-inflammatory activity. The rats were obtained from the animal house colony of the National Research Centre, Dokki, Giza, Egypt. The animals were housed in standard metal cages in an air conditioned room at 22 ± 3˚C, 55 ± 5% humidity and provided with standard laboratory diet and water *adlibitum*. Experiments were performed between 9:00 and 15:00 h. Animal procedures were performed in accordance with the Ethics Committee of the National Research Centre and followed the recommendations of the National Institutes of Health Guide for Care and Use of Laboratory Animals (Publication No. 85-23, revised 1985).

2.5.2. Drugs and Chemicals

Indomethacin cream (1%) was obtained from Ramida Pharmaceutical Industries Co, Egypt. Carrageenan was obtained from Sigma, USA.

2.6. Methods

2.6.1. The preparation of different mixtures for fabric treatment

Different mixtures of the treatment solutions were prepared as described in our previously research achievement [5].

2.6.2. Fabric treatment

The cotton fabric samples were impregnated in the prepared mixtures individually at 60 °C for 20 min. and were padded two (dips and nips) at a wet pickup (100 %). The treated samples were batched at room temperature for 2 hours then dried at (80 °C for 5 minutes) followed by curing at (140 °C for 3 minutes).

2.6.3. Durability evaluation procedure

The treated cotton samples were washed repetitively for 10 and 30 washing cycles according to AATCC test method (124-2006). Then the treated and washed samples evaluated for antibacterial activity, UV-blocking ability, healing and anti-inflammatory properties after 10 and 30 washing cycles respectively.

2.6.4. Dyeing with reactive dyes

The blank and treated cotton fabrics were dyed by Cibacron® Red LS-B (C.I. Reactive Red 270) reactive dye as follow: [Cibacron® Red LS-B dye: (1%) dye, (15) g/l sodium sulphate, (10)g/l sodium carbonate, M:LR 1:50]. The dyeing process was carried out according to Figure (1).

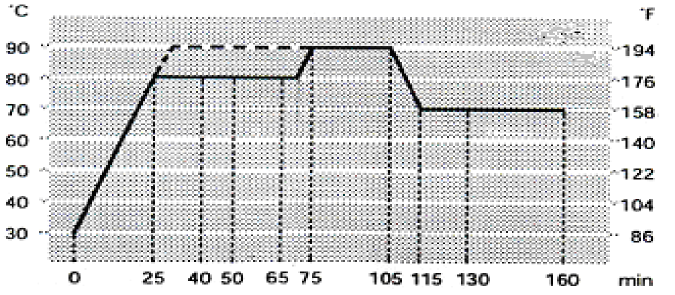


Figure (1): Dyeing process with Cibacron® Red LS-B dye

3. Testing and analysis

3.1. Antibacterial properties

The antibacterial properties were quantitatively evaluated against gram negative bacteria, *Escherichia coli* and gram positive bacteria *Staphylococcus aureus,* according to AATCC test method 100-1993. The reduction in numbers of bacteria was calculated using the following equation:

*Reduction rate (%) = (A-B)/A*100*

Where: A = the numbers of bacterial colonies recovered from untreated fabrics and
B = the numbers of bacterial colonies recovered from treated fabrics.

3.2. Ultraviolet protection factor (UPF)

In vitro testing measures ultraviolet (UVR) transmission and the ultraviolet protection factor (UPF) was calculated according to the Australian/NewZealand Standard (AS/NZS-4399-1996) using UV-Shimadzu 3101-PC-Spectrophotometer. The following equation which based on the percent ultraviolet radiation transmittance through the specimen used to calculate the UPF.

$$UPF = \frac{\sum E\lambda \cdot S\lambda \cdot \Delta\lambda}{\sum E\lambda \cdot S\lambda \cdot T\lambda \cdot \Delta\lambda}$$

Where; Where, $E\lambda$ is the relative erythemal spectral effectiveness, $S\lambda$ is solar spectral irradiance in $W/cm^2/nm$, $T\lambda$ is the spectral transmittance of the fabric (measured), λ is the wavelength in nm and $\Delta\lambda$ is the bandwidth in nm.

3.3. Carrageenan-induced paw oedema for healing and anti-inflammatory properties evaluation

Paw swelling was elicited by sub-plantar injection of 100 µl of 1% sterile carrageenan suspension in saline into the right hind paw [20]. Contra-lateral paw received an equal volume of saline. The oedema component of inflammation was quantified by measuring hind footpad immediately before carrageenan injection and 1-4h after carrageenan injection with a micrometer caliber [21]. Oedema was expressed as a percentage of change from control (pre-drug) values. Rats were divided into fifteen groups each of six. Blank and treated clothes were applied around the hind paw immediately after the injection of the carrageenan suspension. We use indomethacin cream (1%) as a reference anti-inflammatory drug which applied to blank tissue.

3.4. Color strength (K/S)

Color strength (K/S) of the dyed samples; in presence and/or absence of treatment; was assessed using Kubelka – Munk equation: $K/S = (1-R)^2/2R$

Where: K, S, and R are the absorption coefficient, scattering coefficient, and reflectance, respectively [22].

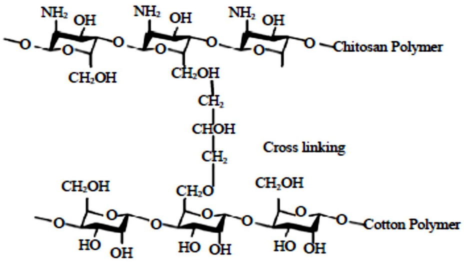
RESULTS AND DISCUSSION

4.1. Effect of treatment durability to repeated washing cycles on antibacterial properties of cotton fabrics

The durability and sustainability of coated samples with chitosan-alginate polymer; in presence and/or absence of ZnO, TiO_2, binder were subjected to assess the antibacterial efficiency against gram negative bacteria *E. coli* and gram positive bacteria *S. aureus.* The antibacterial activities results of coated samples after 10 and 30 washing are shown in Table (1) and could be discussed as following:

i. It can be stated that there are differences in reduction percent(%) between the two types of assessed bacteria regardless the treatment type after 10 and 30 washing cycles. Science, there are differences in the cell walls i.e.*E. coli* has thinner and slack cell walls, and the sensitivity to the finishing agents and/or types after repeated washing process[5,11,23]. As general observation from the obtained results, the antibacterial efficiency of all treated cotton fabrics did not affected by repeated washing cycles. Otherwise, they showed enhancement in bacterial reduction % regardless of the coating type for both *E. coli* and *S. aureus*.

ii. The antibacterial efficiency of samples coated with chitosan showed enhancement in antibacterial reduction %, increased after 10 < 30 washing cycles from 85% to 90% for *E.coli* and 83 to 92% for *S.aureus* respectively. First, it has proposed that chitosan antibacterial activity regarded to its polycationic nature that interferes with bacterial metabolism by stacking the cells' surface in addition to the binding between chitosan and DNA to inhibit mRNA synthesis [5,24]. The improvement and durability of antibacterial efficiency of chitosan coated samples may be due to that chitosan has been cross-linked to cotton fabric by citric acid. In the presence of citric acid; as a cross linking agent; as shown in Scheme (1)[18]; hydroxyl groups of chitosan and cellulose can form covalent bonds with carboxyl groups of polycarboxylic acid in an esterification reaction, thus leading to formation a crosslink between chitosan and cellulose, which greatly improves durability and wash resistance [3,25].



