Materials Science and Engineering C 80 (2017) 29–37

Contents lists available at ScienceDirect

Materials Science and Engineering C

journal homepage: www.elsevier.com/locate/msec

Design of multi-functional cotton gauze with antimicrobial and drug delivery properties

Mohamed Rehan [a], S. Zaghloul [a,*], F.A. Mahmoud [b], A.S. Montaser [a], A. Hebeish [a]

[a] Textile Research Division, Pre-treatment and Finishing of Cellulosic Fibers Department, National Research Centre, 33 Bohoth Street, Dokki, P.O. Box 12622, Cairo, Egypt
[b] Solid State Physics Department, National Research Centre (Scopus affiliation ID: 60014618), 33 Bohoth Street, Dokki, P.O. Box 12622, Cairo, Egypt

ARTICLE INFO

Article history:
Received 16 December 2016
Accepted 14 May 2017
Available online 15 May 2017

Keywords:
Cotton gauze
In-situ AgNPs
Oxytetracyline hydrochloride
Antimicrobial
Drug delivery

ABSTRACT

The ideal biomedical fiber/fabric materials can both promote the drug delivery properties and prevent microbial infection. Herein we present an innovation-based strategy for fabrication of biomedical cotton gauze which concomitantly displays antimicrobial and drug delivery performance properties. The innovative strategy involved three distinct steps: (1) Cationization of cotton gauze by reacting it with 3-chloro-2-hydroxypropyl trimethyl ammonium chloride [Quat-188] or anionization of cotton gauze through partial carboxymethylation. (2) Thus modified samples of cotton gauze along with unmodified blank samples were submitted to in situ formation of silver nanoparticles (AgNPs) using trisodium citrate (TSC) which has three-fold functions: (a) reducing agent for conversion of Ag^{+} to Ag^{o} (atom), (b) stabilizing agent to prevent aggregation of AgNPs and, (c) linker for fixation of AgNPs on the surfaces of the cotton gauze. (3) All the modified and unmodified cotton gauze samples were loaded with oxytetracyline hydrochloride drug. To this end, characterization of the modified and unmodified cotton samples before and after being loaded with drug using state-of-the-art facilities was undertaken. These facilities comprised UV–vis spectroscopy, energy dispersive X-ray, scanning electron microscope and Infrared Spectroscopy by Attenuated total Reflectance (ATR/IR). Evaluation of the antimicrobial and drug release properties of the cotton gauze samples in question was conducting. Results obtained signified that the modified cotton gauze can be used in the area of biomedical textiles particularly as antimicrobial and drug delivery. Also reported were mechanisms entailed in chemical modifications of cotton gauze and interactions of this modified cotton gauze with antimicrobial as well as with drugs.

1. Introduction

A perusal at the literature would reveal that the medical and health care sector demand heavily development of highly efficient textile materials of manifold purposes [1–7]. The fibrous nature of textile materials advocates them to have a dominant role in designing appropriate matrix for the biomedical sector industries [8,9]. However, by virtue of their porosity and large surface area, the textile materials provide humidity and temperature which favor the growth of bacteria on the surfaces of textiles. This, indeed, stimulates the antibacterial finishing treatments which impart biocidal activity to textiles. Accentuation of textile resistance towards microorganisms (to bring into focus antibacterial, antimicrobial and antifungal textile) has become a must at the medical textile industry [10–15]. This is exemplified by silver nanoparticles (AgNPs) which acquire broad antimicrobial activity against Gram positive and Gram negative microorganisms; AgNPs are used as antimicrobial agent in the textile field [16–19].

Textile drug delivery systems were developed with great emphasis on slow - or controlled delivery systems for the sake of attaining optimal therapeutic effect [20,21]. In normal practice, two approaches were devised for preparation of textile drug delivery system in medical textiles. The first approach involved incorporation of the drug via fixation, coating or encapsulation on the final form of fabric/fiber surfaces [22–26]. The second approach for preparation of textile drug delivery systems entailed processing of the drug during the preparation stage of the fabric/fiber [27–29].

Finishing technologies of textiles such as cationization, partial carboxymethlation and the in situ AgNPs formation processes were developed to enhance dyeing and printing ability as well as antibacterial and mechanical properties of the fabric/fiber [30–37]. Nevertheless, to the author's knowledge, no research work has been published so far on the effect of these particular processes on the antibacterial and drug delivery properties. With the above in mind, current work is undertaken with a view to submit cotton gauze, independently, to cationization, partial carboxymethylation and in situ AgNPs formation processes and, the onset of this on antibacterial activity and drug delivery performance of the as processed cotton gauze. The latter is selected on the bases of; being the most common wound dressing, low cost and

* Corresponding author.
E-mail addresses: drsaad_nrc2010@yahoo.com, mrsaad_nrc2003@yahoo.com (S. Zaghloul).

http://dx.doi.org/10.1016/j.msec.2017.05.093
0928-4931/

highly absorptive capacity. World-class facilities are used for characterization of the modified cotton, evaluation of their antimicrobial and drug release properties. Mechanisms involved in chemical modifications of cotton and interactions of this modified cotton with microbial as well as with drug are reported.

2. Materials and methods

2.1. Materials

Scoured and bleached cotton gauze, plain weave, was supplied by El-Nasr Company for spinning weaving and Dyeing, El-Mahallah El-Kubra, Egypt. 3-Chloro-2-hydroxypropyl trimethyl ammonium chloride (69%) of technical grade chemicals (known as Quatt-188) was purchased under the commercial name CR-2000 from Aldrich, Sodium hydroxide, acetic acid, monochloroacetic acid, sodium carbonate, silver nitrate and trisodium citrate were of laboratory grade chemicals. Oxytetracyline hydrochloride antibiotic drug was kindly provided by National Organization for Drug & Control Research, Haram, Giza, Egypt.

