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Characterization and controlled release aloe extract of collagen protein modified cotton fiber

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ABSTRACT

For exploiting the novel multifunctional cotton fibers, a collagen protein modified cotton fiber (CPMCF) was prepared by the oxidation of cotton fiber with sodium periodate solution and subsequent crosslinking reaction with an aqueous solution of collagen protein in acetic acid. Infrared spectra and X-ray photoelectron spectrometry (XPS) analysis of the CPMCF illuminated that the C=N double bond was formed through the imine reaction of the aldehyde group on oxidized cotton fiber with the amino group of collagen protein. X-ray diffractograms indicated that the crystallinity of the oxidized cotton fiber increased from 65.6 to 69.3% after collagen protein treatment. Scanning electron microscopy photographs displayed that the collagen protein combined on the surface of oxidized cotton fiber. The resulting optimum conditions to prepare the CPMCF achieved the sufficient aldehyde groups in oxidized cotton fiber and the collagen protein content on CPMCF, whereas the mechanical strength of the oxidized cotton fiber had no significant change. Meanwhile, a model experiment for the controlled release of aloe anthraquinone extract on CPMCF showed a satisfactory result compared with those release of the original cotton fiber, demonstrated potential application of the synthetic collagen protein–cotton fiber as a carrier for the sustained release of drugs.

1. Introduction

Collagen protein, consisting mostly in the skin, bone, tendon, ligaments and blood vessel of animal, is one of the most abundant natural protein resources inside mammalian body with a large unexplored commercial potential. Collagen has the special three-ply helix construction formed through three polypeptide chains, one is the α(I) chain and the other two are the α(II) chains (Gelse, Pöschl, & Aigner, 2003; Giuseppe, Alexandre, Murray, Rolf, & Robert, 2000). Collagen protein is a vital structural protein of connective tissue which can support the organs and protect the organism, and is also the most important functional protein composing the substrate among cells. In recent years, a number of investigations have been carried out to exploit the potential applications of collagen protein (Cavallaro, Kemp, & Kraus, 1994; Takezawa, Ozaki, & Takabayashi, 2007; Wallace & Rosenblatt, 2003). Since collagen has unique structure, physiological and biological activities, low antigen, predominant biocompatibility and biodegradability, it has been applied widely in biomedical material, drug delivery carrier, tissue engineering, cosmetic, foodstuff and feedstuff, etc. (Brown, 2009; Ellis & Yannas, 1996; Fujioka, Takada, Sato, & Miyata, 1995; Madhan, Muralidharan, & Jayakumar, 2002; Wang, Su, & Chen, 2008). It is well known that the collagen protein not only is similar to the human skin collagenic structure but also has the perfect biocompatibility with the human body, which can supply necessary nutrition for the human skin and various amino acids that are helpful to the human body. In addition, the collagen protein can accelerate the skin tissue metabolism so as to moisten the human skin and delay the aging (Chvapil, Kronenthal, & Winkle, 1973; Stein, Vader, Weitz, & Sander, 2011).

Cotton fiber is a greatly vital natural cellulose fiber, which has been extensively utilized as textile, industrial product, construction material and medical commodity. Presently, lots of chemical modification methods by using crosslinking agents treatment, or irradiation of the gamma ray for bonding collagen protein on the poly(vinyl alcohol) and polypropylene fibers have been necessarily applied to improve the properties and find more versatile applications of these fibers (Lin, Dan, & Dan, 2012; Tyan, Liao, Lin, & Chen, 2003; Wang, Tang, Wu, Xu, & Ye, 2007; Wang, Wu, & Chen, 2011). However, the studies on collagen protein modified cotton fiber have not been reported previously. Our work now report a novel modification method by oxidizing the cotton fiber with sodium periodate, the periodate oxidation cleaves the C-2 and C-3 of the glucose units in the cotton fiber molecule, giving the reactive aldehyde groups in the oxidized fiber (Maekawa & Koshijima, 1991; Potthast, Kostic, Schiehser, Kosma, & Rosenau, 2007), subsequently treated with the

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collagen solution in aqueous acetic acid, then the imine bond produced by the reaction of aldehyde groups on the oxidized cotton cellulose with amino groups of the collagen protein. The resulting collagen protein modified cotton fiber (CPMCF) is a green ecological fiber material, and the preparing process is nonpolluting and eco-friendly without using chemical crosslinking agents.