Scheme (1): Cross-linking of chitosan with cotton fiber polymer

iii. On the other hand, samples coated with alginate only enhanced from fair to good antibacterial efficiency; i.e., from 40% to 50 for *E.coli* and from 37% to 43% for *S.aureus* bacteria after 10 and 30 washes respectively. This may be attributed to the nature of alginate polymer, which has fair antiseptic, haemostatic, and antibacterial properties as well as the superior ability to promote wound healing [5]. Meanwhile, mixing chitosan with alginate it creates a polyelectrolyte complex (PEC) of oppositely charged polymers which have the a dual effectiveness and durability of chitosan when used as antibacterial coating for textile fabrics [5,7]. Consequently, sustained the antibacterial efficiency of the chitosan+alginate mixture coated samples from 93% to 96% for *E. coli* and from 88% to 90% for *S. aureus* respectively after 10 < 30 washes.

iv. It could be observed from Table (1) that, incorporating metal oxides i.e., ZnO and TiO_2 within treatment regime has a significant impact on the durability of the coated samples regardless the bacteria type. This may be attributed to the metal ions behavior, which is toxic to microbes at very low concentration. Since they kill microbes by binding to intracellular proteins, DNA, and lipids damaging them [4]. The sustained and durability of antibacterial efficiency in case of mixing ZnO and TiO_2 with the binder, may be due to that metal oxides are supported by the reaction with functional groups of the binder which cross-linking the whole system on fabric surface [26]. In addition to the removal of metal agglomeration from coated fabrics surface by repeated washing, such agglomeration has positive impact on antibacterial efficiency [5].

v. The maximum antibacterial reduction % that maintained up to 30 washing cycles was achieved with pre-loaded with metal oxides post-coated with chitosan+alginate mixture. Rising to 96% for *E. coli* and 94% for *S. aureus*in case of fabrics pre-loaded with ZnO post-coated with chitosan+alginate mixture; and 97% and94% in case of fabrics pre-loaded with TiO_2 post-coated with chitosan+alginate mixture for *E. coli* and *S. aureus* respectively. The excellent durability of those coated samples may be regarded to Zn and Ti metal ions are loaded and physically trapped firstly to the fabric subsequent coating with durable chitosan+alginate (PEC) polymer created another layer of protective and sustained coating film which promote durable antibacterial efficiency [5,27].

Table (1): Bacterial reduction% of chitosan, alginate, ZnO, and $TiO_2$and their mixtures coated fabrics after 10 and 30 washing cycles

Treatment Type	***Bacterial Reduction %***			
	After 10 washing cycles		***After 30 washing cycles***	
	E. coli	***S. aureus***	***E. coli***	***S. aureus***
Untreated	0%	0%	0%	0%
Chitosan	85%	83%	91%	92%
Alginate	40%	37%	50%	43%
Chitosan+alginate mixture	93%	88%	96%	90%
ZnO	86%	82%	91%	88%
ZnO + binder	89%	86%	95%	92%
ZnO+chitosan+alginate mixture	90%	87%	96%	93%
Pre-loaded with ZnO post-coated with chitosan+alginate mixture	90%	89%	96%	94%
Pre-coated with chitosan+alginate mixture post-loaded with ZnO	84%	82%	88%	85%
TiO_2	89%	88%	92%	90%
TiO_2 + binder	86%	81%	90%	88%
TiO_2+chitosan+alginate mixture	89%	84%	90%	87%
Pre-loaded with TiO_2 post-coated with chitosan+alginate mixture	93%	91%	97%	94%
Pre-coated with chitosan+alginate mixture post-loaded with TiO_2	85%	84 %	89%	87%

4.2. Effect of treatment durability to repeated washing cycles on Ultraviolet Protection Factor (UPF) of cotton fabrics

Textile clothing has the ability to protect the skin from harmful UV radiation, which depending on fiber chemistry, fabric weave, porosity, wetting, shrinkage, stretching, chemical treatments, and color[28].To quantify the protective ability of textiles, the UPF was determined. UPF calculated by measuring both UVA and UVB radiation transmittance, translating these results into mathematical expression. According to AS/NZS 4399:1996 the protection categories are; non-ratable protection UPF <15, good protection UPF>15, very good protection UPF>30, and excellent protection UPF>40, 50, 50+. [9]. The UPF results of all treated fabrics after 10 and 30 washing cycles were calculated in order to evaluate the durability of treated fabrics to repeated washing were presented in Table (2). The rate of UV protection of cotton fabrics was quantified and it is suggested that UPF of apparel and garment application should be at least 40 to 50+[10].

The results in Table (2) showed that UPF values of chitosan, alginate, and chitosan+alginate mixtures enhanced after repeated washing cycles i.e., 30>10 respectively rising there protection category from non-ratable to good protection in the order chitosan+alginate mixture (PEC) polymer > chitosan >alginate. The UPF results enhanced after repeated washing because of laundering process, which reduced the fabric porosity, associated with shrinkage [11]. However, treatment of cotton fabrics with chitosan does not have great effect on UV-blocking function [5,11,29]

