International Journal of Textile Science 2014, 3(4): 64-69
DOI: 10.5923/j.textile.20140304.02

Applications of Transglutaminase in Textile, Wool, and Leather Processing

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Abstract Transglutaminase (TGase) has been used in food industry since it reconstitutes small meat pieces into a steak. In addition, its application in pharmaceutical industry is well investigated and still under further study. However, the application of TGases in textile and leather industry was minimal before a decade. Hence, this paper reviews the potential applications of TGases in textile and leather fabrication. The enzyme recovers the wool and silk damaged during chemical and enzymatic treatment at different stages of wool and silk processing. It enhances the shrink resistance of the wool and it improves the tensile strength of the wool fibers. In addition, TGase allows the grafting of amines or proteins to bring desired properties in wool fibers. Furthermore, smoothed and better color fastness can be obtained from wool treated with TGase. TGase is also used to fill voids with caseins and gelatines in leather industry. TGases play a great role in wool, silk and leather processing.

Keywords Transglutaminase, Wool, Silk, Leather, Shrink resistance, Tensile strength

1. Introduction

TGases (EC 2.3.2.13) are a group of enzymes capable of catalyzing the acyl transfer reaction between the γ-carboxyamide groups in Gln residues of peptide or protein and ε-amino groups in Lys residues, resulting in the formation of ε-(γ-glutamyl) lysine linkages and the release of ammonia [1]. In this reaction, the γ-carboxyamide group of glutamine and the ε-amino group of lysine function are the acyl donor and the acceptor, respectively. The covalent conjugation of polyamines, lipid esterification, or the deamidation of glutamine residues is responsible for posttranslational modification of proteins by protein-to-protein cross-linking using TGase.

TGases have been found in a variety of different organisms such as mammals, plants, crustaceans, fish, a wide range of invertebrates and microorganisms [2, 3]. However, the microbial transglutaminase (mTGase) is more important than others since it can be produced at industrial level easily by simple fermentation. The optimum pH for mTGases activity is between 5 and 8. However, at pH 4 or 9, some activity of mTGase was reported, and was thus considered to be stable over a wide pH range [4]. MTGase from *Streptomyces* sp. exhibited optimal activity in the 6.0–6.5 pH range and a second maximum of activity was observed at pH 10 with both the crude *Streptomyces* sp. enzyme and the commercial enzyme [3]. The optimum temperature for enzymatic activity is 55℃; it maintained full activity for 10 min at 40℃, but lost activity within a few minutes at 70℃; and it was active at 10℃, and retained some activity at near-freezing temperatures [1].

MTGases can catalyze acyl transfer by forming covalent crosslinks among proteins, peptides, and primary amines. Unlike mammalian TGases, the mTGases do not require calcium for activity [2, 5] and have a broader substrate specificity range [2, 5], and can be produced at relatively low cost [5]. These properties are advantageous for industrial applications [5]. Though mTGases were independent of Ca^{+2} concentration, but they were elevated in the presence of K^{+}, Ba^{2+}, and Co^{2+} and inhibited by Cu^{2+} and Hg^{2+}, which suggests the presence of a thiol group in the mTGase's active site [3].

The main applications of mTGase in food industries are well discussed. However, novel potential applications of TGase in biomedical engineering, material science, textiles and leather processing have emerged during the last decade. This paper reviews the trends of TGase application in textile and leather industry.

2. Transglutaminase Application in Wool Industry

Wool is a textile fiber obtained from sheep, goat, rabbits and other animals. It passes through varies processing steps to be ready for wearable clothes. These steps are scouring, carding, gilling, combing, drafting, spinning and twisting. An array of chemicals and enzymes are employed at these

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Published online at http://journal.sapub.org/textile

different stages and the treatments change the wool properties in undesired manner. TGase recovers these damages caused by treatments at different steps.

2.1. Recovering Damages Caused by Proteolytic Treatment

Protease found in biological detergents can successfully remove protein stains and they also hydrolyze natural protein fibres such as wool keratins and silk causing severe and irreversible damage to the garments [6]. In general, application of protease enzyme technology in wool processing results in considerable loss of tensile strength by diffusion of the enzyme into the interior of wool fibers. To overcome this disadvantage, enzymatic activity has been more targeted to the outer surface of the scales by improving the susceptibility of the outer surface scale protein for proteolytic degradation. This has been realized by a pretreatment of wool with hydrogen peroxide at alkaline pH in the presence of high concentrations of salt [7].