Additionally, the aloe anthraquinone is extracted from the succulent leaves of a Chinese crude drug, Aloe vera. Aloe anthraquinone has antibacterial, anti-inflammatory, anti-hypersusceptible, anti-ultraviolet radiation, deodorizing and wound healing activities (Abd-Alla et al., 2009; Genovese et al., 2010; Ji & Jia, 2009; Mwale & Masika, 2010; Ranjani, Rajan, & Brindha, 2010). The loading and controlled release of aloe anthraquinone using our newly synthesized CPMCF would be significant to make full use of aloe extract pharmaceutical functions and achieve the efficacies of antibacteria, diminishing inflammation, moistening skin and anti-ultraviolet light, which is applicable to making underwear, bedding and medical commodity for atopic dermatitis.

2. Experimental

2.1. Materials

Cotton fibers were obtained from Hongxiang Spinning and Dyeing Co. Ltd. (Suzhou, China) and used with preliminary treatment to remove any additives and prevent interference from extraneous substances. Collagen protein with the average molecular weight about 10 kDa and aloe anthraquinone extract were purchased from Mingrang Biotechnology Co. Ltd. (Shanghai, China) and Tokyo Chemical Industry Co. Ltd. (Tokyo, Japan), respectively. All chemicals used for the following investigations were purchased from Sinopharm Chemical Reagent Co. Ltd. (Shanghai, China) and of analytical grade. Deionized water was used throughout the following work.

2.2. Sodium periodate oxidation of cotton fiber

Desired amount of cotton fiber was immersed in aqueous solutions of sodium periodate ranging in concentrations from 0.1 to 5.0 g/l at a liquor ratio 1:100. The cotton samples were stirred gently at 50 °C for 1 h in the absence of light. Then the oxidized cotton fiber was soaked in 0.1% (w/w) excess ethylene glycol solution with stirring for 30 min at ambient temperature to remove the remaining oxidant, and washed with deionized water up to neutral conditions. The oxidized cotton fiber was used for the next reaction without drying.

2.3. Imine reaction of oxidized cotton fiber with collagen protein

A collagen protein solution was prepared by stirring a dispersion of collagen in aqueous acetic acid solution at pH value of 4.0, then the above mentioned oxidized cotton fiber was immersed in 1.0% (w/w) collagen protein solution with constant oscillating for 1 h at 40 °C, subsequently submitted to thermal treatment at 60 °C under vacuum for 3 h, then scoured with deionized water several times. The resultant cotton fiber was air-dried at ambient temperature to produce the collagen protein modified cotton fiber (CPMCF).

2.4. Measurement

2.4.1. Aldehyde group content of oxidized cotton fiber

The aldehyde group content in oxidized cotton fibers was determined by the Schiff base reaction with hydroxylamine hydrochloride (Marte & Owens, 1956). Hydrochloric acid released from hydroxylamine hydrochloride was titrated by 0.03 M NaOH methanol solution which of concentration was determined with the titrimetry of potassium hydrogen phthalate (Qian & Li, 2001). The formula is as follows:

$$\text{Aldehyde group content } (\text{mmol/g}) = 30V/W \quad (1)$$

where V is the volume of sodium hydroxide methanol solution used in titrimetry (l); W is the mass of oxidized cotton fiber sample (g).

2.4.2. Collagen protein content in CPMCF sample

The modified cotton fiber samples were precisely weighed and then suspended in concentrated sulfuric acid (5 ml). Five drops of hydrogen peroxide (30%) was added to the suspension and the mixture was heated under reflux until the solution became transparent and colorless. The resulting solutions were subjected to Kjeldahl nitrogen analysis by the DS-20 analyzer (Tecator Instrument Co. Ltd., Sweden). The collagen protein content in the CPMCF was calculated from the nitrogen percentage on the basis of the calibration curve for the weight of collagen protein and test value.