It can be noted that, inorganic UV absorbers such TiO_2 and ZnOhave unique properties such as; non-toxic and chemical stable for UV-rays exposure. There provide good protection by reflecting and/or scattering most of the UV-rays, additionally they absorb UV radiation because of their semi conductive properties [5]. It is obvious from Table (2) that, the UPF values of all treated fabrics changed after repeated washing cycles. Although, the UPF values were decreased in the order 10 < 30 washing cycles regardless the mixture and metal oxide type. Which may be attributed to that the coating layer slightly loss their efficiency of UV-blocking power i.e., ZnO and TiO_2 particles are partially removed from the fabric [9]; they still achieving UPF rating of 40, 50, and 50+, which assigns the maximum UV protection [10]. Consequently, these results showed that treatment durability un affected by repeated washing, Furthermore, the UPF values of; Pre-coated with chitosan+alginate (PEC) polymer mixture post-loaded with ZnO; TiO_2+binder; and Pre-loaded with TiO_2 post-coated with chitosan+alginate (PEC) polymer mixtures. The UPF values for all coated fabrics in this study even after 30 washing cycles achieved good to excellent protection categories as shown in Table (2). The UPF values in these figures clearly showed that the minimum UPF values were >30 (very good protection) achieved with pre-loading cotton fabric with metal oxides post-coating with chitosan+alginate (PEC) polymer for ZnO. Those findings indicated that, excellent UV protection achieved with metal oxides (TiO_2 and ZnO) treatments, either alone or in mixtures. This obtained result may be attributed to that metal oxides particles were physically trapped and covered the entire fabric surface as well as good adhesion between coating films and fabrics surface, which resulting more area for diffuse reflection, scatting, and absorption of UV radiation [9,30,31]. Additionally, the reason for enhancements of UPF values after 30 washing cycles may be attributed to that prepared mixtures would induce the aggregation of metal oxides particles; which producing a lower scattering efficiency; were eliminated by repeated washing cycles [32,33].

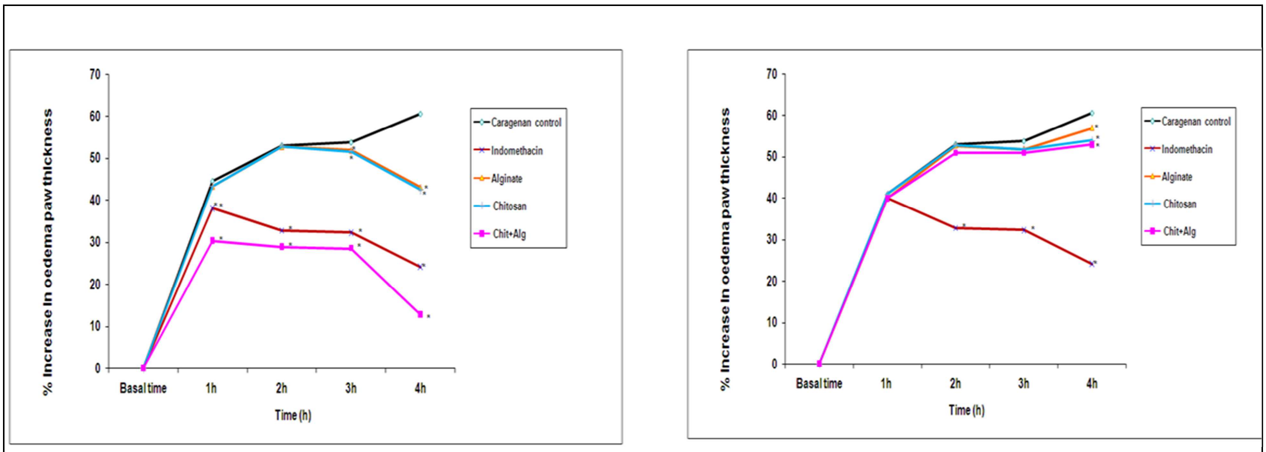
Table (2): Ultraviolet protection factor (UPF) of chitosan, alginate, ZnO, and $TiO_2$and their mixtures coated fabrics after 10 and 30 washing cycles

Treatment Type	**Ultraviolet Protection Factor (UPF)**	
	After 10 washing cycles	**After 30 washing cycles**
Untreated	5	
Chitosan	13	15
Alginate	9	13
Chitosan+alginate mixture	18	22
ZnO	50+ (170)	50+ (140)
ZnO + binder	50+ (100)	50+ (85)
ZnO+chitosan+alginate mixture	47	40
Pre-loaded with ZnO post-coated with chitosan+alginate mixture	31	30
Pre-coated with chitosan+alginate mixture post-loaded with ZnO	50+ (93)	50+ (116)
TiO_2	50+ (333)	50+ (240)
TiO_2 + binder	50+ (249)	50+ (255)
TiO_2+chitosan+alginate mixture	50+ (59)	50
Pre-loaded with TiO_2 post-coated with chitosan+alginate mixture	50+ (61)	50+ (85)
Pre-coated with chitosan+alginate mixture post-loaded with TiO_2	50+ (284)	50+ (230)

4.3. Effect of treatment durability to repeated washing cycles on healing and anti-inflammatory properties of cotton fabrics

I. The sub planter injection of 100µL of 1% sterile carrageenan into the rat hind paw elicited an inflammation (swelling and erythema) and a time-dependent increase in paw oedema by 41, 53 and 54% at 1^{st}, 2^{nd} and 3^{rd} hours respectively, and the paw thickness was maximal by 61% at 4h post-carrageenan injection as compared with pre-carrageenan control values. The results obtained for all coated fabrics after 30 repeated washing cycles are compared with results previously reported achievement [5]. It could be noticed that, healing and anti-inflammatory properties were affected by repeated washing although they still sustain good and adequate healing and anti-inflammatory effectiveness.

II. Coated samples with alginate or chitosan individually showed non-significant inhibition of oedema formation at 1st, 2nd and 3rd hours respectively, while induced a significant oedema inhibition by 6.56 and 11.48 % after 4th hour, respectively. Also, fabricscoated with chitosan+alginate mixture induced a significant oedema inhibition only after4th hours by 13.11% as compared with carrageenan control group at the same time post carrageenan injection; as shown in Figure (2).Data represent the mean value ± S.E. of six rats and % increase in oedema paw thickness. Data were analyzed using one-way ANOVA and LSD comparison test* significantly different from carrageenan control value at respective time point at $P<0.05$.

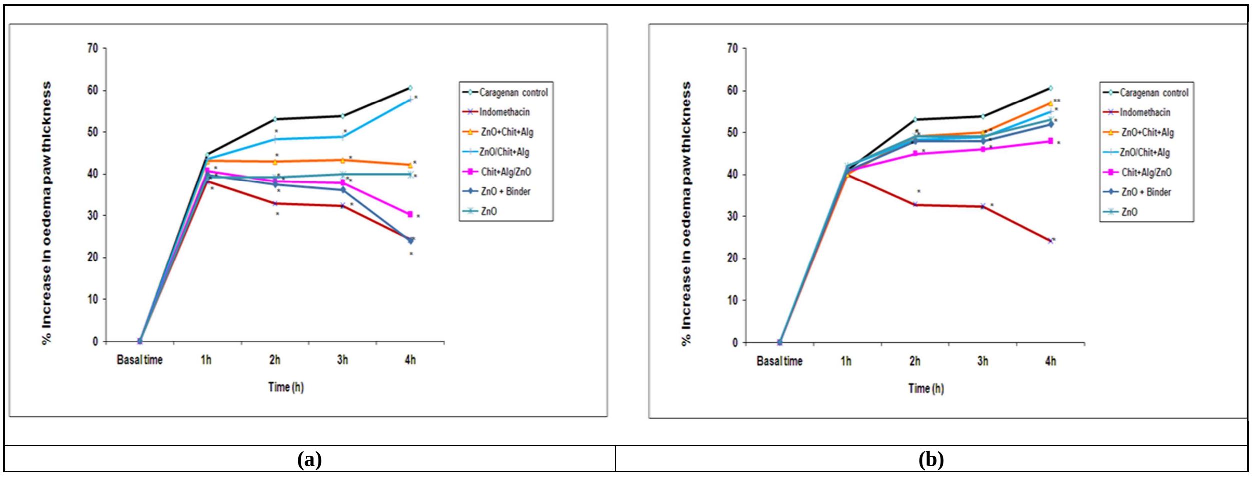


(a) (b)

