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Antibacterial Activity Evaluation of a Treated Cotton by Chitosan Polymer

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***Abstract*— In this study, the synthesis and the application of chitosan hydrogel was explored in an activated textile fabric. The chemical modifications were based on the anionic and the cationic activation of cotton. The pH stimuli-responsiveness behavior of the synthetized polymer was also investigated and then confirmed in terms of regulating water uptake of modified cotton in function of pH. The Surface morphology of treated cotton fabric was studied by SEM analysis and showed that the surface morphology of the modified cotton is slightly depending to the chemical modification process: when carboxyl groups (carboxymethylation) or amine groups (aminisation) were added. Further, the antibacterial activity of the hydrogel-coated samples has been evaluated against Escherichi coli, Listeria monocytogene and Staphylococcus aureus bacteria. Results showed a clear improvement of the antibacterial property against both Gram-positive and Gram-negative bacteria.**

Keywords*— *Chitosan; hydrogel; activated cotton; pH stimuli-responsive; antibacterial activity.

I. INTRODUCTION

Man has adopted antimicrobial substances since ancient times, a fact that is demonstrated by their use in Egyptian mummies and in similar applications in other cultures [1]. In this regard, the protection of textile fabrics, too, have long fulfilled a role of the utmost importance. Today, this need to protect and preserve can be fundamental for some textile application such as for biomedical field. Antimicrobials are protective agents that, being bacteriostatic, bactericidal, fungistatic and fungicidal, also offer special protection against the various forms of textile rotting. Synthetic fibres have almost good antibacterial capacity but they are not completely immune to microorganisms. The problem is more complicated with natural fibres such as cotton or wool, although having different behaviour. Wool is more likely to suffer bacterial attack than cotton, and cotton is more likely than wool to be attacked by fungi. In addition, substances added to fibres, such as lubricants, antistatics, natural-based auxiliaries (for example size, thickener and hand modifiers) and dyes may provide a good food source for bacteria [2]. Subsequently, the textile support must be well protected.

There are various chemical and physical possibilities that can be considered in the production of antimicrobial fabrics. In practice, the antimicrobial effect is obtained through the application of specific chemical products during the finishing stage, or through the incorporation of these substances into chemical fibres during the spinning process. The most important antibacterial agents that are used in textile finishing still quaternary ammonium compounds. Triclosan which is halogen-containing derivative of phenol offers also good antibacterial protection. Some metallic salts, such as copper, zinc and cobalt, have attracted attention as effective antimicrobial agents for textiles [3, 4]. For this example, silver is generally the most widely used in textiles. The application of each cited antibacterial substance makes the treated textile support more pollutant and not compatible with further applications especially in the biomedical filed.

Ecofriendly polysaccharides polymers are the main part of the natural-based hydrogels that have exceptional properties such as biocompatibility, biodegradability, renewability, and non-toxicity [5, 6]. Furthermore, this biopolymer attracts currently a great deal of interest for antibacterial activity mixed by its biodegradability [7, 8].

In this study, the application of the synthetized hydrogel polymer onto activated cotton was elucidated. The pH stimuli-responsiveness behavior of obtained hydrogels was tested and discussed in term of water uptake property. Assessment of the antibacterial activity of cotton fabrics was evaluated in order to confirm the antibacterial capacity of the modified cotton.

II. EXPERIMENTAL

A. *Materials*

Chitosan polymer was supplied by Sigma and used as received. Chemicals employed for activation of bleached cotton supports were sodium hydrosulfite ($Na_2S_2O_4$) monochloracetic acid (CAA), sodium carbonate (Na_2CO_3), caustic soda (NaOH , 38°Be), hydrochloric acid (HCl). They were supplied by Chemi-pharma and were of analytical grade.

B. *Synthesis of hydrogels procedure*

1.0 g of chitosan was dissolved in distilled water containing 2% (o.w.f) of acetic acid. Next, the solution was suspended with Blender. Then 10.0% (w/v) of sodium hydroxide was added to the chitosan solution and the resulting mixture was stirred until pH of the solution reached to 10. The resulting hydrogel was after that filtered, dialyzed against deionized water until the outer solution was neutralized. The hydrogel was finally dried at 25°C for 24 h.

