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Combined Cutinase and Keratinolytic Enzyme to Endow Improved Shrink-resistance to Wool Fabric

Nan Zhang, Panghui Huang, Ping Wang, Yuanyuan Yu, Man Zhou, and Qiang Wang*

Key Laboratory of Science and Technology of Eco-Textile, Ministry of Education, Jiangnan University, Wuxi 214122, Jiangsu, China

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Abstract: The overlapping cuticle scales on the wool surface cause severe felting shrinkage during laundering. However, the conventional wool anti-felting processing mostly adopts dichloroisocyanurate (DCCA), which produces absorbable organic halogen (AOX) with high toxicity in the effluents and cause severe environmental problems. Herein, an environmental-friendly enzymatic method was proposed and investigated to endow satisfactory shrink-proofing property for wool fabrics without severe damages. The cutinase from *Thermobifida fusca* and the keratinolytic enzyme from *Bacillus subtilis* were consecutively used to treat wool fabrics for 4 h and 24 h, respectively. The area shrinkage of the resultant wool fabrics decreased from 12.4 % to 5.86 % for to the cuticle was partly broken, which met the machine-washable requirement with an acceptable strength loss of 7.46 %. Moreover, the dyeability and wettability of the resultant fabric were improved after the combined enzymatic treatments, due to the destruction of the lipid layer and breakdown of keratin by combined cutinase and keratinolytic enzyme according to the analyses such as scanning electron microscope (SEM) and X-ray photoelectron spectroscopy (XPS) and contact angle. Overall, our result revealed that the synergistic actions of cutinase and keratinolytic enzyme treatments could effectively disintegrate cuticles and remove scales.

Keywords: Cutinase, Keratinolytic enzyme, Wool, Bio-antifelting, Functional modification

Introduction

Wool is one of the most important natural fibers and is usually used to manufacture top-grade suit fabrics for its excellent properties like elasticity, fullness, warmth retention, and comfortability [1]. However, wool has a specific overlapping cuticle scale structure on the surface, which causes severe shrinkage when it is subjected to mechanical processes under wet and heated conditions, such as laundering. This shrinkage is attributed to the differential frictional effect (DFE), in which the friction exerted in the direction of scale is lower than that against the direction of the scales [2]. Moreover, wool scale is essential to the mechanical and moisture-absorbing properties of wool fiber. To reduce the felting tendency of wool, the scales' edges should be smoothened or partly removed to reduce the DFE of wool fibers [3].

Wool scale layers are composed of hydrophobic lipid-like substances and highly cross-linked keratin, which contains a high content of disulfide bonds and is inert to general chemicals and proteases [4-6]. Currently, the most extensively applied chlorination/resin anti-felting method can achieve satisfactory shrinkage, while its release of high-toxic absorbable organic halogens (AOX) in the effluents causes severe environmental pollution [7,8]. Considering the ecological and economical restrictions in the textile industry, it is necessary to explore eco-friendly alternatives for wool processing.

To date, various alternative methods were proposed to remove wool scale including reduction [9], plasma [10], and enzymatic treatments [11]. Among them, enzymatic treatments have great potential to replace the chlorination method because enzymes are environmental-friendly and react under mild conditions [8]. Protease is the enzyme that catalyzes the hydrolysis of peptide linkage in proteins. However, owing to the hydrophobic lipid layer, the high-crosslinked disulfide bonds, and closely assembled keratins in wool scales [12, 13], proteases preferentially attack the non-keratinous proteins of the CMC in the cortex by channeling beneath the overlapping cuticle cells, resulting in unacceptable and irreversible fiber damage [14]. Therefore, how to restrict the enzymatic attack on wool cuticle rather than CMC has attracted great attention. Yoon *et al.* found that corrosion of the hydrophobic lipid layer using plasma treatment increased the accessibility of protease to cuticle surface [15]. Some researchers increased the size of proteases by covalently-bonded with polymers like PEG [16], Eudragit S100 [17,18], etc, and the modified proteases endowed the resultant wool fabric improved felting property with less strength loss. However, the modified protease still showed limited ability to degrade wool cuticle due to the hydrophobic lipid layer and the high-crosslinked disulfide bonds in the wool cuticle. The pretreatment using reagents that can effectively cleave the disulfide bonds like H_2O_2, $KMnO_4$, Na_2SO_3 significantly enhanced the effect of protease hydrolysis of the wool cuticle [18]. Levene *et al.* pretreated wool fabrics with sodium sulfite then followed with the treatment of 16 commercial proteases, while only six proteases including esperase, savinase, SP 490, type XXXI, subtilisin, and papain endowing wool fabric

*Corresponding author: qiangwang@jiangnan.edu.cn

satisfactory percent of shrinkage (less than 6 %). Besides, the strength loss of the resultant wool fabrics treated with esperase was about 10-12 %, while that of wool fabrics treated with the other five enzymes was 16-24 % [11]. Therefore, the selection of enzymes for wool anti-felting also affects the damage degree to wool fabrics.

