



Production of Antibacterial Cotton Fabrics via Green Treatment with Nontoxic Natural Biopolymer Gelatin



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Herein we used gelatin, which is an organic, non-toxic, safe and inexpensive substance as antibacterial material for cotton fabrics. There are some studies deals with the topic of finishing of textile materials with gelatin, its antibacterial, antioxidant and anti-fungal activity and its used as bio-material for textiles finishing but these studies neglect the effect of gelatin antibacterial itself which increase our attention to study the antibacterial effect of gelatin itself without any harmful side effects. Pad-dry-cure method was used for cotton fabric which was treated with gelatin in the presence of ammonium dihydrogen phosphate as a catalyst and dimethyloldihydroxyethylene urea as a cross linker. Factors affecting this treatment were studied. Fourier Transform Infrared spectroscopy (FT-IR), physico-mechanical properties, thermal gravimetric analysis and scanning electron microscope analysis were used to characterize the treated cotton fabrics. Disk diffusion method used to assessment antibacterial activity of the treated fabrics. Cytotoxicity of gelatin compared with ciprofloxacin in viable A549 cells was evaluates by using MTT. Results show that treated cotton fabrics show excellent antibacterial activity towards tested bacteria and there is no observed toxic effects on mammalian cells so that gelatin can be used as safe antibacterial finishing agents of cotton fabrics.

Key words: Gelatin, Antibacterial activity, Modification, Cotton fabrics, Cytotoxicity, Green treatment.

1. Introduction

Textile materials form good media for microbial growth so that its antibacterial usage in hospitals without antibacterial properties is limited [1-4].

Surface modification of textiles provide desirable properties to textile without decreasing in both tensile and comfort properties. Nowadays functional finishing used to improve these textile materials with multifunctional properties especially that make these fabrics performance, durable, safe and ecofriendly. Based on these principles natural biopolymer are highly recommended for functional finishing to impart them multifunctional properties with more safety

[5-8].

Chemical finishing of fibers and fabrics has always been an essential part of textile wet processing, but in recent years the demand of 'high tech' textile products have amplified the interest and usage of chemical finishes. A number of chemical finishes is now being used to change textile materials into technical textiles with special properties [9-14].

Many research papers deals with finishing of textile materials with gelatin as natural biopolymer but most of them neglect its own antibacterial activity due to the presence of another antibacterial material such as chitosan [5, 15-17], zeolite [18], mono methoxy poly ethylene

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glycol-poly lactide hydrogel [19, 20], cerium (III) [21] and ZnO [22, 23]. Microencapsulation of gallic acid using agarose/gelatin system used safe antifungal material [24], in addition gelatin was used to encapsulate vitamin C for cosmetic textile applications [25-27]. Based on these studies gelation takes our attention as natural antibacterial biopolymer used for textiles finishing without any harmful side effects due to its functional groups.

amino acids, specially prolin, hydroxyproline and glycine. These amino acids form polypeptide chains by connecting with each other. The polypeptide chains contain more than a thousand amino acid. Gelatin has a stick-shaped molecule structure which composed of primary, secondary and tertiary helicoid structures (Fig. 1) [28]. The amino acids of gelatin and its basic properties are listing in Table 1 [28].

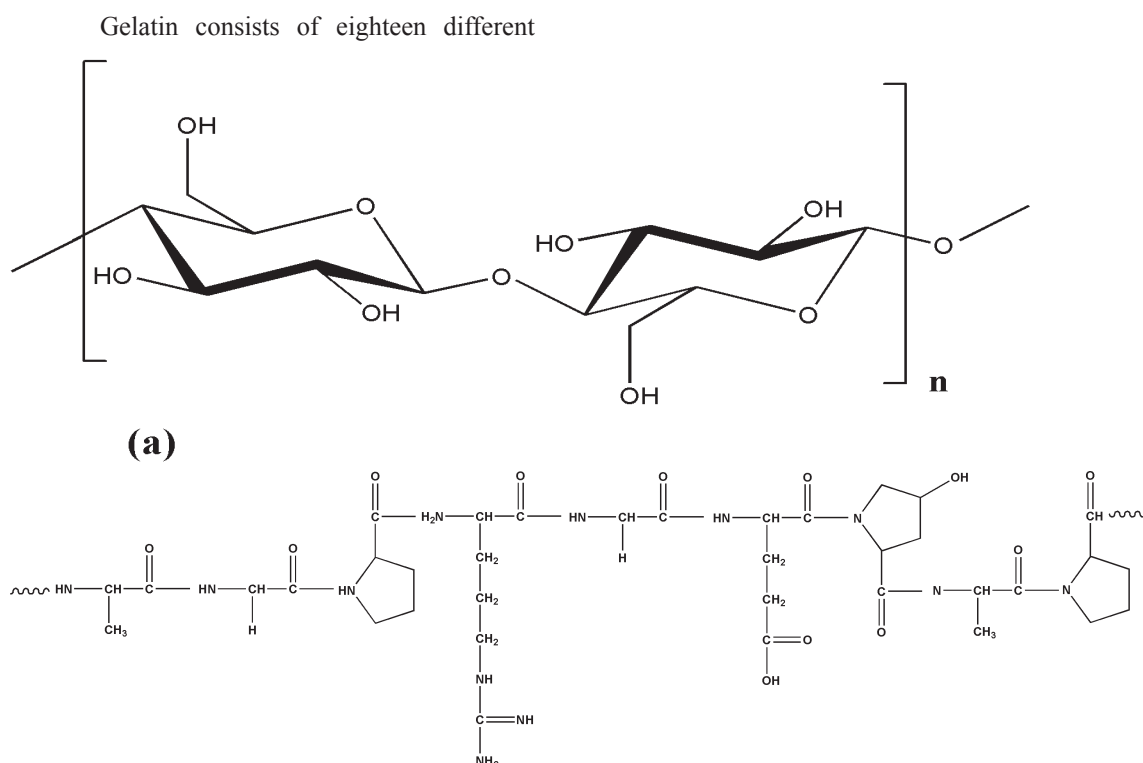


Fig. 1. Chemical structures of (a) cellulose, (b) gelatin

The present work aimed to use gelatin as safe antibacterial finishing material for cotton fabrics via pad-cure method. Several parameters were used to optimized this treatment such as effect of ammonium dihydrogen phosphate concentration, effect of gelatin concentrations, effect of DMDHEU concentrations and effect of the curing temperature and time. Gelatin treated cotton fabrics were characterized. The antibacterial activity of the finished fabrics was evaluated against disk diffusion method using both Gram positive and Gram negative bacteria. Scanning electron microscope (SEM) used to study the surface morphology of treated and untreated cotton fabrics.