Figures (2): Time course of the effects of cotton fabrics coated with alginate and/or chitosan after 10 and 30 washing cycles respectively, on rat paw oedema thickness induced by sub-plantar injection of 1% carrageenan. (a) after 10 and (b) after 30 washing cycles

III. The results of all coated fabrics were obtained after 10 and 30 washing cycles with indomethacin, results. It could be concluded that coated fabric with chitosan+alginate mixture has anti-inflammatory activity more than treated clothes with indomethacin after 10 washes as well as achieving the maximum healing and anti-inflammatory among the other coated mixtures. Since, chitosan+alginates (PEC) polymer mixture has haemostatic properties and can accelerate wound healing [5].Such polymer mixture has specific biological properties like haemostatic, granulation, and epithelization, which form ideal material suitable for application on wounds during the various healing phases. Both polymers are biodegradable and obtained from the natural origin and having a good bio-adhesion, which is necessary for the more retention over the skin. The alginate as acidic linear polysaccharide composed of cell wall and intercellular cementing matrix algae can be converted into hydrophilic gel. This hydrophilic gel mixture provides a moist wound and isolates it from environment, which promotes healing and epidermal regeneration [5,7]. While the decrease in healing and anti-inflammatory properties of coated samples containing alginate after 30 washes may be regarded to the alkali used in repeated washing bath, where alginate nature has insufficient resistance and stability to alkali [34].

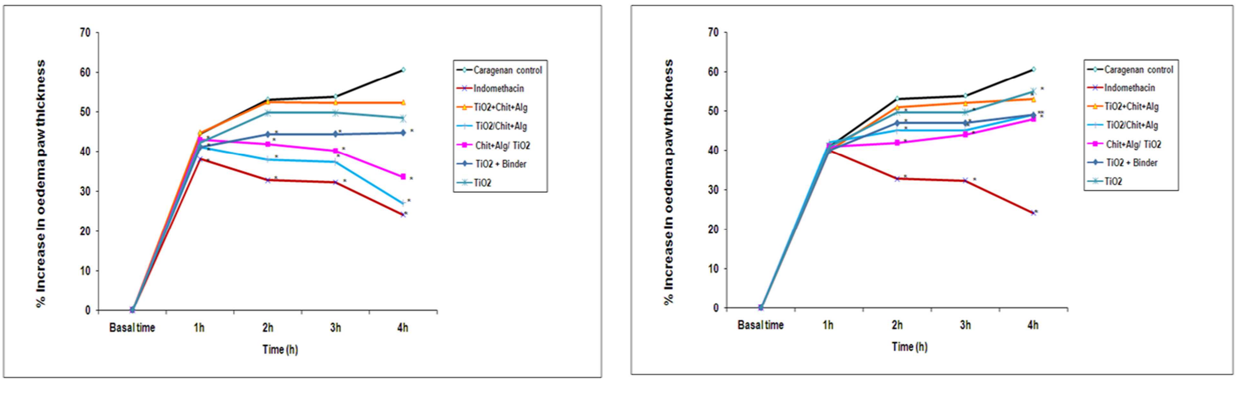
IV. After 30 washes, fabrics coated with ZnO/chitosan+alginate mixture or pre-loaded with ZnO post-treated with chitosan-alginate mixture showed non-significant inhibition of oedema formation at 1st hour, while ZnO+chitosan+alginate mixture induced significant oedema inhibition by 7.55, 7.41 and 6.56 % at 2nd, 3rd and 4th hours respectively. Similarly, samples pre-loaded with ZnO post-treated with chitosan-alginate mixture induced significant oedema inhibition by 8.93, 9.56 and 9.84 % at 2nd, 3rd and 4th hours respectively. In addition, pre-coated fabrics with chitosan+alginate mixture post-loaded with ZnO; coated fabric with ZnO+binder mixture; or ZnO individually; showed non-significant inhibition of oedema formation at 1st hour. While showing a significant oedema inhibition by 15.09, 9.43 and 7.55% at 2nd hour, 14.81, 11.11 and 9.26% at 3rd hour as well as 21.31, 14.75 and 13.11% at 4th hour, respectively, as compared with carrageenan control group at the same time post carrageenan injection; as shown in Figure (3). Data represent the mean value ± S.E. of six rats and % increase in oedema paw thickness. Data were analyzed using one way ANOVA and LSD comparison test* significantly different from carrageenan control value at respective time point at $P<0.05$.



(a) **(b)**

Figure (3): Time course of the effects of cotton fabrics coated with ZnO+chitosan+alginate mixture, pre-loaded with ZnO post-coated with chitosan+alginate, pre-coated with chitosan+alginate post-loaded with ZnO, ZnO+binder or ZnO on rat paw oedema thickness induced by sub-plantar injection of 1% carrageenan. (a) after 10 and (b) after 30 washing cycles

V. Coated fabric with TiO_2+chitosan+alginate mixture showed non-significant inhibition of oedema formation at 1^{st}, 2^{nd} and 3^{rd} hour while induced significant oedema inhibition by 13.11 % at 4^{th} hours. Meanwhile, pre-loaded fabrics with TiO_2 post-coated with chitosan+alginate mixture or pre-coated with chitosan+alginate mixture post-loaded with TiO_2; showed non-significant inhibition of oedema formation at 1^{st} hour while showing a significant oedema inhibition by 15.09 and 20.97% at 2^{nd} hour,16.67and 18.52% at 3^{rd} hour as well as 19.67 and 21.31% at 4^{th} hour respectively. In addition, coated fabrics with TiO_2+binder mixture or TiO_2 individuallyshowed non-significant inhibition of oedema formation at 1^{st} hour while showed a significant oedema inhibition by 11.32and 6.12% at 2^{nd} hour,12.96 and 7.89 at 3^{rd} hour as well as 19.67 and 9.84% at 4^{th} hour respectively. As compared with carrageenan control group at the same time; post carrageenan injection; as clearly showed in Figure (4). Data represent the mean value ± S.E. of six rats and % increase in oedema paw thickness. Data were analyzed using one way ANOVA and LSD comparison test.* significantly different from carrageenan control value at respective time point at $P<0.05$.