Apart from those common proteases, keratinolytic enzymes can also be considered as potential enzymes for wool surface modification, since keratinolytic enzymes were reported to be able to efficiently hydrolyze keratin. Various strains have been screened and modified to produce keratinolytic enzymes with high enzyme activity [19]. The keratinolytic enzymes are often used in producing feather meal or dehairing of leather [20], while a few was used in wool anti-felting [21]. Tu *et al.* combined keratinase H328 and Protease K to treat wool fabric, the result suggested that the protease K strongly decreased the tensile strength while the keratinase H328 treatment does not affect the tensile strength [22]. Therefore, it is necessary to explore keratinolytic enzyme endowing wool fabrics ideal anti-felting property with low strength loss. Wang *et al.* proposed a three-step enzymatic process using cutinase, keratinase, and savinase treatment which endowed wool fabrics satisfied area shrinkage with a strength loss of 14 % [23]. The cutinase-protease treatment also altered the hydrophobic property of wool surface for the cutinase can hydrolyze line-chain aliphatic esters on the wool surface [23,24].

Considering that the lipid layers on the outermost of the scales hindered the accessibility of keratinolytic enzymes to wool cuticles, wool fabrics were pretreated with cutinase to remove the lipid layers to maximize the function of the keratinolytic enzyme. The purpose of this study is to erode the wool cuticle by mild and combined enzymatic treatment without causing severe damages to the physical properties of wool fabric. Then the area shrinkage, dyeability, and wettability of the resultant wool fabrics were examined and analyzed. To further discuss the reaction mechanism between wool and enzymes, the surface morphology and elemental analysis of the treated wool samples were examined and analyzed in detail.

Experimental

Materials

The cutinase from *T.fusca* with an enzyme activity of 99 U/m*l* was kindly supplied by the School of Food Science and Technology, Jiangnan University. The activity of cutinase was determined using p-nitrophenylbutyrate (pNPB) as the substrate. One unit of enzyme activity is defined as the production of 1 μmol pNP per min. The keratinolytic enzyme from *Bacillus subtilis* with an activity of 200 U/m*l* was kindly supplied by the School of Biotechnology, Jiangnan University. The activity of the keratinolytic enzyme was determined using 1 % keratin solution as substrate according to Liu *et al.* [26]. One unit of enzyme is defined as the amount of enzyme that produce 1 ug tyrosine per min at pH 8.0 and 50 °C. The commercial acid dye C.I. Weak Acid Blue 80 was supplied by Dystar Co. (Shanghai, China). The pure wool fabrics (328 g/m^2) were provided by Xiexin Group Co. (Wuxi, China). Wool fabrics or fibers were cleaned with methanol/chloroform 13:87 (v/v) at 65 °C for 4 h to remove the free lipids and other impurities from the wool fibers.

Enzymatic Treatment of Wool

Wool fiber or fabrics samples were pretreated with the cutinase (0-6 U/g fabric) in Tris-HCl buffer (0.05 M, pH 8.0) at 50 °C for 4 h, and then treated with the keratinolytic enzyme (0-800 U/g fabric) in Tris-HCl buffer (0.05 M, pH 8.0) at 50 °C for varying time (0-48 h). The bath ratio was 50:1 in both enzymatic treatments. After the treatment, the three sets of parallel samples were rinsed with deionized water three times and dried at 50 °C.

Evaluation and Characterization

Percent of Area Shrinkage

The percent of area shrinkage of fabrics was determined using Woolmark Test Method TM31: washing of wool textile products and new standard ISO 6330:2012. The wool fabrics were subjected to one 4G wash cycle (equal to 7A wash cycle in IOS 6330:2000) for relaxing shrinkage and three 4N wash cycles (corresponded to 5A wash cycle in ISO 6330:2000) for felting shrinkage using a Y(B) 098D Automatic Shrinkage Testing Machine (Darong Textile Instrument Company, China). The three sets of parallel samples were performed and then the average percent of area shrinkage of wool fabric was calculated [16].