Although commercial biological detergents state on the packaging "Do not wash wool or silk", it is a common misjudgment, either through ignorance or by accident, for household laundry of wool goods to be carried out using biological detergents. Common detergents (in particular alkaline detergents) commonly damage wool garments. Changes in fabric structure are exhibited in wool garments, often resulting in distortions in fabric shape. The fabric becomes weak and holes may appear. All these effects are irreversible, and occur with increased severity when biological detergents containing proteases are used [8].

TGase treatment not only reduced the loss of strength caused by protease treatment but also increased the strength of the yarn in greater extent [9]. The proteolytic treatment in terms of yarn strength resulted 14.9% strength loss and this loss was fully recovered using Guinea pig liver transglutaminase (GPL tTGase) and mTGase [10]. There was no application difference between the two types of transglutaminase in strength loss recovery. However, mTGase was applied at a protein concentration 20-fold greater than that of GPL tTGase.

Treating wool fibres with a protease may enhance the benefits of TGase treatments, since the increase in tensile and elongation properties with a TGase treatment when compared to their respective controls is higher for Savinase treated samples. The reason for this effect may be the opening up of the wool fibre structure by the proteases, which would facilitate TGase crosslinking via ε(γ-glutamyl) lysine bridges [10].

2.2. Recovering Damages Caused by Chemicals Treatment

Different chemical methods are known and widely used commercially to produce shrinkproof wools. The most common methods involve an acid chlorination of the wools or the application of permonossulphuric acid (PMS), followed by a polymer application [11]. Alternatively, coating of the wool fibres using a polymer or monomers that are polymerised on the fibre surface is also used to mask the scalar structure of the fibres. Such methods achieve a significant level of shrink-resistance to wool textiles, but may affect adversely the handle properties, as well as generating damaging substances that may be released into the environment. The oxidative chemical used in such kind of treatment include PMS, chlorine and peroxycarboximidic acid whereas the reducing chemical include sodium sulphite. The use of all these chemicals damages the wool structure in general and wool strength in particular. The effect of a TGase treatment on 100% wool yarns previously treated with chlorine, PMS or sodium sulphite was therefore investigated to determine if the losses in strength and elongation could be recovered [10]. Likewise, felting shrinkage of wool fabrics can be controlled by another oxidative chemical peroxycarboximidic acid [12], however it results strength loss. This loss was recovered by cross-linking keratins in the wool fibers using transglutaminase. TGase transamidation proved to control the dimensional stability of wool with the inference that keratin can self-crosslink and crosslink to proteinous substrates such as body tissue and to a variety of Gly–Lys carboxamide-active agents [13].

Wool bleaching is usually carried out by using an oxidizing agent in classical wool processing and this reduces the tensile strength of wool yarn. However, the application of mTGase to bleached wool restores the tensile strength. For instances, the mTGases treatment on the wool yarns bleached with H_2O_2 improved the tensile strength and whiteness along with the higher alkali resistance [14]. Similarly, KMnO4 pretreatment significantly damaged the scale structure of wool surface, however, the surface morphology was not altered when mTGase was applied after chemical pretreatment [15].

Similarly, the use of TGases can remediate the fibre damage resulting from chemical treatments, and such recovery is similar for 5µg/ml of GPL tTGase and 100µg/ml of mTGase. Treatments with both tissue and microbial TGases lead to significant increases in fibre strength for chlorine, PMS or sodium sulphite treatments [10].

2.3. Upgrading Shrink Resistance (Anti-felt)

Shrink resistance could be upgraded by direct treatment of the wool by transglutaminase or by the incorporation of other proteins like gelatin and casein to change the surface properties of the wool so that the desired property could be obtained. Direct treatment of the wool with transglutaminase was investigated to know its effect on shrink resistance; the felting shrinkage level decreased from 15% for the control sample to 6% for the samples treated with TGase [10]. On the other hand, the incorporation of other proteins to the wool also had significant effect on shrink resistance. For example, TGase-mediated crosslinking of gelatin on the surface of wool and its effect on the properties of wool fabric were investigated by [16] and gelatin treatment for 1 h combined with microbial TGase reduced the area shrinkage of $KMnO_4$-pretreated wool fabric from 6.53 to 1.92 %, which was more effective than that treated with gelatin alone (in which the area shrinkage was reduced to 4.02). Similarly, the incorporation of casein with mTGase reduced the area shrinkage of the $KMnO_4$-pretreated wool fabric from 11.33 to 4.58 % [17]. However, comparing with a kind of traditional resin anti-felting treatment, the tensile strength of the fabrics treated with mTGase was lower than that by resin treatment [18]. On the other study, the felting shrinkage of wool treated with mTGase reduced from 9.3% to 2.3% [19].