Materials and Methods

Materials

Mill-scoured and bleached cotton fabric which was supplied by Misr Spinning and Weaving Co., Mehalla EL-Kobra, Egypt, was used. Gelatin, ammonium dihydrogen phosphate and dimethyloldihydroxyethylene urea (DMDHEU) were at analytical grade. Two bacterial strains from the bacterial lab, botany department, the faculty of women for art, science & Education, Ain shams university, Cairo, Egypt were employed. They include *Staphylococcus aureus* (*S. aureus*) as Gram-positive bacteria and *Escherichia coli* (*E. coli*) as Gram-negative bacteria. These bacterial strains were selected as test cells because they are the most frequent bacteria in the wound infection and represent Gram positive and Gram negative bacteria, respectively. Fresh inoculants for antibacterial assessment were prepared in nutrient broth at 37°C for 24 hours.

TABLE 1. Amino acids listing of gelatin and its basic properties

Amino acid name	Abbreviation ^a	Properties
Alanine	ala (A)	Hydrophobic (Nonpolar)Positively charged
Arginine	arg I	Negatively charged
Asparagine	asn(N)	Hydrophilic
Aspartic acid	asp (D)	Negatively charged
Cysteine	cys I	Hydrophilic
Glutamic acid	glu (E)	Hydrophobic
Glutamine	gln (Q)	Positively charged
Glycine	gly (G)	Hydrophobic
Histidine	his (H)	Hydrophobic
Isoleucine	ile (I)	Positively charged
Leucine	leu (L)	Hydrophobic
Lysine	Lys (K)	Hydrophobic
Methionine	met (M)	Hydrophobic
Phenyl-alanine	phe (F)	Hydrophobic
Proline	pro (P)	Hydrophobic
Serine	ser (S)	Hydrophobic
Threonine	thr (T)	Hydrophobic
Tryptophan	trp (W)	Hydrophobic
Tyrosine	tyr (Y)	Hydrophobic
Valine	Val (V)	Hydrophobic

^a Abbreviation for three and single letter code

Fabric treatment

Pad-dry-cure method was used for treatment and finishing of cotton fabrics with gelatin, where the padding-bath containing DMDHEU (0-80g/L), ammonium dihydrogen phosphate catalyst (1-3 g/L) and the gelatin concentrations (3-10 % w/v) along with a non-ionic wetting agent. The fabric was padded to 100% wet pick up, then fixed, dried at 80 °C for 5 min and cured at different temperatures and times. All cured samples were washed by water at 50 °C for 5 min and conditioned before testing and analysis.

Testing and analysis

- Nitrogen content was determined by ASTM Method E258-67 method.
- The tensile strength (TS) and elongation at break EL. (%) of untreated and treated cotton fabrics were tested in the warp direction according to ASTM D-2256-98.
- Stiffness (S) was determined in the warp direction according to ASTM Test Method D 1388-96 using Jika (Toyaseiki) apparatus.
- Surface roughness (SR) was measured using a Surfacer 1700a.
- Air permeability (AP) was evaluated according to ATSM (D 737-96). The air permeability of a fabric is the air flow passing through that fabric under a given air pressure.
- Thermo gravimetric analysis (TGA) was performed at a temperature starting from 25 °C to 800 °C under inert nitrogen atmosphere with heating rate of 10 °C min⁻¹ using the instrument: SDT Q600 V20.9 Build 20, USA.
- Fourier transforms infrared spectra (FT-IR) measured at a JASCO FT-IR-6100 spectrophotometer using the KBr pellet disk method for transmittance measurements.
- The surface morphology of untreated and treated cotton fabrics were obtained by using Scanning electron microscope (SEM) images, Model Quanta 250 FEG (Field Emission Gun) attached with EDX Unit (Energy Dispersive X-ray Analyses), with accelerating voltage 30 kV, magnification 14× up to 1,000,000 and resolution for Gun, FEI company, Netherlands.

- The disc diffusion method [29, 30] was used for assessing the antibacterial activity of gelatin powder, untreated and treated cotton fabrics. Briefly, discs of 10 mm diameter were cut from the cotton fabrics. Nutrient agar plates were incubated with microbial culture. The cut discs of untreated and treated cotton fabrics were placed onto the surface of inoculated plates. The plates were incubated at 37°C for 48 hours. The inhibition zone (distance from disc circumference in mm) was determined for each disc.

Assay for cytotoxicity test of gelatin (In vitro)

Cell culture

Culture was maintained in Dulbecco's Modified Eagle's medium (DMEM) medium (in case of A549), and supplemented with 10% foetal bovine serum at 37 °C in 5 %CO₂ and 95% humidity, cells were sub-cultured using trypsin versene 0.15 %. Notable, skin normal human cell line (BJ-1) "Immortalized normal foreskin fibroblast cell line" was obtained from Karolinska Center, Department of Oncology and Pathology, Karolinska Institute and Hospital, Stockholm, Sweden. Other cell lines "were obtained from Vacsera (Giza, Egypt).

Cell viability assay

After about 24 h of seeding 20000 cells per well in case of A-549 cells per well (in 96 well plates), the medium was changed to serum-free medium containing a final concentration of the extracts of 100 µg/ml in triplicates. The cells were treated for 24 hours 100 µg/ml doxorubicin was used as positive control and 0.5 % distilled water was used as negative control. Cell viability was determined using the MTT (3-(4, 5-dimethylthiazol-2-yl)-2, 5-diphenyltetrazolium bromide) assay as described by Mosmann 1983 with minor modifications [31].

$$(1 - (Av(x) / (Av(NC)))) \times 100$$

Where Av: average, X: absorbance of sample well measured at 595 nm with reference 690 nm, NC: absorbance of negative control measured at 595 nm with reference 690.

Results and Discussion

Treatment of cotton fabrics with gelatin

Cotton fabrics were reacted with gelatin in the presence of dimethyloldihydroxyethylene urea (DMDHEU) as binder and dihydrogen phosphate as catalyst to impart it new desirable properties such as antibacterial, mechanical and thermal properties. Herein we study the reaction parameters to optimize it, these reaction parameters are: concentrations of ammonium dihydrogen phosphate (0.5-3.0 g/L), gelatin (3-10 w/w %), DMDHEU (0-80 g/L), curing time (1-5 min.) and temperature (120-160 °C). Scheme 1 shows the extent of reaction between cotton fabrics and gelatin in the presence of ammonium dihydrogen phosphate as catalyst at different curing time and temperature.

As we see in Scheme 1 that gelatin reacts with cellulose via nucleophilic substitution reaction in slow step and all other steps are fast steps which mean that equation 2 is the rate determining step. So that we can follow the reaction progress quantitatively via estimation of nitrogen content which reflect amount of gelatin modified cellulose.

Effect of ammonium dihydrogen phosphate concentration

Figure 2 shows the dependence of the extent of the reaction, expressed as nitrogen content, between cotton fabric and gelatin (7% w/v) in the presence of DMDHEU concentrations (60 g/L) and ammonium dihydrogen phosphate as a catalyst at different concentrations (0.5-3.0 g/L); dry at 80°C for 5 min and cure at 160 °C for 3 min.