Tensile Properties

The tensile property of wool fabric was evaluated using a YG(B) 026D-250 Electronic Fabric Strength Tester (Darong Textile Instrument Company, Wenzhou, China) following the procedures described in ISO 13934.-1:2013 [16]. Briefly, the wool fabric was cut into 5 cm (weft direction)×30 cm (warp direction) and fixed using the clamps with a gauge length of 20 cm. The tensile breaking strength was recorded with a testing speed of 20 mm/min. Each sample was measured 5 times to obtain the average value. The percentages of strength loss were calculated using equation (1).

$$\text{Percent of strength loss (\%)} = (N_0 - N_i)/N_0 \times 100\% \quad (1)$$

where N_0 and N_i are the tensile strengths before and after enzymatic treatment, respectively.

Surface Morphology Observation

The morphological analysis of wool fiber surfaces before and after treatments was visualized using a Quanta 200 scanning electron microscope (FEI, Holland) at 1000× magnification and operating at a typical accelerating voltage

of 10 kV. The samples were sputter-coated with gold before observation.

XPS Experiment

XPS experiments were conducted using an RBD upgraded PHI-5000C ESCA system (Perkin Elmer, USA) with Mg Ka radiation (h=1253.6 eV) or Al Ka radiation (h=1486.6 eV). The X-ray anode was run at 250 W with a high voltage of 14.0 kV and a detection angle of 54 °. The pass energy was fixed at 23.5, 46.95, or 93.90 eV to ensure sufficient resolution and sensitivity. The base pressure of the analyzer chamber was 5×10^{-8} Pa. Data were analyzed using the RBD AugerScan 3.21 software provided by RBD Enterprises [1].

Wettability

The wettability of the wool samples was evaluated through the contact angle, which was tested using a JC2000D4 contact angle tester (Zhongchen Digital Technical Apparatus Company, Shanghai, China). Distilled water droplet of 20 μ*l* was injected onto the surface of wool fabrics using a fixed steel needle. The images were captured once per minute until the droplet retains on samples for 20 min. Contact angles were measured at three different points for each sample.

Dyeability

The wool samples were dyed with C.I. Acid Blue 80 (C.I.61585, 1 % o.w.f., on the weight of fabric) at pH 4.5 and 90 °C for 1 h with a liquid ratio of 100:1. The dyeability of wool was determined using a UV-802S spectrophotometer (Unico, China) and expressed in terms of dyeing exhaustion. The wavelength of maximum absorbance (λ_{max}) of the dye used in this study was 550 nm. The dyeing exhaustion was calculated using equation (2):

$$E\,(\%) = (A_0 - A_t)/A_0 \times 100\% \quad (2)$$

Where E is the exhaustion percentage; A_0 and A_t are the absorbances of the dye bath before and after dyeing at 550 nm, respectively.

The color depth (K/S values) of wool samples was examined using a Gretag Macbeth Color-Eye 7000A spectrophotometer (Datacolor, New Windsor, USA).

Results and Discussion

Optimization of the Dosage and Treatment Time of Cutinase and Keratinolytic Enzyme

Cutinase was used to pretreat the wool fabrics to degrade the lipid on the surface of wool fibers. The wettability of the wool fabrics was used to evaluate the extent of lipid removal from the wool surface. As shown in Figure 1(a) and (b), the control wool fabrics had poor wettability with a contact angle of 139.44 ° and a wetting time of 25.57 min. With increasing the dosage of cutinase and the treatment time, the wettability of wool fabrics becomes better. When the dosage of cutinase was higher than 6.0 U/g fabrics, the downward trend of the contact angle flatted out (Figure 1(a)). As shown