2.4. Incorporation of Amines or Proteins into Wool Fibres

TGase may also be used to incorporate a functional alkyl amine moiety into wool proteins that give desired or beneficial effects to the wool, thus increasing its potential in the wool industry [20]. Amines are attached by the incorporation of fluorescein cadaverine, thus raising the prospect of novel and exciting finishes [10]. Active functional agents that have the requisite primary amine group may be incorporated in this way to provide a beneficial effect, e.g. anti-microbial agents, sunscreens, water repellents and perfumes. If such compounds do not have an alkyl amino side group, it may be possible to chemically modify them to include one, making such compounds potential TGase substrates which can then be used for incorporation into wool fibres. For instance, the role of transglutaminase for the covalent bonding of *o*-phosphorylethanolamine to wool and its resistance to washing is investigated by [21]. As a result, it was found that the covalent bonding was resistant for four washings. However, the addition of *o*-phosphorylethanolamine with transglutaminase did not affect the tensile strength of the fabric even though the elongation property was dramatically increased.

The incorporation of proteins helps to bring desired wool properties in textile industries. For instances, silk serecin proteins was grafted to wool fabrics using mTGase [2]. The TGase mediated grafting of these proteins led to a significant effect on the properties of wool yarn and fabric, resulting in increased bursting strength, as well as reduced levels of felting shrinkage and improved fabric softness.

2.5. Making the Wool Softer (Smoothed)

One of the problems associated with wool is its tactile discomfort (itchiness). However, softness of the wool is one of the criteria that guide wool quality in the market. Improvement in softness and the handle of wool can be achieved by addition of various chemical agents such as silicone softener or by the addition of proteolytic enzymes [22]. The cost of these improvements may be greater than the moderate benefits achieved. Changes in one property of wool can affect other properties, sometimes adversely. For example, protease treatments normally have adverse effects on strength and weight of wool material. The addition of proteins or amines to the wool improved the surface texture of the wool. The incorporation of gelatin [16] and casein [17] to the wool fibers using TGase smoothed the wool fiber surface by coating or filling the raised scales of the wool. In the same manner, the addition of *o*-phosphorylethanolamine to the wool with transglutaminase made the wool whiter with a softer feel than the untreated fibre [21].

2.6. Wettability of Wool Fabrics

TGase could improve wool fabrics wettability. With the increase of mTGase concentrations, the contact angles of treated samples greatly reduced and the time of water penetration shortened obviously [20]. When wool fabric was treated by protease and then treated with 32% (owf) of TGase, the wettability could be distinctly improved [23]. The wettability is one factor in color fastness and hence it also plays a great role in wool dyeing.

2.7. Effects on Color Fastness

The dyeing properties of three natural dyes (curcumin, gardenia yellow and lac dye) on wool fabric after treatment with mTGase have been investigated [24] and it was found that after 120 min of mTGase treatment, compared with the fabric only pretreated with chemical and protease, the colour strength of curcumin, gardenia yellow and lac dye increased from 8, 7.5 and 22 to 12.8, 11.7 and 27, respectively. The values of wash fastness for dyed wool fabrics increased from 2 to 4 after mTGase treatment, but the light fastness was not obviously improved [24].

In the other study, TGase treatments have no influence on the washing fastness for samples dyed with sappan [25]. However, treatments with TGase enhance *K/S* value of wool dyed subsequently without influencing rubbing fastness [23, 25]. Furthermore, the use of mTGase could improve the dyeing properties of treated wool fabrics. With an increase in its concentration, the initial dye exhaustion increased and the time to reach the dyeing equilibrium was also shortened. It was evident that the improvement of dyeing properties was closely related to the improvement of wettability performance of wool using mTGase [20].

2.8. Tensile Strength

Tensile strength is the maximum stress sustained under a tensile force without fracture [26]. In other definition, it is a measure of a steady force that is necessary to break a fibre [21]. TGase-mediated crosslinking of gelatin on the surface of wool recovered the tensile strength from 267 to 335 N [16]. For casein and TGase treated wool, the tensile strength increased from 275 to 315 N [17]. In addition, the strength of wool fibers treated with mTGase increased by 30% compare untreated control [19].

2.9. Antibacterial Functionalization

The wool fibers commonly suffer from degradation and infection due to proliferation of microorganisms under relatively hot and humid condition. This problem has been solved using inorganic and organic antibacterial agents [27] and promising results have been obtained. ε-Poly L-lysine (ε-PL) is a homo-polypeptide of L-lysine with the amide linkages between the ε-amino and carboxyl groups and it is now industrially produced by *Streptomyces albulus*. Positively charged ε-PL molecules generally inhibit the proliferation of microorganisms including yeasts, fungi, and gram-positive or gram-negative bacterial species [28]. Recently, ε-poly-L-lysine was incorporated into wool using mTGase and the result showed that ε-poly-L-lysine endued the wool with better antibacterial properties [29]. Similarly, promising antimicrobial rates of 96.98% and 97.93% against *Escherichia coli* and *Micrococcus luteus*, respectively in wool grafted with ε-polylysine was found [30].