Scheme 1. Suggested equations for the reaction of gelatin with cotton fabrics in the presence of catalyst (DMDHEU)

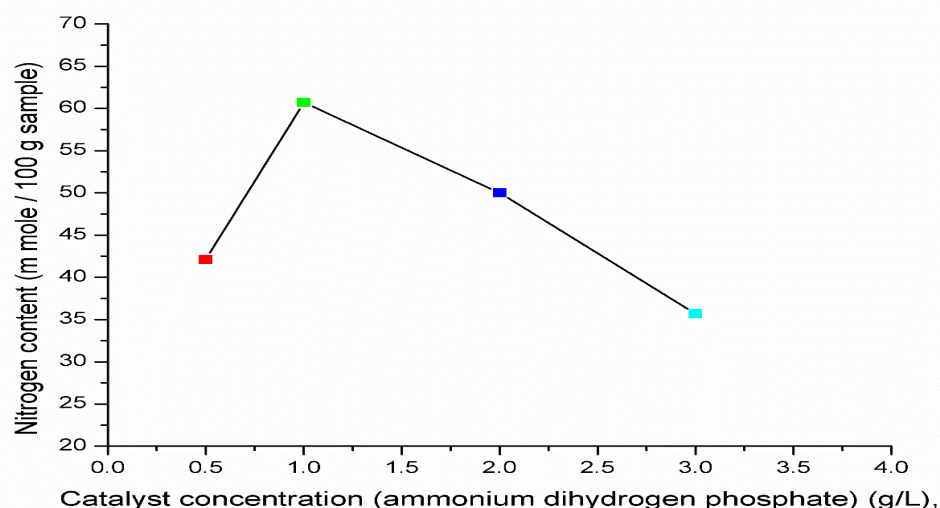


Fig. 2. Effect of ammonium dihydrogen phosphate concentrations

Reaction condition: Gelatin concentration (w/w) 7%; DMDHEU concentrations 60 g/L; dry at 80°C for 5 min and cure at 160°C for 3 min.

Figure 2 shows that increasing of the ammonium dihydrogen phosphate concentration up to 1 g/L, causes a significant enhancement in the extent of the reaction (equations 1 and 2).

On increasing the ammonium dihydrogen phosphate concentration, the liberated H_3PO_4 acid increase which acts as a self-catalyst and increase the reaction extent. Whereas a further increase in the ammonium dihydrogen phosphate concentration accompanied by decrease in the reaction extent which may be due to the increase the concentration of the liberated acid which can form side reaction products as shown in equations 3-5 [32].

Effect of gelatin concentrations

The effect of gelatin concentration was obtained by treating cotton fabric with different concentrations of gelatin (3-10 % w/v) in the presence of DMDHEU concentrations 60 g/L and ammonium dihydrogen phosphate 1(g/L); dry at 80°C for 5 min and cure at 160°C for 3 min.

Figure 3 illustrates the effect of gelatin concentration where increasing the gelatin concentrations from 3% up to 10% was accompanied by increasing the nitrogen content. This indicates that, the reaction between gelatin and treated fabric increased as shown in equation 2. Higher concentrations of gelatin more than 10 % cause higher rigidity of the fabric, which is not acceptable. So that we can't use concentration higher than 10%.

Effect of dimethyloldihydroxyethylene urea (DMDHEU) concentrations

DMDHEU is more attractive in textile finishing due to its durability and low cost. The effect of DMDHEU concentration was obtained by treating cotton fabric with different concentrations (0-80 g/L) in the presence of gelatin concentration 7% and ammonium dihydrogen phosphate 1(g/L), dry at 80°C for 5 min and cure at 160°C for 3 min. Figure 4 shows that increasing of the dimethyloldihydroxyethylene urea (DMDHEU) concentrations from 0 up to 60% was accompanied by increasing the nitrogen content which is due to the cross-linking with cellulose. This is the result of conversion of some hydrogen bonds to covalent bonds [33]. This indicates that the reaction between gelatin and fabric increase as shown in equation 6 (Scheme 2). There is a slight increasing of the nitrogen content by increasing the dimethyloldihydroxyethylene urea (DMDHEU) concentrations to 80 %.

Effect of the curing temperature and time

Figure 5 shows the effect of temperature and time on the extent of the reaction occur between cotton fabric and gelatin (7%) in presence of DMDHEU concentrations (60g/L) and ammonium dihydrogen phosphate (1g/L) at different curing temperatures (120–160 °C) and for different curing of times (1 –5 min).

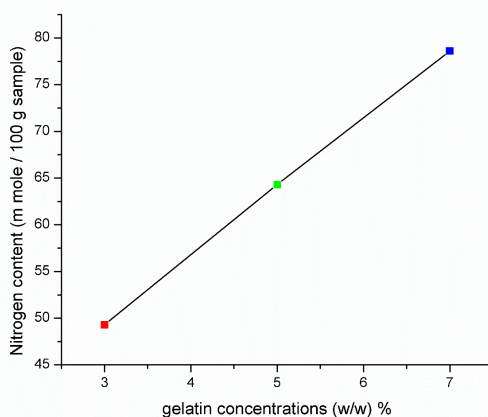
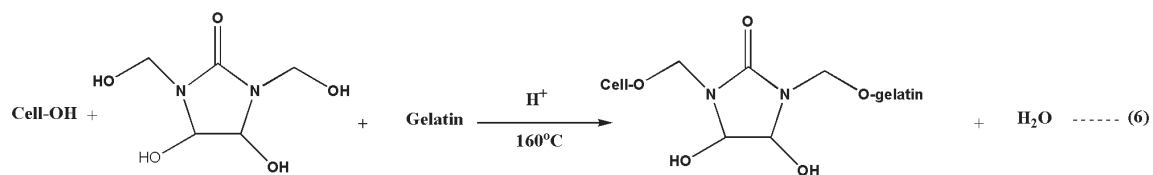


Fig. 3. Effect of gelatin concentrations

Reaction condition: concentration of ammonium dihydrogen phosphate 1 g/L, DMDHEU concentrations 60g/L dry at 80°C for 5 min and cure at 160°C for 3 min.



Scheme 2. reaction of cellulose with gelatin via dimethyldihydroxyethylene urea (DMDHEU)

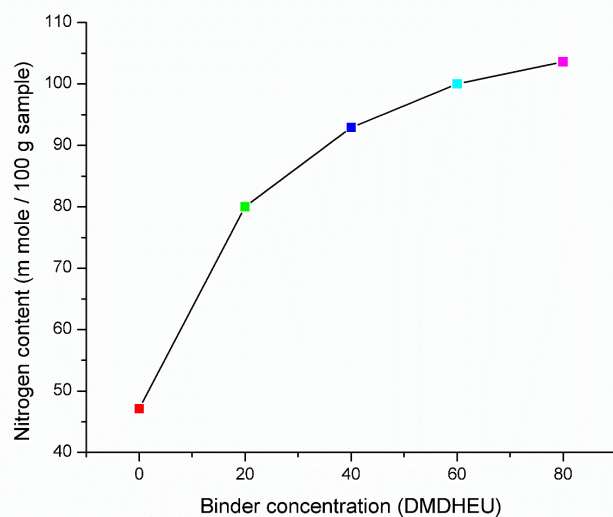


Fig. 4. Effect of binder DMDHEU concentrations

Reaction condition: concentration of ammonium dihydrogen phosphate; 1 g/L, Gelatin concentration (w/v) 7%; dry at 80°C for 5 min and cure at 160°C for 3 min.