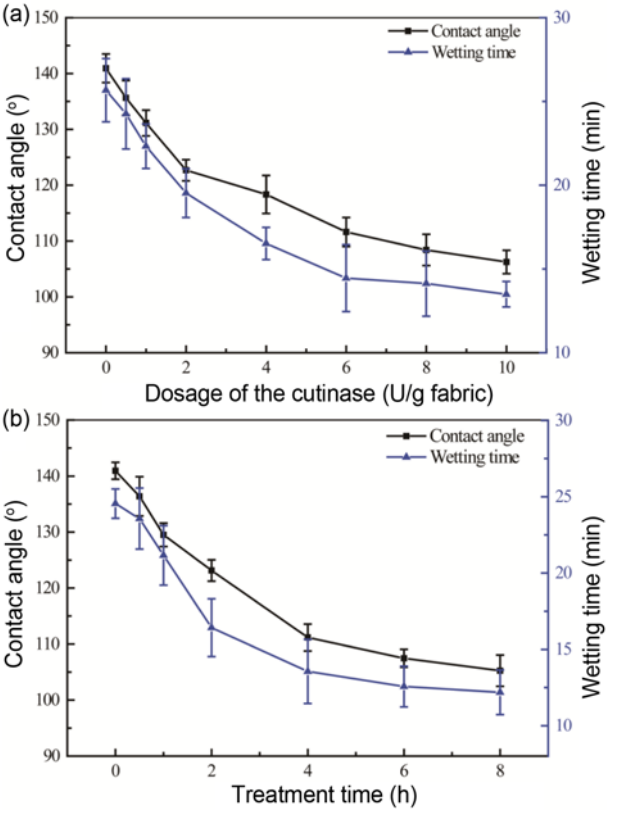

Figure 1. Relationship between (a) dosage and (b) treatment time of cutinase and wettability of wool fabrics.

in Figure 1(b), the result shown that when the processing time was 4 h, the contact angle and wetting time of the resultant wool fabrics greatly decreased to 111.17 ° and 13.56 min, respectively. Therefore, wool fabrics were pretreated with cutinase (6.0 U/g fabrics) at 50 °C for 4 h.

As shown in Figure 2(a) and (b), the keratinolytic has the maximum enzymatic activity at 50 °C and pH 8.5. In this study, the treatment condition was pH 8.0 and 50 °C, where the relative enzyme activity was about 96.06 %. Wool fibers contained approximately 10 % of cuticle scales on the surface [27]. Partly removal of wool cuticle scale resulted in damage to the structure of wool fibers and strength loss of wool fabrics. Therefore, the percent area shrinkage and the strength loss of the resultant wool fabrics were the key evaluation criterion.

As shown in Figure 2(c), with increasement the dosage of the keratinolytic enzyme, the shrinkage area of the wool fabrics decreased. Besides, the shrinkage area of wool fabrics pretreated with cutinase was lower than that of unpretreated wool fabrics. The area shrinkage of wool samples treated with sole cutinase decreased from 10.3 % to 9.7 %, with a slight strength loss of approximately 1.3 %. These results indicated the individual cutinase treatment slightly improve the anti-felting property of wool fabrics.

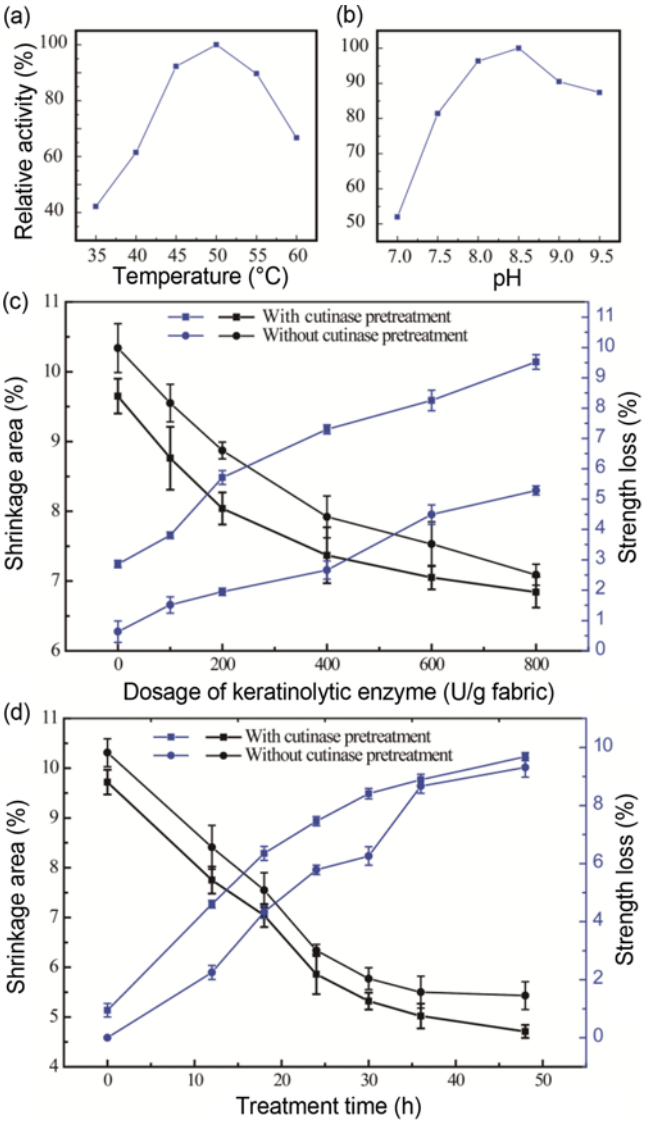

Figure 2. Effect of (a) temperature and (b) pH on the activity of the keratinolytic enzyme, (b) optimized pH of the keratinolytic enzyme, effect of dosage of (c) keratinolytic enzyme and (d) treatment time on area shrinkage and percent of strength loss of wool fabrics after pretreatment with cutinase.