C. Cotton surface activation procedure

Ionic character conferred to cotton cellulose facilitate the incorporation of chitosan hydrogel. The anionic cotton consisted of reaction with monochloracetic acid to give partially carboxymethylated cellulose. This process is presented by Equation 1. Cationic cotton was obtained by a reactive dye followed by a reductive cleavage of the fixed dye. The process is described by Equation 2.

$$Cellulose-OH\ NaOH+ClCH_2COONa \rightarrow Cellulose-O-CH_2COONa+NaCl+H_2O \quad (1)$$

$$Cellulose-OH\ NaOH+R-NR'^{3+} \rightarrow Cellulose-O-R-NR'^{3+}H_2O \quad (2)$$

D. Chitosan hydrogel application process

Chitosan was applied onto samples by impregnation in acidic chitosan solution (pH adjusted to 3.6) during 2 hours, and following this by a gelation reaction with the cross-linking with the cotton support at pH 10 at room temperature.

E. SEM Characterization

The surface morphology of treated cotton by chitosan hydrogel was examined using the Scanning Electron Microscope (SEM) operating at 5 kV, and compared to the surface morphology of non modified cotton.

F. pH stimuli-responsiveness measurements

The evaluation of the hydrogel responsiveness to pH changes (swelling/deswelling) was investigated by means of the gravimetric measurements. The samples were immersed into solutions of different pH values (4, 6, 7 and 9) and by determining the water uptake (%) at room temperature with varying immersion duration The water uptake WU(t)(%) was calculated from the Equation 3:

$$WU(t)(\%)=\frac{WUt-WU0}{WU0} \quad (3)$$

Where WU0 is the weight of the sample before immersion in the solution and WUt is the weight of the sample after the immersion.

G. Antibacterial Activity Evaluation

Antibacterial activity experiments were based on the intended application, the environment in which the textile product is to be used and on the surface properties of the textile properties. The antibacterial properties of functionalized fabrics were evaluated according to the disc diffusion method ASTM E2149-01. Discs of 6 mm diameter were cut from the tested fabric. Nutrient agar plates were incubated with microbial culture. The cut discs of fabric were placed onto the surface of inoculated plates, which were then incubated at 37°C for 48 hours. The inhibition zone (distance from disc in mm) was determined for each disc. Three bacterial cultures were used for assessing the antibacterial activity of prepared samples (Escherichi coli ATCC 35218, Listeria monocytogene ATCC 19115 and Staphylococcus aureus ATCC 25923). These microorganisms were obtained from the culture collection of the Laboratory of Analysis, Treatment and Valorization of Pollutants of the Environment and Products, Faculty of Pharmacy of Monastir.

III. RESULTS AND DISCUSSIONS

A. Stimuli-responsiveness of chitosan hydrogel coated cotton

In this section, the stimuli-responsivness was investigated by controlling the water uptake as function of pH. As described elsewhere, cotton supports were firstly activated either negatively or positively. Figure 1 shows the evolution of water uptake in function of pH for untreated, cationized and anionized fabric in both cases: (a) without chitosan hydrogel incorporation and (b) chitosan hydrogel incorporation.

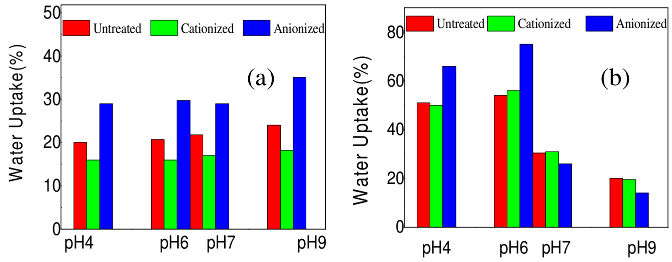

Fig. 1. Water Uptake (%) for untreated, cationized and anionized fibers: (a) without chitosan hydrogel (b) with hydrogel chitosan.

As presented in figure1 (a) and (b), the obtained water uptake values for non-activated cotton compared to treated cotton with hydrogel samples showed important Water Uptake increase in acidic medium (pH 4 and pH 6). This expected behavior was attributed to the pH responsiveness of chitosan as the consequence of the protonation of amino groups in acidic medium. In contrast samples treated with hydrogel showed lower Water Uptake in neutral and alkaline medium (pH 9) because chitosan does not contribute to swelling at this medium. This result indicates that the pH-dependency swelling of the chitosan hydrogel was successfully transformed into fabric.

The most obvious pH sensitivity of the functionalized cotton fabric was observed for anionic treated fabric (WU=75% at pH 6, WU=15% at pH 9). This assumption is in good agreement with the observation of the surface morphology by SEM (Figure 2).