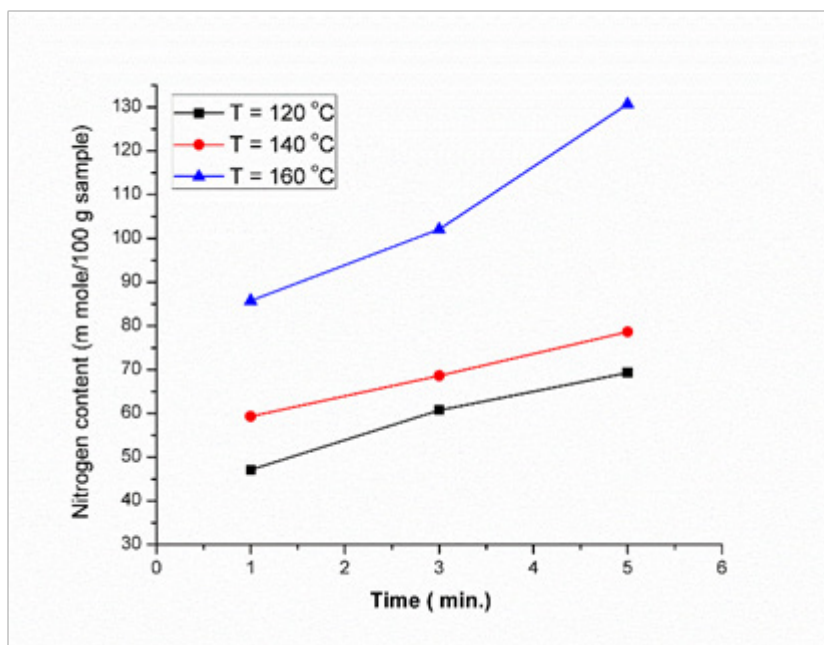


Fig. 5. Effect of the curing temperature and time

Reaction conditions: Ammonium dihydrogen phosphate (1 g/L), Gelatin concentration (w/v) (7%); DMDHEU concentrations 60 g/L and dry at 80°C for 5 min

A close examination of Fig. 5 would reveal the following: The nitrogen content increases by increasing the curing temperature and time and the maximum value of the nitrogen content is obtained at 5 min with respect to all samples at different temperatures specially at 160°C.

Characterization of gelatin treated cotton fabrics

FT-IR spectra were obtained from gelatin powder, untreated cotton fabric and cotton fabric treated with gelatin. As shown in Fig. 6. The FT-IR spectra show typical bands of gelatin (Fig. 6.a) where it formed by four individual peaks at amide-A and free water at 3282 cm^{-1} , amide-I at 1630 cm^{-1} , amide-II at 1547 cm^{-1} , amide-III at 1239 cm^{-1} [34, 35]. Figure 6.b shows the cotton fabrics' typical characteristic peaks which showed typical ones as for pure cellulose. Broad C-H stretching band appears from 2800 to 3000 cm^{-1} region. Peaks at 2918 and 2849 cm^{-1} corresponding to the asymmetric and the symmetric stretching of methylene ($-\text{CH}_2-$) groups in long alkyl chains. It can be found that the two new absorption peaks appear at 1694 and 1760 cm^{-1} respectively, assigned to CO absorption peaks of hydanto in ring structure. Other information can be obtained in the two peaks at 1600-1400 cm^{-1} for the asymmetric and the symmetric stretching of COO^- ion, respectively [21, 36, 37]. The FT-IR spectra of gelatin treated cotton fabrics show appearance of all characteristic bands of both cellulose and gelatin with slight shifts of these peaks which proof that gelatin bonded with cotton fabrics [38].

Table 2 shows the extents of some physico-mechanical properties of the treated cotton fabric with gelatin. It is obvious that the treated cotton fabric has an increasing in fabric stiffness (S), surface roughness (SR), tensile strength (TS), and elongation at break (EB) but there is a decrease in air permeability (AP). Gelatin ingredient onto the fabric structure leads to variation in extents of some properties [39-41].

Table 2 shows that TS increased from 29 to 33 Kg/m^2 due to the interaction between gelatin and cotton fabrics and entanglement of gelatin through the fabrics at the same time the EB decreased from 25 to 21% due to the crosslinking between the gelatin and cellulose biopolymers [42-44]

Also, air permeability measured for both untreated cotton fabric and treated one with gelatin and its value decreased from 16.28 to 8.30 ($\text{cm}^3/\text{cm}^2.\text{s}$) due to induction of compact and condense protein networks by crosslinking [44]

Thermal stability of the untreated and treated fabric with gelatin evaluated based on thermo gravimetric analysis (TGA). Figure 7 shows the TGA evaluations expressed on weight loss % with temperature for untreated and treated fabric with gelatin which show similar behavior with four main thermal degradation stages. The first stage of low M. Wt compounds such as water at 40 - 130 °C [45]. The second stage for glycerol and structurally H_2O is bounded at 170 to 280 °C [46]. For the third stage, cellulose and gelatin chains has been last at 280-380 °C. The fourth stage of more thermal stability structure at temperature is higher than 500 °C [47]. Figure 7 shows that the untreated fabric shows decompositions at temperatures 298, 369 and 528 °C whereas the treated one with gelatin shows the corresponding decompositions at 298, 391 and 605 °C, so that the treated cotton fabrics with gelatin has a higher thermal stability than an untreated one [44].

a

Figure 8 shows the scanning electron microscope (SEM) of untreated cotton fabrics and gelatin treated cotton fabrics (Fig. 8 (a and b) respectively); the cotton fabrics were treated with gelatin via chemical crosslinking reaction. Figure 8 shows the SEM of cotton fabrics before and after being treated with gelatin. Where it can be clearly seen that gelatin deposited homogeneous on the fiber surface and no aggregation formed. In addition, it confirms the fixation of gelatin onto the fiber surface because of etherification reaction.

Antibacterial activity of gelatin powder and fabric treated with gelatin

The antibacterial activity data of gelatin powder compared with antibiotic ciprofloxacin were summarized in Table 3. These data estimated invitro via disk inhibition zone method. *S. aureus* and *E. coli* were used as an example of Gram-positive and Gram negative bacteria. The antibacterial data confirm that gelatin itself has excellent antibacterial activity towards tested bacteria.

Figure 9 shows the antibacterial activity of gelatin treated cotton fabrics, evaluated invitro via disk diffusion method against *S. aureus* as Gram-positive bacteria and *E. coli* as Gram-negative bacteria.

The results show that gelatin treated samples shows antibacterial activity with concentrations 3% and 5% of gelatin in the fabrics as shown in Fig. 9.