2.4.3. Instrumental measurements

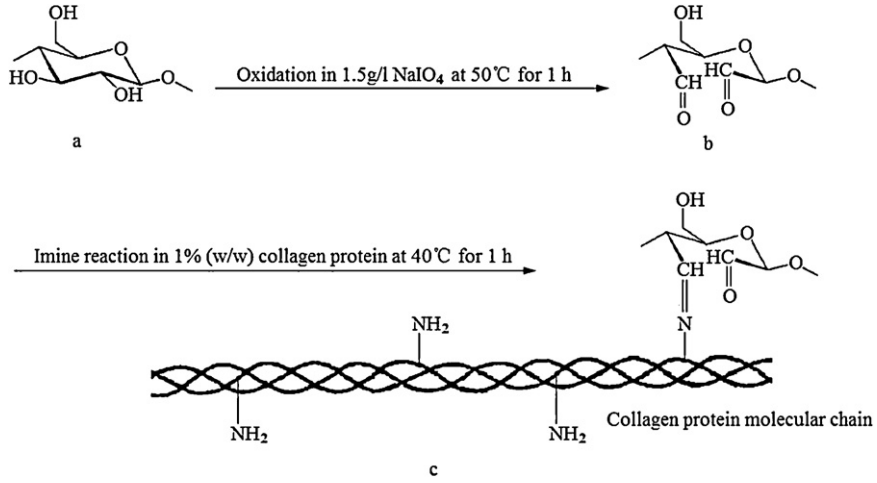
The Fourier transform infrared spectra of the oxidized fiber and the CPMCF were recorded with a NEXUS-870 FT-IR spectrometer (Nicolet Instrument Co. Ltd., USA) using a Globar source. Approximately 1 mg of sample was pressed into discs of variable thickness of potassium bromide, and samples were analyzed in transmittance, with accumulation of 50 scans and a resolution of 2/cm. Scanning electron microscopy photographs were taken on a Japan Hitachi S-4800 electronic instrument operating at 2–8 kV after sputtering the samples with gold. The X-ray diffractometry profile was recorded for dry pellet of the samples in reflection mode using a Japan RINT 2027 X-ray generator equipped with a Cu Kα target and β Ni filter. Diffractometer scans at a rate of 2°/min and a 2θ range of 5–45°. The breaking strength of the cotton thread was measured with a YG-021A electronic automatic graph tension tester (Changzhou Textile Instrument Factory, China). The length of the sample thread was 250 mm, the rate of extension was 100 mm/min, each sample was measured 10 times and the tests were performed at 20 °C and a relative humidity of 65%.

2.5. XPS of the oxidized cotton fiber and the CPMCF

X-ray photoelectron spectrometry (XPS) of the oxidized fiber and the CPMCF were performed in FAT mode with the X-ray gun of Mg target (1253.6 eV) at the power of 12 kV × 15 mA, and the analytical background vacuum of 2×10^{-7} Pa, the channel energy of 100 eV and the step length of 0.1 eV/s, using a Kratos XSAM-800 multifunctional spectrometric apparatus (VG Science Instrument Co. Ltd., Britain). The surface of samples were sputtered with the Ar^+ ion to eliminate interference from contaminated substances and subsequently preserved in high vacuum.

2.6. Loading and controlled release aloe extract of CPMCF

The CPMCF (2.5 g) was added into the 2.0% (w/w) aloe anthraquinone extract solution and stirred slightly for 2 h at 60 °C, subsequently dried in vacuum oven at the same temperature for 1 h, then the colored cotton thread was vigorously washed with deionized water to remove the aloe extract adsorbed physically on the surface of the fiber. The original cotton thread (2.5 g) was also treated by aloe extract using the same procedure to produce the control sample. The aloe extract-treated thread and the control sample were separately added to an isotonic sodium chloride solution (200 ml) in two Erlenmeyer flasks. Each flask was shaken at 37 °C for different days (4, 6, 8 and 10 days), and the solutions were replaced with fresh sodium chloride solutions every 24 h. The aloe anthraquinone extract released into the sodium chloride



Scheme 1. Proposed schematic reaction for crosslinking collagen protein into cotton fiber oxidized by $NaIO_4$ solution.

solution was measured with a Japan Shimadzu UV-3600 ultraviolet spectrophotometer at the maximum observation wavelength of 297 nm.