While wool fabrics were treated with the subsequent keratinolytic enzyme for 12 h, the area shrinkage of the resultant wool fabrics decreased with the increment of the keratinolytic enzyme. When the dosage of the keratinolytic enzyme was up to 400 U/g fabrics, the area shrinkage of the resultant wool fabrics decreased slowly. The strength loss of the wool fabrics increased remarkably from 7.5 % to 9.8 %. Here, the optimized dosage of the keratinase was 400 U/g fabrics. As shown in Figure 2(d), the shrinkage area of wool fabrics increased with the prolongation of treatment time, while the strength loss showed a contrary trend. Wool fabrics without cutinase pretreatment require more enzyme (600 U/g fabric) or longer processing time (30 h) to gain an ideal anti-felting property. When the wool fabric with cutinase pretreatment was treated with keratinolytic enzyme for 24 h, the area shrinkage of wool fabrics was 5.86 % meeting the requirement of machine washable with an acceptable strength loss of 7.46 %. Therefore, the optimized treatment time of the keratinolytic enzyme was 24 h. The percent of strength loss of wool fabrics treated with the combined cutinase and keratinolytic enzyme was much higher compared to the wool fabrics treated with the individual keratinolytic enzyme. These results demonstrated that cutinase pretreatment could enhance the action of keratinolytic enzyme on wool in the subsequent treatment [23]. The cutinase only hydrolyzed the ester bonds of the wool surface lipid layer but did not break the scale structures, thus, the pretreatment merely influenced the shrink-proofing property of wool samples. However, the removal of the surface lipid layer facilitated the following hydrolysis of keratinolytic enzyme, resulting in the decrease in the percentage of area shrinkage when the wool fabrics undertook cutinase and keratinolytic enzyme treatments in succession. The lipid structure of the epicuticle was disrupted by the cutinase, improving the accessibility of the keratinolytic enzyme to the cuticle.

Surface Characterization of Wool Fibers

Morphology of Wool Fibers (SEM)

The surface morphologies of wool fibers treated with different treatments were shown in Figure 3. The untreated wool fibers showed characteristic overlapping scale layers structure, and the edge of each cuticle has a clear boundary (Figure 3(a)). After the wool fibers were treated with cutinase, little scale damage can be observed for the partial hydrolysis of the lipid layer scarcely affected the keratins in wool scales (Figures 3(b)). Regarding of the wool fibers treated with a single keratinolytic enzyme, remarkable scale damages and the flake shape of cuticle edges were observed due to the gradual hydrolysis of the exocuticle (Figure 3(c)). After wool fibers underwent the combined treatment of cutinase and keratinolytic enzyme, and some cuticle fragments attached on wool surfaces were detected (Figure 3(d)), indicating the scales were partially removed.

Surface Element Analysis with XPS

The XPS can detect the relative concentration of elements presenting at a depth of approximately 10 nm can be determined, which is almost the same as the depth of the lipid layer of the wool surface [28]. Therefore, the XPS was used to investigate the variation of the wool surface after wool underwent different enzymatic treatments. The main elements detected on the wool surface were C, O, N, and S. The XPS spectra of untreated wool and wool treated with various enzymatic treatments were shown in Figure 4.

The XPS spectra showed that the primary elements detected on both wool samples were carbon (C), oxygen (O),