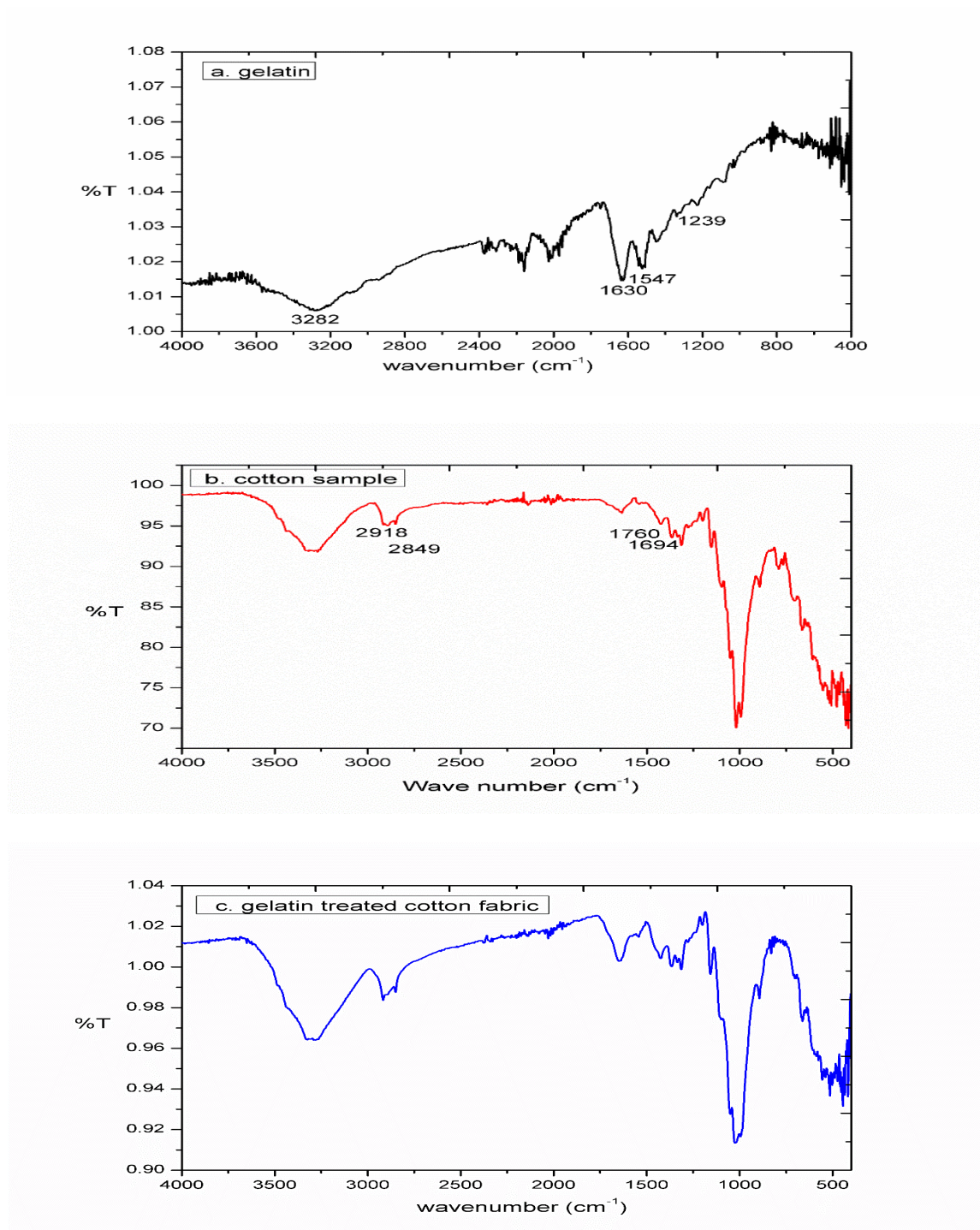


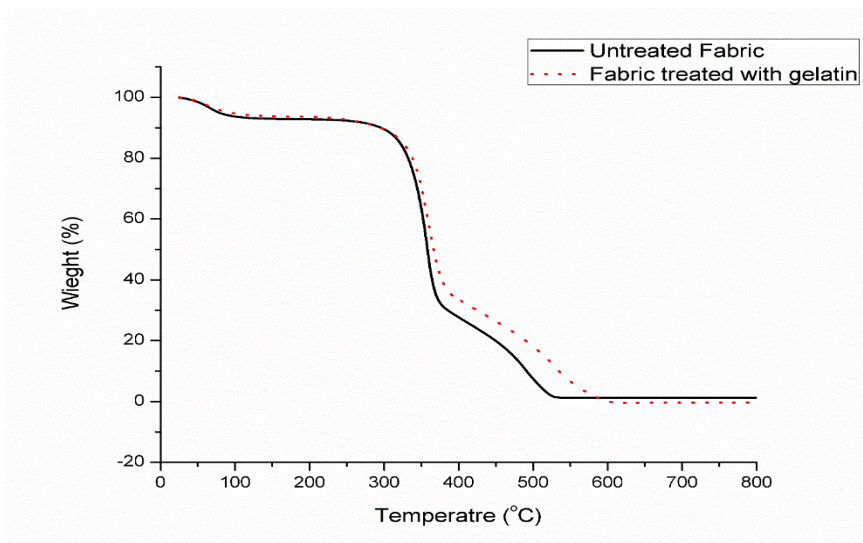
Fig. 6. FTIR chart of cotton, gelatin and gelatin treated cotton fabric samples

Reaction conditions: Ammonium dihydrogen phosphate (1 g/L), Gelatin concentration (w/v) (7%); DMDHEU concentrations 60g/L and dry at 80°C for 5 min and cure at 160 °C for 3 min.

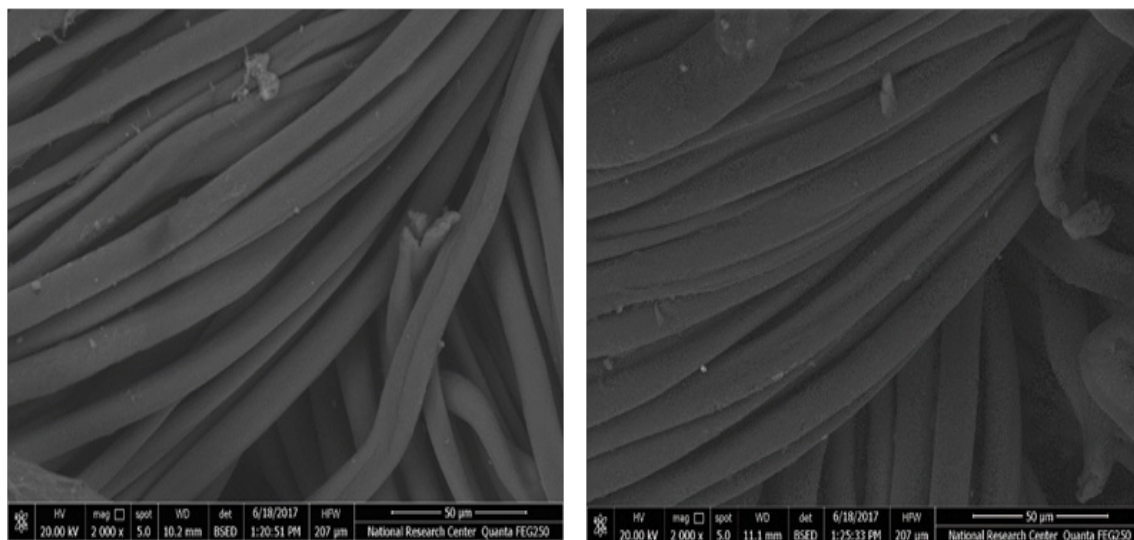
TABLE 2. Effect of gelatin treatment on some performance functional properties of treated fabric