(a) **(b)**

Figure (4): Time course of the effects of fabrics coated with TiO_2+chitosan+alginate mixture, pre-loaded with TiO_2 post-coated with chitosan+alginate, pre-coated with chitosan+alginate post-loaded with TiO_2, TiO_2+binder or TiO_2on rat paw oedema thickness induced by sub-plantar injection of 1% carrageenan. (a) after 10 and (b) after 30 washing cycles.

VI. The healing and anti-inflammatory effectiveness of metal oxides may be regarded to the fact that ZnO helps to restore the disturbed skin-barrier function in eczematous diseases and enhances wound healing, it considered safe to use, since it does not penetrate the skin, even with disturbed barrier function. Moreover, ZnO has excellent anti-inflammatory; drying; mild astringent; and antiseptic properties hence, help in wound healing process[5].

4.4. Effect of dyeing process on antibacterial properties of coated samples

The effect and possibility of coloration process with Cibacron® Red LS-B (C.I. Reactive Red 270) reactive dye on untreated cotton fabric as well as those coated with different mixtures on the antibacterial efficiency against *Escherichia coli* (*E. coli*) (Gram negative bacterium) and *Staphylococcus aureus* (*S. aureus*) (Gram positive bacterium) was evaluated. Based on the data on Table (3), it can be concluded that even the samples dyed with reactive dye alone without any treatment application afforded antibacterial activity (17% and 12%) against *E.* coli and *S. aureus* respectively. In addition, it could be noticed from the obtained data that post dyeing process had a significant positive impact on the bacterial reduction % regardless the used treatment type. Consequently, the optimum bacterial reduction % was afforded by;pre-loaded with TiO_2 post-coated with chitosan+alginate mixture (98%) against *E.* coli and chitosan polymer coating (95%) against *S. aureus*. This behavior may be due to the inherent antibacterial characteristics of this class of bi-functional reactive dye, which is anionic dye and has sulfonate groups ($-SO^{3-}$, as water-solubiizing groups), bromine, fluorine, and chlorine groups in their structures. Halogen biocides such as Cl_2, Br_2 and I_2 are powerful oxidizing agents which having bactericidal and fungicidal activity. Halogen compounds affect microorganisms by attacking the cell membrane to get into the cytoplasm and affect the enzymes of the microorganisms, hence enhance antibacterial properties of dyed and pre-treated post-dyed cotton fabrics [17].

Table (3): Bacterial reduction percentage before and after dyeing with Cibacron® Red LS-B (C.I. Reactive Red 270) reactive dye

Treatment Type	*Bacterial Reduction %*			
	Treated Only		Pretreated Post-dyed	
	E. coli	*S. aureus*	*E. coli*	*S. aureus*
Untreated	0%	0%	17%	12%
Chitosan	87%	84%	97%	95%
Alginate	44%	41%	60%	58%
Chitosan+alginate mixture	93%	88%	96%	93%
ZnO	87%	83%	91%	87%
ZnO + binder	89%	86%	90%	88%
ZnO+chitosan+alginate mixture	91%	87%	95%	93%
Pre-loaded with ZnO post-coated with chitosan+alginate mixture	90%	88%	94%	91%
Pre-coated with chitosan+alginate mixture post-loaded with ZnO	91%	89%	93%	92%
TiO_2	89%	88%	92%	88%
TiO_2 + binder	92%	89%	94%	92%
TiO_2+chitosan+alginate mixture	92%	88%	93%	90%
Pre-loaded with TiO_2 post-coated with chitosan+alginate mixture	94%	91%	98%	95%
Pre-coated with chitosan+alginate mixture post-loaded with TiO_2	95%	88 %	92%	90%

Table (4): Ultraviolet protection factor (UPF) before and after dyeing with Cibacron® Red LS-B (C.I. Reactive Red 270) reactive dye

Treatment Type	Ultraviolet Protection Factor (UPF)	
	Treated Only	Pretreated Post-dyed
Untreated	5	50+ (57)
Chitosan	12	50+ (70)
Alginate	7	26
Chitosan+alginate mixture	15	50+ (55)
ZnO	50+ (185)	50+ (240)
ZnO + binder	50+ (123)	50+ (150)
ZnO+chitosan+alginate mixture	45	50+ (100)
Pre-loaded with ZnO post-coated with chitosan+alginate mixture	32	50+ (75)
Pre-coated with chitosan+alginate mixture post-loaded with ZnO	50+ (88)	50+ (125)
TiO_2	50+ (390)	50+ (418)
TiO_2 + binder	50+ (241)	50+ (261)
TiO_2+chitosan+alginate mixture	50+ (65)	50+ (120)
Pre-loaded with TiO_2 post-coated with chitosan+alginate mixture	44	50+ (135)
Pre-coated with chitosan+alginate mixture post-loaded with TiO_2	50+ (325)	50+ (400)

4.5. Effect of dyeing process on Ultraviolet Protection Factor (UPF) of coated samples

In order to assess the effect of dyeing process on UV-blocking ability, all the pre-treated and untreated fabric samples were dyed with Cibacron® Red LS-B (C.I. Reactive Red 270) reactive dye; subsequently their UPF values were calculated. The results of UPF calculated values are presented in Table (4). It is obvious that, white fabric sample dose not posses any UV protection and not suitable to protect against UV radiation. On the other hand, the UPF value of the untreated fabric sample was significantly enhanced after dyeing process, achieving excellent UV protection category 50+ (57). The obtained result may be attributed to the fact that color has a great effect on UV protection. The dyes used to color textile materials can have a considerable influence on their permeability to ultraviolet radiation. Depending on not only their specific chemical structural attributes, absorptive groups present in the dyestuff, but also dye molecular geometry, since the absorption band of many dyes extends in to the ultraviolet spectral region [25]. As a result, such dyes acts as ultraviolet absorbers and increases the UPF of the fabric. Meanwhile, dyes disrupt ultraviolet light because they include "conjugated" molecules that disrupt ultraviolet radiation, hence enhancing UPF of textiles[25]. Otherwise, dyes itself are susceptible for fading, deterioration and loss their fastness because of the degradation action of UV. Hence, such UV-blocking treatments are necessary to protect the dyed textile fabrics. It is expected that coating films contain metal oxides (TiO_2 and ZnO) would significantly prolong the lifetime of outdoor textile products by effectively blocking UV rays on the fiber surfaces

[35]. It is also worth mentioning that the UPF values of the pretreated post-dyed fabric samples were increased and have excellent UV protection category (50+) regardless the treatment type, except the pretreated with alginate only fabric sample. The increasing of UPF values of pretreated post-dyed fabric samples is reasonable, and could be discussed in terms of the dual action of protective coating layer, as well as the dyeing process abilities to block and protect these fabric samples against UV radiation.