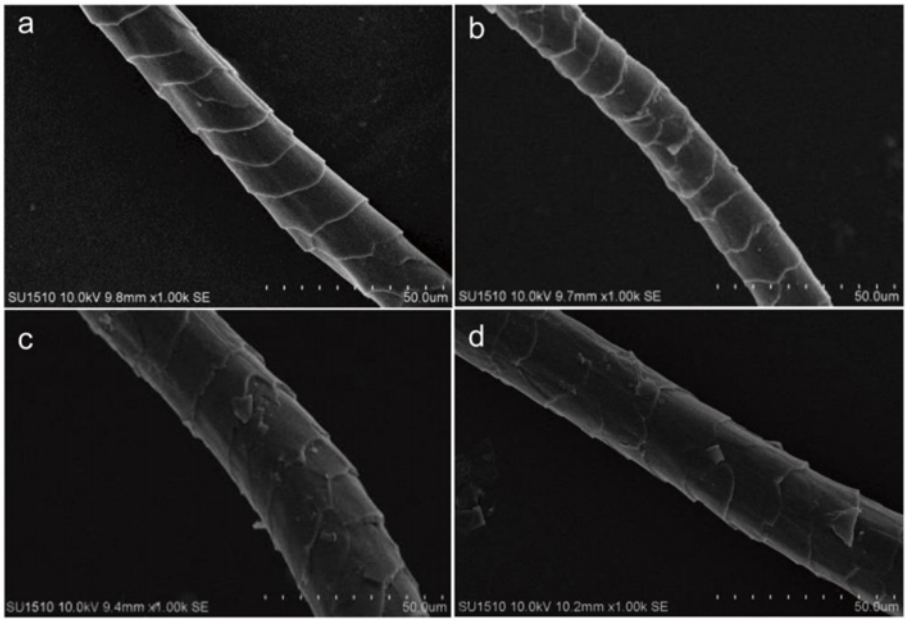

Figure 3. SEM images of (a) untreated wool fibers and wool fibers treated with (b) cutinase, (c) keratinolytic enzyme, and (d) combined cutinase and keratinolytic enzyme.

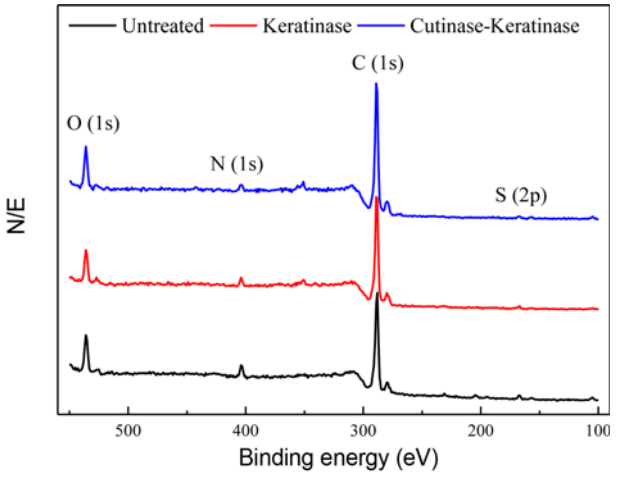

Figure 4. XPS spectra of untreated wool and wool treated with combined cutinase and keratinolytic enzyme.

Table 1. Elemental composition (%) of untreated wool and wool treated with combined cutinase and keratinolytic enzyme

Sample	C1s	O1s	N1s	S2p
Untreated	85.05	9.83	2.45	1.23
Keratinolytic enzyme-treated	83.18	10.19	2.45	0.61
Cutinase-Keratinolytic enzyme	79.91	11.32	5.69	0.70

nitrogen (N), and sulfur (S). The major peak centered at 288.0 eV with a shoulder at lower binding energies was ascribed to C 1s. While the signal peaks of O 1s, N 1s, and S 2p were located at 536.0, 404.0 eV, and 167.0 eV, respectively [29]. As shown in Table 1, the carbon content of the untreated wool was 85.05 %, which was more than that of the whole wool (50-55 %) due to the presence of a lipid layer on the wool surface [30]. Compared to the virgin wool, the wool fabric treated with the individual keratinolytic enzyme and combined cutinase and keratinolytic enzymes showed a decrease of 1.87 % and 6.04 % in carbon content. Besides, the content of N on the surface of wool fabric treated with combined cutinase and keratinolytic enzyme (5.69 %) was higher than that of untreated and keratinolytic enzyme-treated wool fabric (2.45 %). These results indicated that the scales layer of wool fibers was partially damaged and removed due to the removal of surface lipid with the cutinase and disruption of the keratin in the cuticle scale with the keratinolytic enzyme. The underlying hydrophilic groups like amino ($-NH_2$), hydroxyl (-OH), and carboxyl (-COOH) groups were exposed, resulting in the change of elemental composition of the wool surface [30].