Test	Untreated Fabric	Fabric Treated with Gelatin
Stiffness ; S (mg)	1103.6	1708.8
Surface Roughness; SR (μm)	15.75	16.39
Air Permeability; AP ($\text{cm}^3/\text{cm}^2.\text{s}$)	16.28	8.30
Tensile Strength; TS (Kg/m^2)	29	33
Elongation at Break; EB (%)	25	21

Reaction conditions: Ammonium dihydrogen phosphate (1 g/L), Gelatin concentration (w/v) (7%); DMDHEU concentrations 60 g/L and dry at 80°C for 5 min and cure at 160 °C for 3 min.

**Fig.7. Thermal Gravimetric Analysis of untreated and treated cotton fabric with gelatin**

Reaction conditions: Ammonium dihydrogen phosphate (1 g/L), Gelatin concentration (w/v) (7%); DMDHEU concentrations 60 g/L and dry at 80°C for 5 min and cure at 160 °C for 3 min.

**Fig. 8. SEM of (a) untreated cotton fabrics, (b) gelatin treated cotton fabrics**

Reaction conditions: Ammonium dihydrogen phosphate (1 g/L), Gelatin concentration (w/v) (7%); DMDHEU concentrations 60 g/L and dry at 80 °C for 5 min and cure at 160°C for 3 min.

TABLE 3. Antibacterial activity of gelatin compared with well known antibiotic ciprofloxacin via disk inhibition zone (all samples evaluated at 20 µg/ml concentration)

Sample	Inhibition zone, mm	
	<i>S. aureus</i>	<i>E. coli</i>
gelatin	23.0	20.5
ciprofloxacin	24.0	22.5

In addition, the concentration of ammonium dihydrogen phosphate has no effect on antibacterial activity of the gelatin treated cotton fabrics. The antibacterial activity of these samples is due to the presence of amino, amide and carboxylic groups in the gelatin structure, which adsorb onto bacterial surface, to penetrate cell membrane, finally destruct cell membrane causing bacteria death. Also these samples showed higher antibacterial activity toward Gram-positive bacteria (*S. aureus*) more than Gram-negative bacteria (*E. coli*) due to their cell wall structure [48, 49].

Effect of durability of gelatin treated cotton fabrics towards washing cycles on antibacterial activity

Figure 10 illustrates the effect of durability of gelatin treated cotton fabric towards washing cycles (from 5 to 25 washing cycle). These durability depends on some types of bonding of gelatin with cotton fabrics.

Figure 10 shows that the treated fabrics were durable towards washing cycles up to 25 cycles; where it has antibacterial activity even after 25 washing cycle. In addition, this durability decreases as washing cycles increases. From the data we can say that the treated cotton fabrics were effective even until 25 cycles and more.

Cytotoxicity of gelatin compared with ciprofloxacin via MTT assay

The main aim of this study that use of safe gelatin as antibacterial agent for cotton fabrics. Herein the cytotoxicity of gelatin was evaluated via MTT assay compared with common use antibiotic ciprofloxacin to illustrate and confirm the safety of natural polymer gelation. Various concentrations of both gelatin and ciprofloxacin ranged from 10-300 mg/l were used for the cultured cells incubation for to determine their cytotoxicity by MTT assay that used for viable A549 cells evaluation and expressed in mitochondrial activity decrement as shown in Fig. 11.

Figure 11 shows the MTT protocol to evaluate the cytotoxic effects of gelatin compared with common antibiotic ciprofloxacin. At low concentration (from 10-50 mg/l) there is weak toxic effect of ciprofloxacin and there is no effect for gelatin. At 100 mg/l concentration the viable cells numbers decreased to it half of initial values. As the concentration of 200 mg/l there are about 30% only viable cells, at the higher concentration of 300 mg/l the viable cells represents only less than 20% of the original one. Where are the is change in viable cells for gelatin as the concentration increased from 10-30 mg/l (Fig. 11).

Conclusion

Gelatin was used as antibacterial material for cotton fabrics via pad-dry-cure method. The optimum condition for fabric treatment were gelatin concentration at 7%, DMDHEU concentration at 60 g/l and ammonium dihydrogen phosphate at 1 g/l which was dried at 80 °C for 5 mints and cured at 160 °C for 3 min. FT-IR spectroscopy confirm that gelatin bonded with cotton fabrics. At the optimum reaction condition, there are an increasing in fabric stiffness, surface roughness, tensile strength, elongation and thermal stability but there is a decrease in air permeability. SEM confirm the fixation of gelatin onto the fiber surface because of etherification reaction. Gelatin shown higher antibacterial activity comparable with ciprofloxacin and can be used as safe powerful natural antibacterial activity for cotton fabrics. The treated cotton fabric showed higher antibacterial activity toward Gram-positive bacteria more than Gram-negative bacteria. In addition we found that these treated fabrics were durable upto 25 washing cycles. There is no change in viable cells in case of natural polymer gelatin compared with common used antibiotic ciprofloxacin which enriched its usage as antibacterial finished material.

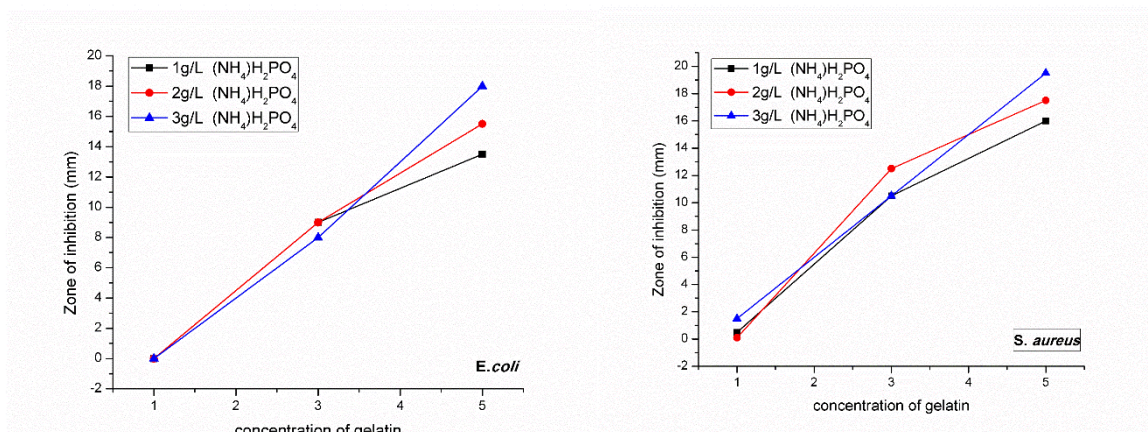


Fig. 9. Antibacterial Activity of gelatin treated cotton fabrics towards *S. aureus* and *E. coli* at different experimental conditions

Reaction conditions: Ammonium dihydrogen phosphate (1 g/L), Gelatin concentration (w/v) (7%); DMDHEU concentrations 60 g/L and dry at 80°C for 5 min and cure at 160°C for 3 min.