4.6. Effect of dyeing process on healing and anti-inflammatory properties of coated samples

I. The sub planter injection of 100µL of 1% sterile carrageenan into the rat hind paw elicited an inflammation (swelling and erythema) and a time-dependent increase in paw oedema by 46, 57 and 58 % at 1st, 2nd and 3rd hours respectively, and the paw thickness was maximal by 68 % at 4h post-carrageenan injection as compared with pre-carrageenan control values.

II. The results of fabrics coated with alginate and/or chitosan induced a significant oedema inhibition by 8.70, 13.04 and 8.70 % after1st hour 31.58, 40.35and 33.33 % after 2nd hour 34.48, 43.10 and 36.21 % after3rd hour and 55.88, 63.24 and 57.35 % after4th hour respectively, as compared with carrageenan control group at the same time post carrageenan injection; as shown in Figure (5).Data represent the mean value ± S.E. of six rats and % increase in oedema paw thickness. Data were analyzed using one way ANOVA and LSD comparison test *significantly different from carrageenan control value at respective time point at $P<0.05$.

III. Fabrics coated with indomethacin induced a significant oedema inhibition by 13.04, 42.40, 44.24and 64.39 % after 1st, 2nd, 3rdand4th hour, respectively, as compared with carrageenan control group at the same time post carrageenan injection.

IV. Blank dyed fabrics showed non-significant inhibition of oedema formation at 1st, hours respectively, while induced a significant oedema inhibition by 5.26,6.90 and 22.06 % after 2nd, 3rdand4th hour, respectively, as compared with carrageenan control group at the same time post carrageenan injection.

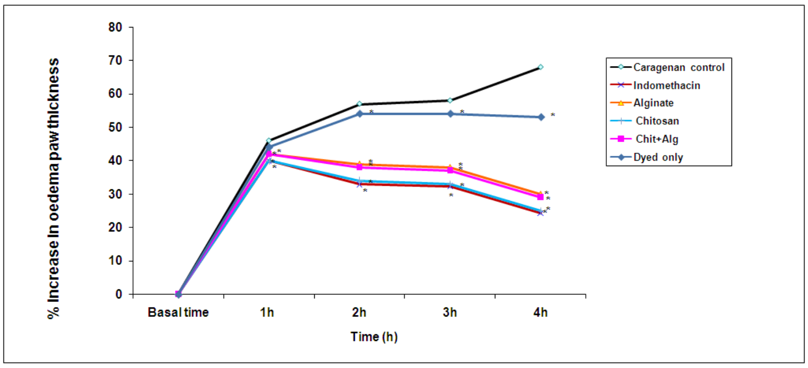


Figure (5): Time course of the effects of pre-coated fabrics with alginate and/or chitosan post-dyed with Cibacron® Red LS-B (C.I. Reactive Red 270) reactive dye on rat paw oedema thickness induced by sub-plantar injection of 1% carrageenan

V. Fabrics coated with ZnO+chitosan+alginate mixture induced significant oedema inhibition by 13.04, 49.12, 51.72 and 70.59% at 1st, 2nd, 3rd and 4th hours respectively, similarly preloaded with ZnO post-treated with chitosan+alginate mixture induced significant oedema inhibition by 8.70, 47.37, 50.00 and 69.12 % at 1st , 2nd, 3rd and 4th hours respectively. Meanwhile, pre-coated fabrics with chitosan+alginate mixture post-loaded with ZnO; showed a significant oedema inhibition by 6.52, 40.35, 43.10 and 64.71% at 1st, 2nd, 3rd and 4th hours. Also, coated fabrics with ZnO+binder mixture; or ZnO individually showed a significant oedema inhibition by 10.87, 49.12, 51.72 and 69.12% as well as 8.70, 31.58, 34.48 and 55.88% at 1st , 2nd, 3rd and 4th hours respectively, as compared with carrageenan control group at the same time post carrageenan injection; as shown in Figure (6). Data represent the mean value ± S.E. of six rats and % increase in oedema paw thickness. Data were analyzed using one way ANOVA and LSD comparison test* significantly different from carrageenan control value at respective time point at $P<0.05$.

VI. Fabrics coated with TiO_2+chitosan+alginate mixture induced significant oedema inhibition by 6.52, 10.53, 10.34 and 30.88% at 1st, 2nd, 3rd and 4th hours respectively, similarly pre-loaded with TiO_2 post-treated with chitosan-alginate mixture induced significant oedema inhibition by 10.87, 40.35, 43.10 and 64.71 % at 1st,2nd, 3rd and 4th hours respectively. Meanwhile, pre-coated fabric with chitosan+alginate mixture post-loaded with TiO_2; showed a significant oedema inhibition by 13.04, 45.61, 48.28 and 69.12% at 1st, 2nd, 3rd and 4th hours. Also, coated fabric with TiO_2+binder mixture; or TiO_2 individually showed a significant oedema inhibition by 10.87, 38.60, 41.38 and 64.71% as well as 10.87, 15.79, 18.97 and 45.59% at 1st, 2nd, 3rd and 4th hours respectively, when compared with carrageenan control group at the same time post carrageenan injection; as shown in Figure (7).Data represent the mean value ± S.E. of six rats and % increase in oedema paw thickness. Data were analyzed using one way ANOVA and LSD comparison test * significantly different from carrageenan control value at respective time point at $P<0.05$.

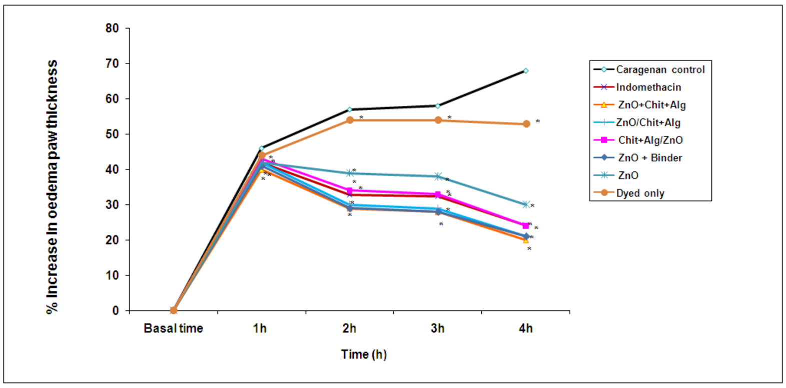


Figure (6): Time course of the effects of pre-coated with ZnO+chit+alg, ZnO/chit+alg, ZnO+chit+alg, chit+alg/ZnO, ZnO + binder or ZnO post-dyed withCibacron® Red LS-B (C.I. Reactive Red 270) reactive dye on rat paw oedema thickness induced by sub-plantar injection of 1% carrageenan