3. Results and discussion

3.1. Preparation and characterization of CPMCF

The preparation process of CPMCF is clarified in Scheme 1. Periodate oxidation of cotton fiber resulted in a ring-opened product containing two aldehyde groups formed by selectively cleave the 2,3-vicinal diol groups of the anhydro D-glucopyranose residues in cotton cellulose molecules (a), obtaining the 2,3-dialdehyde cellulose (b). Subsequently, the oxidized cotton was reacted with a collagen protein solution in acetic acid, the resulting active aldehyde group in the oxidized cotton fiber would combined with an amino group of collagen protein to give the CPMCF (c).

The synthetic reaction process was monitored by infrared spectroscopy. Fig. 1 shows the spectra of the original cotton fiber, the oxidized cotton fiber and the CPMCF, respectively. The characteristic absorption band of the oxidized cellulose clearly appeared at 1729.8 cm^{-1} owing to the stretching vibration of the C=O double bond of the aldehyde group, disappeared after imine reaction with collagen protein. Furthermore, an absorption peak at near 898.5 cm^{-1} in the infrared spectrum (b) of the oxidized fiber indicates the formation of a cyclic hemiacetal linkage (Maekawa & Koshijima, 1991), is not apparent as shown in the IR spectrum (a) and (c). After treatment by collagen protein, the characteristic absorption peak at 1714.7 cm^{-1} in the spectrum of the CPMCF due to C=N stretching of the Schiff base reveals that the imine bond was formed between the aldehyde group of oxidized fiber and the amino group on collagen protein, meanwhile the absorption band at 1521.5 cm^{-1} in the CPMCF spectrum corresponds to the C—N stretching of the primary amine in collagen protein. These infrared spectroscopic results suggest that collagen protein has been coupled on the oxidized cotton fiber through the imine reaction of aldehyde groups of the oxidized cotton fiber with amino groups of collagen protein.

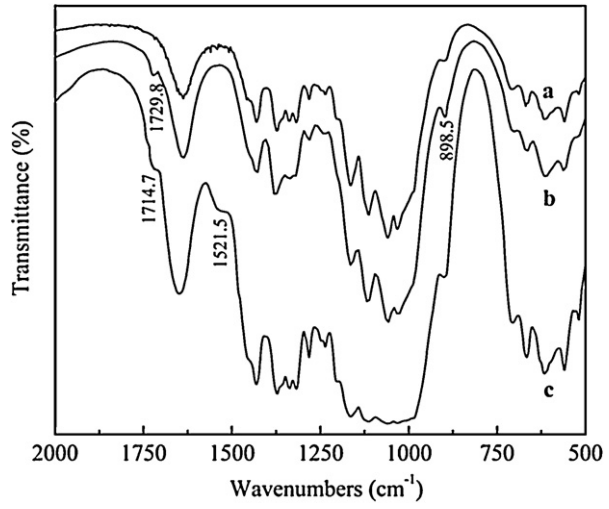


Fig. 1. FT-IR spectra of: (a) original cotton fiber; (b) oxidized cotton fiber; (c) CPMCF.

3.2. Scanning electron microscope of CPMCF

The longitudinal surfaces of the oxidized fiber and the CPMCF were morphologically observed by SEM. Many long and narrow stripes were displayed on the surface of the oxidized cotton fiber as shown in Fig. 2A. This information is explained that cotton fibers were suffered from mild corrosion of sodium periodate in the oxidation process (Janjic et al., 2009). In addition, as shown in Fig. 2B, the surface of the CPMCF had a different morphological figuration from the oxidized cotton fiber that showed a smooth surface. Meanwhile, lots of bulk and filmy substances existed on the CPMCF surface after the reaction with collagen protein. From the SEM finding, it was concluded that the collagen protein was introduced onto the surface of the oxidized cotton fiber and deposited in the crevices of the fiber to cover with a layer of collagen through a series of reactions, which provide a suitable carrier for bonding and releasing the aloe anthraquinone extract.

3.3. Confirmation of collagen protein on CPMCF by XPS

We further examined the crosslinkage between the collagen protein and the oxidized cotton cellulose by the X-ray photoelectron spectrometry (XPS) that has been widely used in chemical elements analysis. Before the XPS testing, the samples were scoured fully with deionized water to eliminate the collagen protein adsorbed on the fiber surface. Since the molecular structures of both oxidized cellulose and collagen protein contain the C—O atomic bond, therefore, the carbon absorption peak of the C—O bond at 286.6 eV is used as the standard to revise the binding energy position for the testing samples of XPS.