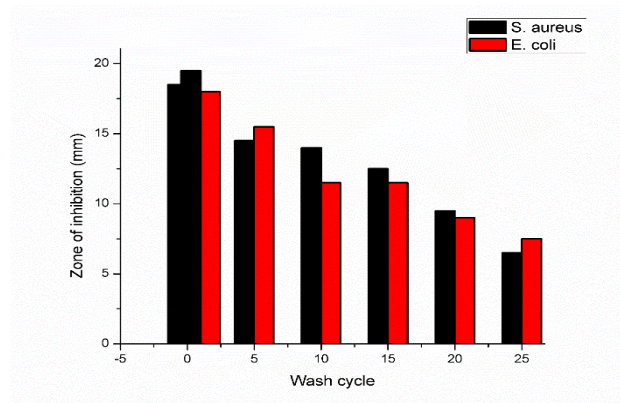


Fig. 10. Effect of durability of gelatin treated cotton fabrics towards washing cycles on antibacterial activity

Reaction conditions: Ammonium dihydrogen phosphate (1 g/L), Gelatin concentration (w/v) (7%); DMDHEU concentrations 60 g/L and dry at 80°C for 5 min and cure at 160°C for 3 min.

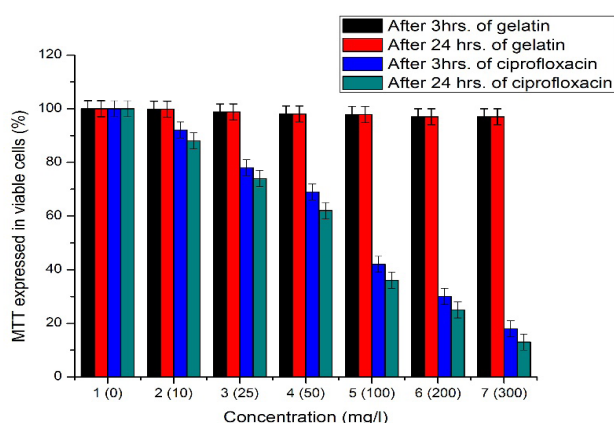


Fig. 11. Mitochondrial metabolic activity (MTT) assay expressed in viable cells of gelatin and ciprofloxacin after 3 and 24 hr, cell culture

Reaction conditions: Ammonium dihydrogen phosphate (1 g/L), Gelatin concentration (w/v) (7%); DMDHEU concentrations 60 g/L and dry at 80°C for 5 min and cure at 160°C for 3 min.

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References

1. Ibrahim, H.M., et al., Preparation of chitosan antioxidant nanoparticles as drug delivery system for enhancing of anti-cancer drug, in *Key Engineering Materials*. 2018. p. 92-97.
2. El-Bisi, M.K., et al., Super hydrophobic cotton fabrics via green techniques. *Der Pharma Chemica*, 2016. **8**(19): p. 57-69.
3. Farag, S., et al., Impregnation of silver nanoparticles into bacterial cellulose: Green synthesis and cytotoxicity. *International Journal of ChemTech Research*, 2015. **8** (12): p. 651-661.
4. Ibrahim, N.A., et al., Enhanced antibacterial properties of polyester and polyacrylonitrile fabrics using Ag-Np dispersion/microwave treatment. *AATCC Journal of Research*, 2014. **1** (2): p. 13-19.
5. Liu, J., et al., Study on the grafting of chitosan–gelatin microcapsules onto cotton fabrics and its antibacterial effect. *Colloids and Surfaces B: Biointerfaces*, 2013. **109**: p. 103-108.
6. Ibrahim, H.M., et al., Preparation of Cotton Gauze Coated with Carboxymethyl Chitosan and its Utilization for Water Filtration. 2019. **11** (1).
7. Aysha, T., et al., Synthesis, spectral study and application of solid state fluorescent reactive disperse dyes and their antibacterial activity. *Arabian Journal of Chemistry*, 2019. **12** (2): p. 225-235.
8. Fouda, M.M. and H. Fahmy, *Multifunctional finish and cotton cellulose fabric*. *Carbohydrate polymers*, 2011. **86** (2): p. 625-629.
9. Mohamed, F.A., et al., Synthesis, application and antibacterial activity of new reactive dyes based on thiazole moiety. 2018. **47** (3): p. 246-254.
10. Paul, R., *Functional Finishes for Textiles: Improving Comfort, Performance and Protection*. 2014: Elsevier.
11. Mohamed, F.A., et al., Improvement of dyeability and antibacterial properties of gelatin treated cotton fabrics with synthesized reactive dye. 2018. **15** (4): p. 4403-4408.
12. Hashem, M., et al., Improving easy care properties of cotton fabric via dual effect of ester and ionic crosslinking. *Carbohydrate Polymers*, 2011. **86** (4): p. 1692-1698.
13. Mohamed, F.A., H.M. Ibrahim, and M.M. Reda, *Eco friendly dyeing of wool and cotton fabrics with reactive dyes (bifunctional) and its antibacterial activity*. *Der Pharma Chemica*, 2016. **8** (16): p. 159-167.
14. Ibrahim, H.M., M.M. Saad, and N.M. Aly, Preparation of single layer nonwoven fabric treated with chitosan nanoparticles and its utilization in gas filtration. *International Journal of ChemTech Research*, 2016. **9** (6): p. 1-16.
15. Gómez-Estaca, J., et al., Antimicrobial activity of composite edible films based on fish gelatin and chitosan incorporated with clove essential oil. *Journal of Aquatic Food Product Technology*, 2009. **18** (1-2): p. 46-52.
16. Mosaad, R.M., A. Samir, and H.M. Ibrahim, Median lethal dose (LD50) and cytotoxicity of Adriamycin in female albino mice. *Journal of Applied Pharmaceutical Science*, 2017. **7** (3): p. 77-80.
17. Nawalakhe, R.G., et al., Development of electrospun iminochitosan for improved wound healing application. *Journal of Engineered Fibers and Fabrics*, 2012. **7** (2): p. 47-55.
18. Ninan, N., et al., Antibacterial and wound healing analysis of gelatin/zeolite scaffolds. *Colloids and Surfaces B: Biointerfaces*, 2014. **115**: p. 244-252.
19. Yang, H. and W.J. Kao, Thermoresponsive gelatin/monomethoxy poly (ethylene glycol)–poly (D, L-lactide) hydrogels: formulation, characterization, and antibacterial drug delivery. *Pharmaceutical Research*, 2006. **23** (1): p. 205-214.
20. Hebeish, A., et al., Highly effective antibacterial textiles containing green synthesized silver nanoparticles. *Carbohydrate Polymers*, 2011. **86** (2): p. 936-940.
21. Yin, R., et al., Preparation and characterization of novel gelatin/cerium (III) fiber with antibacterial activity. *Materials Letters*, 2009. **63** (15): p. 1335-1337.