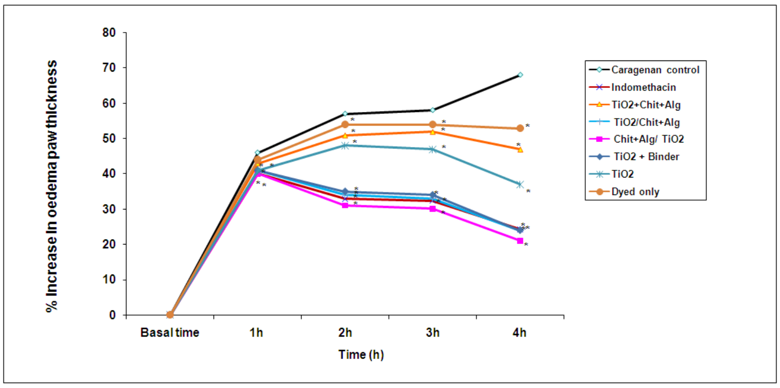


Figure (7): Time course of the effects of pre-coated fabrics with TiO_2+chit+alg, TiO_2/chit+alg, chit+alg/ TiO_2, TiO_2 + binder or TiO_2post-dyed withCibacron® Red LS-B (C.I. Reactive Red 270) reactive dye on rat paw oedema thickness induced by sub-plantar injection of 1% carrageenan

VII. By comparing the results of all pre-coated fabrics post-dyed; using Cibacron® Red LS-B (C.I. Reactive Red 270) reactive dye; with carrageenan control indomethacin and, results revealed that dyed only fabric samples showed a little improvement in healing and anti-inflammatory properties. Moreover, pre-coated post-dyed fabric samples showed significant and sustained enhancement in healing and anti-inflammatory properties regardless the coating mixture type. These obtained results may be thanked to this behavior may the inherent antibacterial characteristics of this class of bi-functional reactive dyes. Which is anionic dyes, has sulfonate groups ($-SO3-$, as water-solubiizing groups),Most suitable of this type of dye is Cibacron® LS range. Where, two hetro-bifunctional groups mixed to form multifunctional reactive dye, having several different types of reactive groups. Monoflourotriazine and monochlorotriazine in combination with sulphone can be used [16]. Halogen biocides are powerful oxidizing agents which having bactericidal and fungicidal activity. Halogen compounds affect microorganisms by attacking the cell membrane to get into the cytoplasm and affect the enzymes of the microorganisms [17]. These excellent antibacterial activities in turn, accelerate the process of wound healing and promote anti-inflammation properties.

4.7. Effect of different treatment types on K/S values

There is a point of view, that dye-ability of finished fabrics are quite crucial. In a complex scenario, using different finishing and dyeing substances and treatments can make a considerable difference in end products. There is no experienced evidence to shed light on dye-ability of fabrics after treatments, and any possible chemical interaction with structural properties (especially under exhaustive finishing and dyeing process) of the cotton fabrics [36]. The results in Table (5) represented the effect of different pre-treatment mixtures on the dyeing affinity and color strength (K/S) of cotton fabric. It is obvious from the data obtained that cotton samples pretreated with chitosan biopolymer achieved the maximum K/S values over the untreated post-dyed and the all treatment type (10.5). Moreover, the K/S values of pre-treated and/or simultaneously treated with chitosan showed significant enhancement and were higher than the dyed only fabric according to the following order; ZnO+chitosan+alginate mixture (8.4) > pre-coated with chitosan+alginate mixture post-loaded with ZnO (7.8) >pre-coated with chitosan+alginate mixture post-loaded with TiO_2 (7.5) > TiO_2+chitosan+alginate mixture (7.3) > dyed only (6.7).These enhancements may be attributed to treatment of cotton fiber with chitosan, which is an aminization technique to introduce cationic site within the fiber polymer structure and increase the hydroxyl group in the fiber for dye absorption. Cotton fiber forms a crosslink with chitosan, resulting positive dye sites on the fiber surface. Chitosan can easily adsorb anionic dyes such as reactive dyes by electrostatic attraction due to its cationic nature. Hence, offering the possibility of increasing the hydroxyl group for the formation of covalent bond with reactive dye and develop desired level of exhaustion [17,18,19]. On the other hand, the pre-treatment with chitosan+alginate mixture; and pre-loaded with ZnO post-coated with chitosan+alginate mixture; insignificantly affected the K/S values after dyeing process.

Otherwise, it can be conclude also from Table (5) that K/S values of dyed fabrics after alginate; ZnO; ZnO+binder; TiO_2; TiO_2 + binder; pre-loaded with TiO_2 post-coated with chitosan+alginate; mixtures finishing were lower than the dyed only fabric. The decrease in K/S values can be explained on the basis that some hydroxyl groups of cotton fibers were consumed or blocked by treatments[37].

Table (5): Effect of different treatment mixtures on K/S values of coated cotton fabric samples

Treatment Type	Color Strength Value (K/S)
Dyed only	6.7
Chitosan	10.5
Alginate	3
Chitosan+alginate mixture	6.7
ZnO	6.1
ZnO + binder	5.1
ZnO+chitosan+alginate mixture	8.4
Pre-loaded with ZnO post-coated with chitosan+alginate mixture	6.4
Pre-coated with chitosan+alginate mixture post-loaded with ZnO	7.8
TiO_2	5
TiO_2 + binder	4.2
TiO_2+chitosan+alginate mixture	7.3
Pre-loaded with TiO_2 post-coated with chitosan+alginate mixture	5.6
Pre-coated with chitosan+alginate mixture post-loaded with TiO_2	7.5