The XPS spectra for C 1s and S 2p regions of untreated wool and wool treated with a single keratinolytic enzyme and combined cutinase and keratinolytic enzyme were shown in Figures 5. Three components were fitted to the C 1s spectrum centered at 284.6, 286.3, and 287.7 eV (Figure 5(a), (b)). The peak at 284.6 eV was ascribed as C-C, C-H, and C-S bonds in the hydrocarbon backbone of covalently bound fatty acids and side groups of the amino acids [29]. While the peak at 286.3 eV corresponded to the presence of C-O and C-N bonds associated with proteins. The peak at 287.7 eV was due to the carboxyl and amide group. The C 1s spectrum of wool undergoing the cutinase-

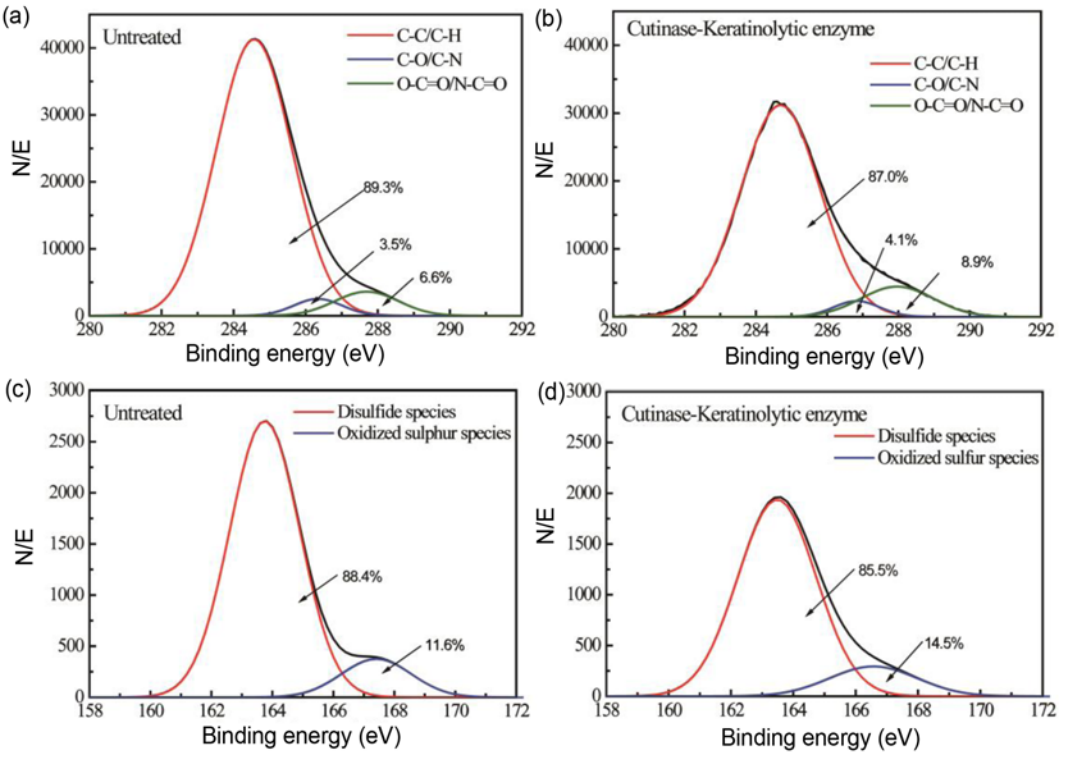

Figure 5. XPS C 1s and S 2p spectra of untreated wool and wool treated with combined cutinase and keratinolytic enzyme.

keratinolytic enzyme treatment showed similar resolved peaks. However, compared to the untreated wool, the contents of C-C and C-H bonds decreased evidently while the content of (N)O-C=O bonds increased in the wool treated with combined cutinase and keratinolytic enzyme because the the cutinase effectively hydrolyzed and remove the lipid layer.

The S 2p spectra of both wool samples consisted of two components peak at about 164 eV and 168 eV (Figure 5(c) and (d)) which were assigned to disulfide species and oxidized sulfur species, respectively [31]. Compared to the untreated wool sample, the relative content of disulfide species slightly decreased while that of oxidized sulfur species increased in wool treated with combined cutinase and keratinolytic enzyme. Those results indicated that some disulfide bonds in wool surface were cleaved and finally formed the final oxidation product after the wool lipid layer was disrupted by cutinase.