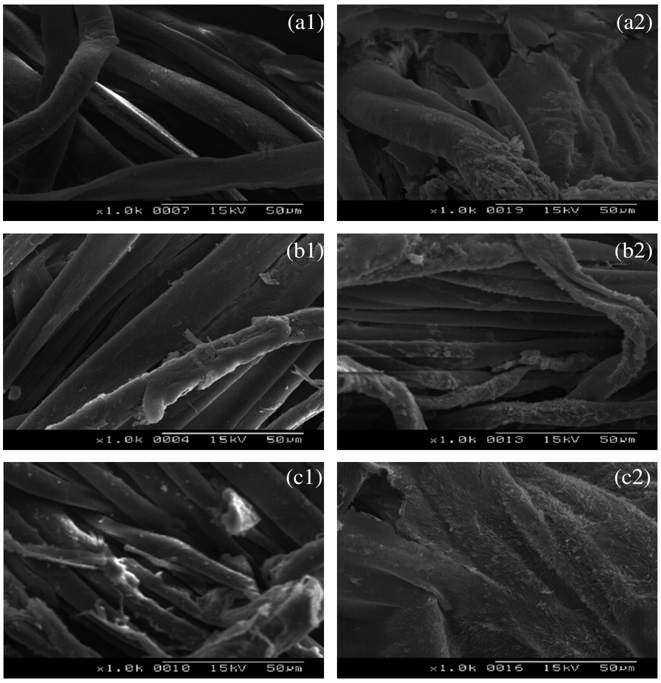

Fig. 2. SEM images of (a1) non-activated cotton (NAC), activated cotton (b1) AC and (c1) CC, and cotton with incorporated chitosan hydrogels (CH): (a2) NAC- CH, (b2) AC-CH and (c2) CA-CH.

Generally, the cotton fibers are slightly anionic in aqueous medium and then the cationic polymers can be absorbed onto the fibers by electrostatic attractions specifically by ionic bonds. In our case, after the cationic treatment the cotton makes ionic interactions with chitosan. The electrostatic attraction that take place between cellulose fibers (host material), and chitosan (absorbent) allowed a higher amounts of chitosan absorbed leading to a higher amount of amino groups and so a higher pH-sensitive swelling of chitosan.

B. Antibacterial Evaluation

The antibacterial activity is a complicated process that differs between Gram-positive and Gram negative bacteria especially due to cell surface characteristics of each bacteria. In our study, the antibacterial activity of the cited bacteria for activated and non-activated cotton fabrics before and after hydrogel treatment was carried out by the disc diffusion method (ASTM E2149-01). The inhibition zone diameters for each sample are presented in Table 1.

According to Table 1, the chitosan hydrogel improve the antibacterial activity of fabrics by increasing the inhibition diameter. The chitosan polymer has an antibacterial efficiency against both Gram-positive and Gram-negative bacteria based on the important inhibition zone diameter.

A high antibacterial activity against E.coli was reached (Gram-negative bacteria) with a maximum of inhibition diameter about 14 mm for treated and cationized fabrics. We attributed this result to the cationic character of chitosan providing higher affinity to negatively-charged biological membranes [9]. This can lead the leakage of intracellular substances [10, 11]. In fact, the mechanism involves interactions of chitosan polymer positively charged molecules with negatively charged constituents of microbial cell walls and membranes interrupting normal cell metabolism [12].

TABLE I. ANTIBACTERIAL BEHAVIOR AGAINST L.MONO, S.AUREUS AND E.COLI BACTERIA.

		Inhibition zone diameter (mm)		
		Gram positive bacteria		***Gram negative bacteria***
Sample		***L.mono***	***S.aureus***	***E. coli***
Without hydrogel	***Untreated***	11	19	7
	Cationized	17	10	7
	Aminized	11	17	6
With hydrogel	***Untreated***	15	25	7
	Cationized	20	17	12
	Aminized	18	17	14

The table shows also that inhibition zone diameter against Gram positive bacteria S. aureus and L.mono increased, compared to the reference untreated fibers. The highest zone of inhibition was about 25 mm with a non-activated cotton. This indicates that chitosan was sufficient to inhibit the growth of the used Gram positive bacteria. In addition to the cationic effect, antibacterial behavior can be improved by the release of the salicyladehyde by hydrolysis. This reagent has also antibacterial properties as detailed in the literature [13].

IV. CONCLUSION:

Results presented in this study proved that the concept of functional finishing of cotton via responsive hydrogel with antibacterial property could lead to the development of technical cotton. The swelling behavior of the obtained textile materials showed highly attractive feature of responsiveness to the pH environmental stimuli. Water Uptake increased in acidic medium (pH below pKa of chitosan) and decreased in neutral and alkaline medium (pH above pKa of chitosan).

Antibacterial tests showed that fabrics functionalized by chitosan hydrogel had better antibacterial capacity against either Gram positive or Gram negative bacteria.

Nevertheless, the encouraging obtained results, improving performance and cost-effectiveness, having application that is more homogeneous on the modified textile support with better linkages and finally meeting environmental and toxicity requirements, will continue to challenge this study.

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