2.2. Methods

2.2.1. Cationization of cotton gauze

Chemical modification of the cotton gauze through cationization was carried out as per the pad-dry-cure method [38]. The experimental procedures adopted were as follows: 3-Chloro-2-hydroxypropyl trimethyl ammonium chloride (Quat-188) was mixed with sodium hydroxide solution at a NaOH/Quat-188 M ratio of 2:1. The cotton gauze was padded in this mixture in two dips and two nips, and then squeezed to a wet pick-up of about 100%. The cotton gauze was dried at 40 °C for 10 min and cured at 120 °C for 3 min. Thus treated cotton gauze was washed with cold water and 1% acetic acid, followed by several washing cycles and finally dried under the normal laboratory conditions.

2.2.2. Partial carboxymethylation of cotton gauze (PCMC)

Cotton gauze was partially carboxymethylated to yield (PCMC) by a method similar to those previously reported [39]. PCMC was produced in a two-stage process. The first stage refers to a mercerization process in which, bleached cotton gauze was impregnated with 15 wt% aqueous NaOH for 5 min at room temperature, squeezing to a wet pick up of 100% then dried at 60 °C for 5 min. Etherification is the second stage in which the alkali treated samples were steeped in aqueous solution of sodium salt of monochloroacetic acid (3 mol) for 5 min at room temperature. These samples were then squeezed to 100% wet pick up, sealed in plastic bags and heated at 80 °C for 1 h then washed and dried at room temperature.

2.2.3. In-situ Ag NPs incorporation into cotton gauze

In a typical experiment, samples ($10 \times 16\ cm^2$) of both the blank and modified cotton gauze were immersed in 200 ppm of silver nitrate solution with a liquor ratio of 1:50 and placed in water bath at 90 °C. To this end, 10 ml of 3% trisodium citrate (TSC), as reducing and capping agent, was added drop wise to each 100 ml silver nitrate solution and kept at 90 °C for 30 min. Subsequently, the cotton gauze samples were removed, squeezed and rinsed by running tap water and left to dry at ambient condition, meanwhile the absorbance of the residual solutions was measured by UV–vis spectra [4].

2.2.4. Loading of drug

The drug was selected based on the antibiotic sensitivity test. 1% Oxytetracyline hydrochloride antibiotic drug was dissolved separately in 100 ml of ethanol. Cotton gauze and modified cotton gauze samples were immersed in the drug solution and allowed to remain stand still for 1 h in the solution. The samples were then taken out and dried at room temperature [40,41].

2.3. Testing and analysis

2.3.1. Ultraviolet-visible (UV–vis) spectra

The absorption spectra of the residual solutions were measured using UV–vis multi-channel spectrophotometer (T80 UV/VIS, d = 10 mm, PG Instruments Ltd., Japan) ranged from 250 to 600 nm.

2.3.2. X-ray diffraction (XRD)

PANalytical X'pert PRO PW 3040/60 (Netherlands) X-ray diffraction fitted with a Cu Kα (λ = 0.154 nm) radiation source in range 2θ = (10°–80°) was used for phases identification and crystal structural analysis. The averaged crystallite size has been calculated using Deby-Scherrer formula; (Eq. (1)):

$$D = 0.9\,\lambda/\beta\,\cos\theta \tag{1}$$

where β is the observed angular width at half maximum intensity.

2.3.3. Scanning electron microscopy measurements and EDX

Microscopic investigations on fabric samples were carried out using a Philips XL30 scanning electron microscope (SEM) equipped with a LaB6 electron gun and a Philips-EDAX/DX4 energy-dispersive spectroscope (EDS). Images were taken at different magnifications (from 1509 to 3,0009), using secondary electrons (SE) in accordance with the clarity of the images. Fabric samples were fixed with carbon glue and metalized by gold vapor deposition to record images.

2.3.4. ATR/IR

ATR/IR (JASCO INFRARED SYSTEM) spectroscopes were used to record and analyze the blank and treated samples containing silver nanoparticles (AgNPs) and/or Oxytetracycline drug.

2.3.5. Color measurement

Colorimetric analysis of the colored fabrics was recorded using a spectrophotometer with pulsed xenon lamps as light source (UltraScan Pro, Hunter Lab, USA) 10° observer with D65 illuminant, d/2 viewing geometry and measurement area of 2 mm. All measurements were conducted at λ425 nm wavelength. The corresponding color strength value (K/S) was assessed by applying the Kubelka Munk (Eq. (2)) [42].

$$K/S = \frac{(1-R)^2}{2R} \tag{2}$$

where R is the decimal fraction of the reflection of the colored fabric, K is the absorption coefficient and S is the scattering coefficient.

Nowadays, in a lot of dye houses, there is a data match system which helps colorist to obtain different shades and to judge about the acceptance of these shades against a particular standard. The total color difference (ΔE) can be calculated from the CIE LAB color space data. In the present work, the color space (L*, a*, b*) of colored samples was measured by the same spectrophotometer used for measuring the color strength at the same set up, and then the color difference was calculated using (Eq. (3)) [43].

$$\Delta E = \left[(\Delta L^*)^2 + (\Delta b^*)^2 + (\Delta a^*)^2\right]^{1/2} \tag{3}$$

where, ΔE is the total difference between the sample and the stander, L^* is lightness from black (0) to white (100), a^* is a red (+)/green (−) ratio and b^* is yellow (+)/blue (−) ratio.

2.3.6. Evaluation of antimicrobial activities

The antimicrobial activity of untreated and treated cotton gauze was tested against *Escherichia coli* as Gram negative bacteria, *Staphylococcus aureus* as Gram positive bacteria and *Candida albican* as fungi. The antimicrobial test was performed quantitatively using the standard test