2.10. Environmentally Friend

The textile industries are suffering from the discharge of high organic load pollutants to water bodies or soil. Now days, the sector is forced to increase the cost for waste treatment. However, enzymatic applications in different stages of textile processing decrease the organic load of the wastes so that waste treatment related costs could be minimized. For instance, the chemical oxygen demand of waste water released from wool industry that used mTGase treatment was half of resin treated one [18]. Similarly, the COD of treating bath with mTGase was only one third of that of the traditional resin treatment [19]. Sericin, usually degummed in silk processing, substantially increase organic discharge [2] and it has been recovered by ultrafiltration for effective application in cosmetic formulation due to their moisturizing effect. However, the sericin pollutant has been incorporated into wool so that, in first hand, the organic load is controlled, in the other hand, it results good wool properties in terms of moisture absorption, antistatic properties, softness, and comfort [2].

2.11. Transglutaminase Limitations in Wool

TGase is industrially very important enzyme in wool fabrication since it recovers wool damaged by chemicals and protease. In addition, it helps to incorporate amines, proteins, and antimicrobials to wool to bring desired properties in wool. However, the amount of lysine and glutamine residues available may be limited, especially on the surface of wool fibre and this restrict the extent of enzyme reaction. Therefore, this may affect the wide application of TGases on wool [31].

3. Transglutaminase Application in Silk Industry

The wearing properties of silk fibers can be seriously affected by silk fiber tendency to cockle and deform. In fact, such defect is determined by the micro structure of the silk fiber. Instead of being linked by relatively strong chemical bonds, there are only weak hydrogen bonds and Van der Waals force exists in the intro- and inter- silk fibroin molecules, which can be destroyed easily [32]. Especially when they are driven by some outside force, fibroin molecules will slip reciprocally and may well form new links in the new location. Therefore, silk fabric has a poor elastic recovery. This problem has been solved by treating silk with TGase.

Both solo mTGase treatment and treatment with mTGase followed by hydrogen peroxide, protease and ultrasonic exhibited that mTGase can improve the crease resistance of silk fabric. It also enhanced its tensile breaking strength or amended damage in the tensile breaking strength caused by pretreatments. It was found that combined treatment of mTGase followed by ultrasonic exerted a better coordinated effect and conferred better performances compared to other treatments [33]. This was responsible for increment of the wrinkle recovery angle by 17.4% and improvement of tensile breaking strength by 11.2% respectively. Like wool fibers, both hydrogen peroxide and protease can demolish the structure of silk fiber to a certain degree and this is the main target mTGase crosslinking. The result may suggest that certain pretreatment will possibly open up the fiber structure and thus, increases accessibility and the catalysis of mTGase. In another study, tensile strength, elongation rate at breakage and thermal stability of the silk samples were remarkably improved by using transglutaminase-mediated polymerization to restore thermal aged silk [34].

4. Leather Industry

Filling is one of essential steps in leather processing and it is defined as the introduction of materials into the voids that exist between the fibres of the leather to smoothen any veiny or other irregularities on the leather surface. Filling materials often remain in place during the later processing steps. The most commonly natural products used as fillers are glucose, flour and gum. In addition, gelatin and casein could also be used effectively as filling materials [35]. In these investigations, glutaraldehyde was grafted with gelatins and these resulted highly polymerized gelatines that were able to fill the leather. In addition, they remained bound to the leather during washing steps. Treatment of casein with transglutaminase gave similar results, suggesting that with mTGase treatment, inexpensive proteinaceous industrial byproducts could replace the more expensive filling materials currently used [36].

5. Conclusions and Future Directions

TGase is a potential enzyme in textile and leather development. Recovering wool and silk damages caused by chemicals and enzymes, enhanced shrink resistance, smoothed wool, better wool wettability, and good color fastness are attributes of TGase treatment in textile industry. In wool processing, it is applied commonly after protease treatment. Although protease badly affects wool properties in different ways, it enhances the TGase action [9], probably by making sites free for TGase action. Here the mechanism is not clearly illustrated and this is open for study. The enzyme is also used to fill holes in leather production. The lysine and glutamine available in wool may be limited to expand the TGase application [31] and this area requires further investigation to know exactly the limitations and find out solution for it. Furthermore, the waste treatment cost that can be reduced when TGase is applied is not well documented to know exactly the economical benefit that can be obtained in this direction and this is also an attractive research area open for investigators.

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