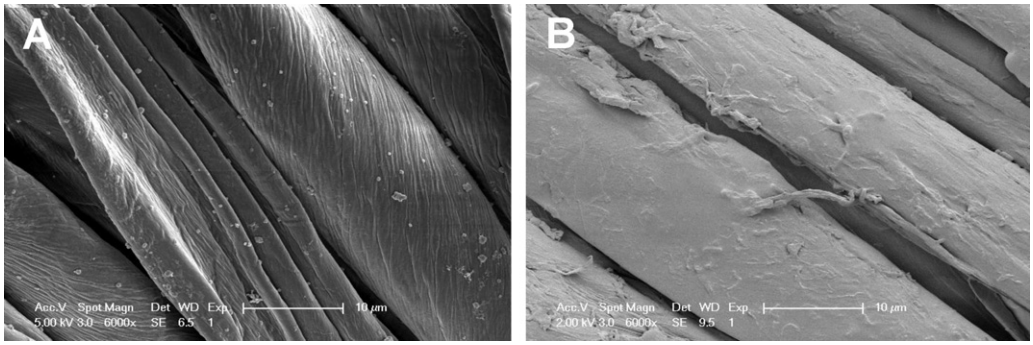


Fig. 2. SEM photomicrographs of: (A) oxidized cotton fiber; (B) CPMCF.

The XPS spectrum of the oxidized fiber shows that the oxidized cellulose had almost no absorption peaks of the nitrogen element except the absorption peaks of carbon and oxygen, due to the oxidized cotton fiber still consisting of the cellulose and containing no nitrogen elements. After modification by collagen protein, although the surface of the CPMCF was sputtered with the ion gun to remove the exterior contaminated layer, the XPS spectrum of CPMCF clearly shows a nitrogen characteristic absorption peak around 402.3 eV as shown in Fig. 3b. Additionally, the oxygen absorption peak of the CPMCF shifted from 531.6 to 532.5 eV on account of the electron attraction effect of carboxy groups in collagen protein. Meanwhile, the percentage of areas corresponding to the absorption peaks of carbon and oxygen of the CPMCF in Fig. 3b increased from 90.28 to 96.59%, suggesting the collagen protein molecules contain a mass of carbon and oxygen elements. So the XPS analysis illuminated that the crosslinking reaction occurred between the amino group of the collagen protein and the aldehyde group on the oxidized cellulose, and the collagen molecules through the C=N covalent bond of the Schiff base fixed on the oxidized cotton fiber.

3.4. X-ray diffraction analysis

The X-ray diffractograms of the oxidized cotton fiber and the CPMCF are shown in Fig. 4. It is discovered that the X-ray diffractive curve form and the diffractive angles corresponding to the diffraction characteristic peaks of the oxidized fiber were similar to the diffractogram of CPMCF. These phenomena indicated that the aggregating structure inside the oxidized cotton fiber had nearly no changes after the collagen protein modification.

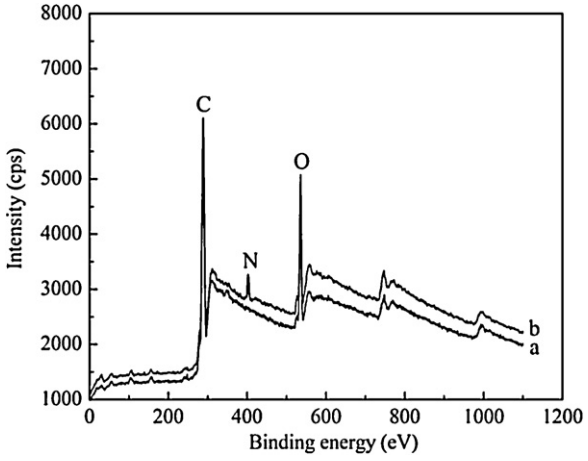


Fig. 3. XPS spectra of: (a) oxidized cotton fiber; (b) collagen protein modified cotton fiber.