22. Shankar, S., et al., Preparation, characterization, and antimicrobial activity of gelatin/ZnO nanocomposite films. *Food Hydrocolloids*, 2015. **45**: p. 264-271.
23. Abdel-Halim, E., et al., Incorporation of chlorohexidin diacetate into cotton fabrics grafted with glycidyl methacrylate and cyclodextrin. *Carbohydrate Polymers*, 2010. **79** (1): p. 47-53.
24. Lam, P.-L., et al., Non-toxic agarose/gelatin-based microencapsulation system containing gallic acid for antifungal application. *International journal of Molecular Medicine*, 2015. **35** (2): p. 503-510.
25. Cheng, S.Y., et al., Cosmetic textiles with biological benefits: gelatin microcapsules containing vitamin C. *International Journal of Molecular Medicine*, 2009. **24** (4): p. 411-419.
26. Farag, S., et al., Comparative study for bacterial cellulose production Using Egyptian *Achromobacter* sp. *Research Journal of Pharmaceutical, Biological and Chemical Sciences*, 2016. **7** (6): p. 954-969.
27. Ibrahim, H.M. and E.M.R. El-Zairy, Carboxymethylchitosan nanofibers containing silver nanoparticles: Preparation, Characterization and Antibacterial activity. *Journal of Applied Pharmaceutical Science*, 2016. **6** (7): p. 43-48.
28. Hanani, Z.N., Y. Roos, and J. Kerry, Use and application of gelatin as potential biodegradable packaging materials for food products. *International Journal of Biological Macromolecules*, 2014. **71**: p. 94-102.
29. Ibrahim, H., et al., *Carboxymethyl Chitosan Electrospun Nanofibers: Preparation and its Antibacterial Activity*. *Journal of Textile & Apparel Technology & Management (JTATM)*, 2015. **9** (2).
30. Mohamed, F.A., et al., Improving dye ability and antimicrobial properties of cotton fabric. *Journal of Applied Pharmaceutical Science*, 2016. **6** (2): p. 119-123.
31. Maneerung, T., S. Tokura, and R.J.C.p. Rujiravanit, Impregnation of silver nanoparticles into bacterial cellulose for antimicrobial wound dressing. 2008. **72**(1): p. 43-51.
32. Khalil, M., et al., Some studies on starch–urea–acid reaction mechanism. *Carbohydrate Polymers*, 2002. **48** (3): p. 255-261.
33. Mortazavi, S.M. and P. Esmailzadeh Boukany, *Egypt. J. Chem.* **62**, No. 9 (2019)
- Application of mixtures of resin finishing to achieve some physical properties on interlining cotton fabrics: I-effect of stiffening and cross-linking agents. *IRANIAN POLYMER JOURNAL.*, 2004. **13**: p. 213-218.
34. Muyonga, J., C. Cole, and K. Duodu, Fourier transform infrared (FTIR) spectroscopic study of acid soluble collagen and gelatin from skins and bones of young and adult Nile perch (*Lates niloticus*). *Food Chemistry*, 2004. **86** (3): p. 325-332.
35. Hanani, Z.N., Y. Roos, and J. Kerry. Fourier Transform Infrared (FTIR) spectroscopic analysis of biodegradable gelatin films immersed in water. In *11th International Congress on Engineering and Food. Congress conducted at Athens, Greece*. 2011.
36. Chung, C., M. Lee, and E.K. Choe, Characterization of cotton fabric scouring by FT-IR ATR spectroscopy. *Carbohydrate Polymers*, 2004. **58** (4): p. 417-420.
37. Wang, Q., et al., Characterization of bioscoured cotton fabrics using FT-IR ATR spectroscopy and microscopy techniques. *Carbohydrate Research*, 2006. **341**(12): p. 2170-2175.
38. Cheng, X., et al., Antimicrobial coating of modified chitosan onto cotton fabrics. *Applied Surface Science*, 2014. **309**: p. 138-143.
39. Fahmy, H., A. Aly, and Z. Mohamed, Synthesis of poly (N-vinyl-2-pyrrolidone)/pyrodextrins adducts and their utilization in functionalization of cotton fabric. *International Journal of Chem Tech Research*, 2016. **9** (9): p. 96-109.
40. Fahmy, H., A. Aly, and S.M. Sayed, Graft copolymerization of N-vinylpyrrolidone onto stearyl alcohol to impart water repellency and antibacterial properties for cotton/polyester fabric. *Progress in Organic Coatings*, 2017. **105**: p. 176-182.
41. Fahmy, H., et al., Evaluation of Functional and Comfort Properties of SA/TDI/PEG1000 Adduct Treated Cotton/Polyester Blended Fabric.
42. Mu, C., et al., Preparation and properties of dialdehyde carboxymethyl cellulose crosslinked gelatin edible films. *Food Hydrocolloids*, 2012. **27**(1): p. 22-29.
43. Hosseini, S.F., et al., Preparation and functional properties of fish gelatin–chitosan blend edible films. *Food Chemistry*, 2013. **136** (3): p. 1490-1495.
44. Guo, J., et al., Periodate oxidation of xanthan gum

and its crosslinking effects on gelatin-based edible films. *Food Hydrocolloids*, 2014. **39**: p. 243-250.

45. Mu, C., et al., Freezing/thawing effects on the exfoliation of montmorillonite in gelatin-based bionanocomposite. *Journal of Applied Polymer Science*, 2013. **128** (5): p. 3141-3148.

46. Soares, R., et al., Thermal degradation of biodegradable edible films based on xanthan and starches from different sources. *Polymer Degradation and Stability*, 2005. **90** (3): p. 449-454.

47. Guo, J., et al., Freezing–thawing effects on the properties of dialdehyde carboxymethyl cellulose crosslinked gelatin-MMT composite films. *Food Hydrocolloids*, 2013. **33** (2): p. 273-279.

48. Liu, S., J. Ma, and D. Zhao, *Synthesis and characterization of cationic monoazo dyes incorporating quaternary ammonium salts*. *Dyes and Pigments*, 2007. **75** (2): p. 255-262.

49. Yazdimamaghani, M., et al., Green synthesis of a new gelatin-based antimicrobial scaffold for tissue engineering. *Materials Science and Engineering: C*, 2014. **39**: p. 235-244.