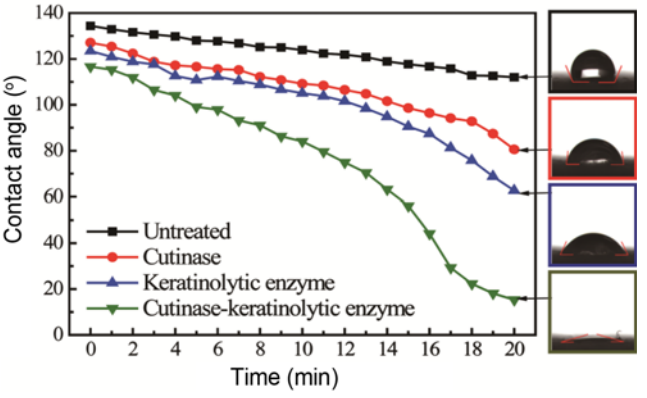

Figure 6. Contact angle of untreated wool fabric and wool fabrics treated with single cutinase, keratinolytic enzyme, and combined cutinase and keratinolytic enzyme (the illustrated pictures were the contact angle after the water droplets contacted the surface of fabric samples for 20 min).

Wettability and Dyeability of Wool Fabrics with Enzymatic Treatment

Wettability

Untreated wool fabrics possess strong hydrophobicity owing to the overlapping cuticle structure and the hydrophobic lipid layer on the surface of wool fibers [32]. Therefore, the degree of scale removal could be assessed in terms of the wettability of the wool fabrics. As shown in Figure 6, the contact angle of untreated wool was 134.4 ° and decreased to 122.4 ° within 20 min, showing a strong hydrophobicity and poor wettability. After the wool fabrics were treated with single cutinase, single keratinolytic enzyme, and combined cutinase and keratinolytic enzyme, the initial contact angle decreased slightly to 127.1 °, 122.1 °, and 116.3 °, respectively; and the contact angles at 20 min were 80 °, 60.8 °, 15.37 °, respectively. Those results

Table 2. Dyeing behaviors of untreated wool fabrics and wool fabrics treated with cutinase, keratinolytic enzyme, and combined cutinase and keratinolytic enzyme

Sample	Exhaustion (%)	K/S
Untreated	12.42±0.51	3.284
Cutinase-treated	18.45±0.37	4.190
Keratinolytic enzyme-treated	26.80±0.45	4.951
Cutinase-Keratinolytic enzyme-treated	35.64±0.39	6.492

indicated that a single enzymatic treatment slightly improved the wettability of wool fabrics. The cutinase treatment partially destroyed the lipid layers on the surface of wool fibers but did not further damage the cuticle scales. While the keratinolytic enzyme showed low accessibility without destroying the lipid layers in advance. The combined enzymatic treatment greatly improved the wettability of the resultant wool fabrics. This improvement was due to the cutinase pretreatment facilitated the accessibility of keratinolytic enzyme to the surface of wool fibers and promoted keratin hydrolytic reactions.

Dyeing Behaviors

Theoretically, the adsorption and diffusion of dye molecules in the fibers should be improved as the wettability of wool fabrics was improved after enzymatic treatments. The untreated wool fabrics and enzymatically treated wool fabrics were dyed with C.I. Acid Blue 80 and their dyeing properties were listed in Table 2. The dye exhaustion percentage was only 12.42 % and the K/S value of the untreated wool was 3.284, indicating untreated wool fibers had a poor dyeability. Owing to the impenetrable and hydrophobic cuticle scales, water-soluble dye diffused in CMC from the gaps between scales then spreads into the whole fiber along with the CMC [33]. The dyeing properties of wool samples treated with single cutinase, keratinolytic enzyme, and combined cutinase and keratinolytic enzyme increased by degrees. The dyeability of wool fabrics that underwent combined enzymatic treatment was better than that of wool fabrics that underwent individual enzymatic treatments. The *K/S* value and exhaustion percentage of wool fabric treated with combined enzymes reached 6.492 and 35.64 %, respectively.

Conclusion

In this work, we investigate the effect of the single keratinolytic from *B. subtilis* and combined cutinase and the keratinolytic enzyme on surface modification of wool fabrics. The combined enzymatic treatments can remarkably modify the surface structure and improve the properties of wool fabric. The resultant wool fabrics showed satisfactory shrink-proofing property to wool fabrics with strength loss of about 7.46 %. SEM micrographs showed that the scales of wool fibers were eroded and the XPS analysis indicated the elemental composition of the wool surface was changed after combined enzymatic treatments. Thus, the wettability and dyeing properties of the resultant wool fabrics were greatly improved. The combined enzymatic anti-felting method is eco-friendly and can endow wool fabrics with better properties and performances, having great potential to be an alternative to the traditional chemical anti-felting processes.

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