The X-ray diffraction peak intensity at $2\theta = 22.7^\circ$ of the CPMCF diffractogram was higher than that of the oxidized cotton fiber, meanwhile the crystallinities of the cellulose samples were calculated through utilizing the PeakFit software program to separate the diffraction crystalline peaks and imitate the diffraction curves as shown in Fig. 4A and B respectively, and the baseline of Gaussian deconvolution is used and the peak numerical fitting ratio is 99.99% in the diffractive curve imitation by the PeakFit software. The oxidized cotton fiber has a crystallinity of 65.6%, as compared to 69.3% crystallinity for the CPMCF. This slight increase in the crystallinity of CPMCF may demonstrate that the collagen protein molecules crosslinked mainly with the amorphous region and the exposed surface of the oxidized cellulose, which intensified the intermolecular hydrogen bonding in the oxidized cotton fiber macromolecules and led to improving the crystalline structure of the CPMCF. However, the imine reaction with collagen protein did not have a significant impact on the microcrystalline structure of the oxidized cotton fiber.

3.5. Optimizing preparation condition of CPMCF

For seeking an optimum condition to prepare the CPMCF, the mechanical property of the oxidized cotton fiber and the collagen protein content on the CPMCF calculated by the Kjeldahl nitrogen analysis under different periodate concentrations were tested as a measure of the amount of aldehyde groups produced in the oxidized fiber as shown in Figs. 5 and 6. Theoretically, the aldehyde group content of the oxidized cellulose should increase with the time, temperature and oxidant concentration in the oxidation process, whereas the periodate ion is difficult to penetrate and diffuse into the inner crystalline phase of cotton fiber, which results in long time and mass oxidant required to the periodate oxidation at normal temperature. Consequently, a little higher temperature may quicken the oxidative reaction and cut down the oxidation time and oxidant dosage (Varma & Kulkarni, 2002). We can get high collagen protein content when reacted with 1% collagen protein solution at 40 °C for 60 min due to the high aldehyde content subsequently enhancing the amount of the collagen protein linked to the oxidized fiber, which is very favorable for the CPMCF to load and release aloe extract. However, the periodate oxidation breaks to some extent the crystalline structure of cellulose in the original cotton fiber, therefore, the fiber mechanical property may be weakened with aldehyde groups elevation.

Figs. 5 and 6 denote the effects of the concentration of preceded oxidation on the breaking strength and aldehyde content of the oxidized cotton fiber and the collagen protein content of the CPMCF. As an expecting result, it was observed that the breaking strength of the oxidized cotton fiber did not change remarkably during the oxidation for the periodate concentration less than 1.8 g/l, whereas