CONCLUSION

In present research coated cotton fabrics were manipulated to obtain multifunctional, durable, and colored textiles and apparel products. The fabrics coated with chitosan and/or alginate, TiO_2, ZnO, and their mixtures were investigated to evaluate their durability to repeated washing. *Escherichia coli* (*E. coli*) and *Staphylococcus aureus* (*S. aureus)* were used to investigate the antibacterial activity to repeated washing cycles and the obtained results showed excellent reserved bacterial reduction % up to 30 washes regardless the coating mixtures and the maximum values were found in case of coating with pre-loaded with TiO_2 post-coated with chitosan+alginate mixture (97%) and (94%) against *E. coli* and *S. aureus* respectively after 30 washes. Moreover, UV-blocking properties of coated fabrics after 30 washes maintained their excellent protection category (40-50+), in case of incorporating TiO_2 and ZnO with chitosan and alginate mixtures. Although healing and anti-inflammatory properties of all coated fabrics using the carrageenan-induced rats paw oedema test maintained excellent up to 10 washes then decreased after 30 washes, they still achieve adequate requirements. As well as dyeing of untreated as well as all coated fabrics with Cibacron$^{®}$ Red LS-B (C.I. Reactive Red 270) reactive dye was done and the results showed that there were positive effects of this class of dye on the aforementioned protective properties, specially healing and ant-inflammatory properties which significantly increased after dyeing achieving better oedema inhibition more than indomethacin and the optimum healing activity was achieved in case of chitosan pre-coated and post-dyed fabric. Meanwhile, there wear enhancement in color strength (K/S) values of pre-treated post-dyed cotton fabrics especially in case of chitosan pre-coated and post-dyed fabric. Otherwise, there was a little decrease in K/S values specially induced in case ofpre-coated post-dyed Alginate fabric sample. Accordingly, it can be concluded from all the obtained results in this investigation that, durability of coated cotton fabrics in addition to their excellent behavior to post-dyeing with reactive dye make the final fabric suitable for different applications and endues such as medical, industrial, and apparel.

REFERENCES

[1] ME Üreyen; A Dog˘an; AS Koparal, *Textile Research Journal*, **2012,** 82(17), 1731–1742.
[2] S Kathirvelu; L D’Souza; B Dhurai, *Indian Journal of Science and Technology,* **2008, (**1), 1-12.
[3] B Simoncic; B Tomsic, *Textile Research Journal***2010**, 80(16), 1721–1737.
[4] Y Gao; R Cranston, *Textile Research Journal,* **2008**, 78(1), 60–72.
[5] RM Kotb; NA Elsayed; AA Salama, *Journal of Chemical and Pharmaceutical Research,* **2014**, 6(11), 900-912.
[6] Y Qin, *International Wound Journal,* **2005**, 2(2), 172-176
[7] OL Shanmugasundaram; R Gowda, *Journal of Industrial Textiles,* **2012**, 42(2), 156-175.

---

[8] R Rai; CR Srinivas, *Indian J. Dermatol. Venereol. Leprol.*, **2007**, 73(2), 73–79.
[9] H Yang; S Zhu; N Pan, *Journal of Applied Polymer Science*, **2004,** 92(5), 3201–3210.
[10] D Mihailovic´; Z Šaponjic´; M Radoicˇic´; T Radetic´; P Jovancˇic´; J Nedeljkovic´; M Radetic, *Carbohydrate Polymers*, **2010**, 79(3), 526-532.
[11] MS Khalilabad; ME Yazdanshenas, *Textile Research Journal*, **2013**, 83(10), 993-1004.
[12] Z Zhang; L Chen; J JI; Y Huang; D Chen, *Textile Research Journal*, **2003**, 73(12), 1103-1106.
[13] VA Dehabadi; H Buschmann; JS Gutmann, *Textile Research Journal*, **2013,** 83(18) 1974–1995.
[14] DT Chun; GR Gamble, *The Journal of Cotton Science*, **2007**, (11), 154–158.
[15] H Meinanderz; M Varheenmaa, Clothing and textiles for disabled and elderly people, 1st Edition, Julkaisija Utgivare Publisher, Finland, **2002**, 19-20.
[16] K Singha; S Maity; M Singha, *International Journal of Textile Science*, **2012**, 1(6), 69-77.
[17] M Hosseini; M Montazer; R Damerchely, *Journal of Engineered Fibers and Fabrics*, **2013**, 8(3), 102-111.
[18] MA Rahman Bhuiyan; A Shaid; MM Bashar; P Haque; MA Hannan, *Open Journal of Organic Polymer Materials*, **2013,** 3, 87-91.
[19] MA Rahman Bhuiyan; A Shaid; MA Khan, *Chemical and Materials Engineering*, **2014**, 2(4), 96-100.
[20] CA Winter; EA Risley; GW Nuss, *Proc. Soc. Exp. Biol. Med.*, **1962**, 111, 544-52.
[21] MG Obukowicz; DJ Welsch; WJ Salsgiver; CL Martin-Berger; KS Chinn; KL Duffin; A Raz; P Needleman, *J. Pharmacol. Exp. Ther.*, **1998**, 287(1), 157-66.
[22] F Jahangiri; A Ashjaran; B Mehravani, *World Applied Sciences Journal*, **2012**, 19(1), 63-69.
[23] D Hua; K Cheuk; Z Wei-ning; W Chen; X Chang-fa, *Transactions of Nonferrous Metals Society of China*, **2007**, 17, 700-703.
[24] T Ristić1; LF Zemljič; M Novak1; MK Kunčič1; S Sonjak; NG Cimerman; S Strnad, *Science against microbial pathogens: Communicating current research and technological advances*, **2011**, 1(3), 36- 51.
[25] D Enescu, *Roumanian Biotechnological Letters*, **2008,** 13(6), 4037-4048.
[26] V Čechalová; A Kalendová, *Journal of Physics and Chemistry of Solids*, **2007**, 68, 1096-1100.
[27] T Mikołajczyk; D Wołowska-Czapnik, *Fibers & Textiles in Eastern Europe*, **2005**, 13, 3(51), 35-40.
[28] MA Sharmila; RI Sathya, *Indian Journal of Applied Research*, **2014**, 4(12), 556-557.
[29] X Chen; Y Liu; H Shi; X Wang; K Qi1; X Zhou; JH Xin, *Textile Research Journal*, **2010**, 80(20), 2214–2222.
[30] BR Das, *The Open Textile Journal*, **2010**, 3, 14-21.
[31] M Miraftab; J Barnabas; JF Kennedy; R Masood, *Journal of Industrial Textiles*, **2011**, 40(4), 345-360.
[32] CH Xue; ST Jia; HZ Chen; M Wang, *Science and Technology of Advanced Materials*, **2008**, 9(3), 1-5.
[33] A. Elshafei; HT Elzanfaly, *Asian Journal of Applied Sciences*, **2011**, 4, 585-595.
[34] W Stęplewski; D Wawro; A Niekraszewicz; D Ciechańska. *Fibers & Textiles in Eastern Europe*, **2006**, 14, 4(58), 25-31.
[35] CH Goh; PW Heng; LW Chan, *Carbohydrate Polymers*, **2012**, 87(2), 1796– 1802.
[36] E Hezavehi; S Shahidi; P Zolgharnein, *AUTEX Research Journal*, **2014**, 15(2), 104–111.
[37] AM El-Naggar; MH Zohdy; MS Hassan; EM Khalil, *Journal of Applied Polymer Science*, **2003**, 88(5), 1129–1137.