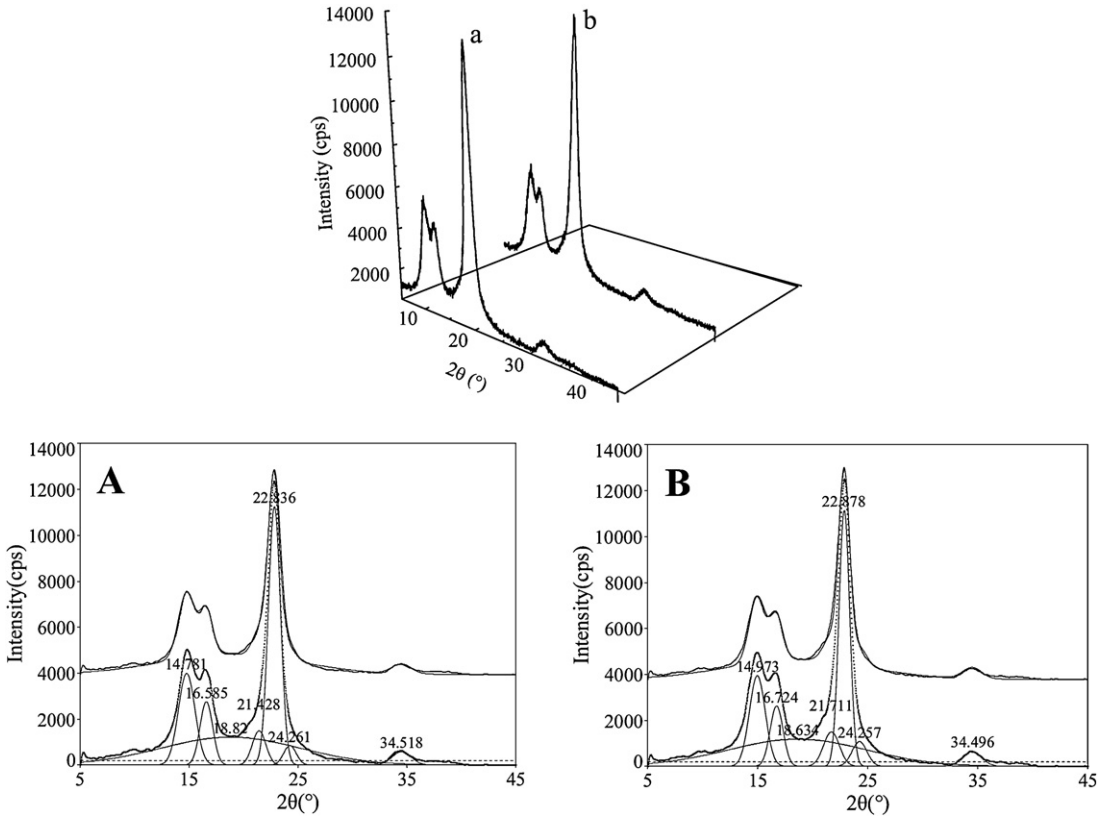


Fig. 4. X-ray diffractograms of oxidized cotton fiber (a) and CPMCF (b), peaks separation and curves imitation of 'a' diffractogram (A) and 'b' diffractogram (B).

decreased dramatically at the oxidant concentration over 2.0 g/l, probably on account of breaking down the crystallization and the intermolecular interaction of cotton fiber.

Meanwhile, Fig. 6 shows that low periodate concentration yielded adequate aldehyde content for the oxidized fiber to achieve sufficient amount of collagen protein integrated on the oxidized cotton fiber. The collagen protein content increased with the oxidant concentration during the initial oxidation stage, then decreased at higher concentrations of sodium periodate till the collagen protein content became nearly constant, and the maximum collagen protein content of the CPMCF was 1.75% of the weight of cotton fiber while the collagen protein content of the original cotton fiber was only 0.05%. This finding is speculated to the difference in the reaction site of the periodate oxidation and imine bond formation in the cotton fiber (Hou, Liu, Liu, Duan, & Bai, 2008; Kim & Kuga, 2000), and the small periodate ion may access into the amorphous and crystalline regions of cotton fiber to oxidize the cotton fiber surface and inside structures. However, the collagen protein due to large molecules is hard to arrive in the fiber inner area, so that the reaction with collagen protein proceeds mainly on the surface of the oxidized cotton fiber. From above condition analysis, the oxidation of cotton fiber in 1.5 g/l sodium periodate solution at 50 °C for 1 h was determined to reach such aldehyde group content.

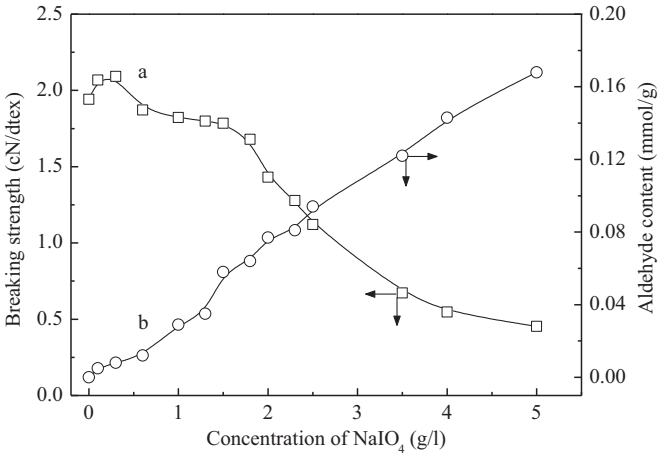


Fig. 5. Change in breaking strength (a) and aldehyde content (b) of the oxidized cotton fiber against the $NaIO_4$ concentration.

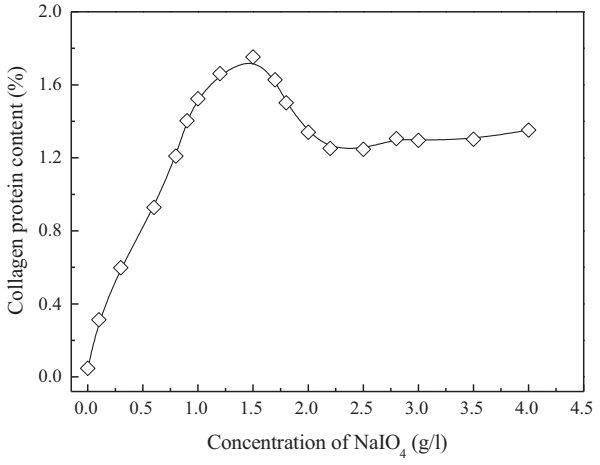


Fig. 6. Relationship between the collagen protein content of the CPMCF and the $NaIO_4$ concentration in preceded oxidation.

Table 1
Controlled release of aloe anthraquinone extract adsorbed on the cotton fibers.

Release time (days)	Absorbency	
	Original cotton fiber	CPMCF
4	0.098	0.172
6	0.086	0.140
8	0.075	0.126
10	0.052	0.121
Residual aloe extract[a]	0.169	1.013

[a] Residual aloe anthraquinone extract on the cotton fibers were extracted with ethanol and measured in the same conditions.

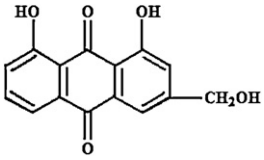


Fig. 7. Molecular structure of aloe anthraquinone.

3.6. The controlled release aloe anthraquinone on CPMCF

The time-course release of aloe anthraquinone extract on the CPMCF and the original cotton fiber is examined at different days. After the same treatment of the CPMCF and the original cotton fiber in an aqueous aloe extract solution, the release aloe anthraquinone into an isotonic aqueous sodium chloride and the residual aloe anthraquinone on the fibers extracted with ethanol after 10 days were respectively subjected to ultraviolet spectrophotometric analysis. These testing results are summarized in Table 1, which indicated that a larger amount of aloe anthraquinone was released from the CPMCF; moreover, the more aloe anthraquinone extract remained on the CPMCF. Compared with the original cotton fiber in release aloe extract, approximately 3.3 times as much aloe anthraquinone was fixed on the CPMCF and then slowly released after the 10-days experiment. This phenomenon is explained that the sufficient polycationic collagen protein residues of the CPMCF may interact with the acidic functional groups in the aloe anthraquinone molecule as shown in Fig. 7, whereas a few aloe anthraquinone adsorb simply on the original cotton fiber. Accordingly, using the CPMCF as a carrier to release the aloe anthraquinone has an expansive foreground in producing antibacterial underwear.

4. Conclusions

The studies here reported obtained a chemical synthesized cotton fiber with the collagen protein modification (CPMCF) by the imine reaction between the aldehyde groups of oxidized cotton fiber and the amino groups in collagen protein. In order to confirm that the amount of collagen protein crosslinked on the oxidized cotton fiber is sufficient to retain the mechanical property of the oxidized cotton fiber and achieve the controlled release aloe anthraquinone on the CPMCF, the selective oxidation of cotton fiber in the sodium periodate aqueous solution with a concentration of 1.5 g/l at 50 °C for 1 h is chosen as the optimal condition, and yielding the aldehyde group content of 0.058 mmol/g in the oxidized cotton fiber and the collagen protein content of 1.75% on the CPMCF.

FT-IR spectra and XPS analysis show that the collagen protein molecule through the C=N double bond is crosslinked with the oxidized cotton fiber, meanwhile X-ray diffractograms of the cotton fibers suggest that the aggregating structure inside the oxidized cotton fiber has almost no changes, and the calculations by the diffraction peaks separation and the diffraction curves imitation confirm that the crystallinity of the oxidized cotton fiber slightly increased after reaction with the collagen protein.

In this way the CPMCF surface crosslinked with the collagen protein displays physiological and biological activities, which improve the affinity and comfort with the human body. Since the chemical reactivity of both the amino group and the carboxyl group in CPMCF are stronger than the hydroxyl group of the original cellulose, the resulting CPMCF possesses more potential for further chemical modification. The CPMCF prepared under optimum conditions for release aloe anthraquinone showed a satisfactory result in controlled release of the liliaceous medicine aloe anthraquinone, and SEM photomicrographs denote the collagen protein formed the membrane on the CPMCF, which indicated that the ability of aloe extract combination enhances greater. Therefore, crosslinking the collagen protein onto the oxidized cotton fiber would be feasible to utilize as a sustained release drug carrier.

Acknowledgements

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