

## Laccase-mediated dye-free coloration of wool fabric

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In this study, an investigation on a novel coloring technique, based on laccase-mediated radical coupling of amino acid molecules of wool fibres, has been carried out. Firstly, the influence of temperature, incubation time, and pH on the *K/S* value and hue angle (*h*) of the colored wool fabrics is studied. Analysis of levelness of coloration, wash fastness, rub fastness, and UV-protection factor of the colored wool fabrics has been done. Then, the surface morphology and structure of the wool fibres are analyzed by scanning electron microscopy (SEM) and Fourier transform Infrared spectroscopy (FTIR). The enzymatic coloration processes are carried out in an acetate-sodium acetate buffer medium (pH 5) at 50 °C for 24 h and the colored wool fabrics show good color fastness and uniformity. The results obtained are as per the requirements of textile color fastness. SEM study shows that no particles are adhered to the surface of the wool fibres. The results of FTIR and ultraviolet-visible spectroscopy show that the obvious oxidation coupling reactions take place between the molecules in the polypeptide chains of the wool.

**Keywords:** Dyeing, Enzymatic colouration, Laccase, UV-protection, Wool

### 1 Introduction

Generally chemical dyes, such as reactive dyes, acid dyes, disperse dyes, and cationic dyes, are conventionally used for dyeing of various textiles<sup>2</sup>. However, these dyes cause severe damage to the environment due to large amounts of wastewater generated from these traditional dyeing processes<sup>3,4</sup>. Recently, there has been a special emphasis on the use of environment-friendly dyeing processes due to customers' awareness for environmental conservation. For instance, natural dyes have replaced the traditional synthetic dyes for dyeing and finishing of textiles, since they are dependent on renewable resources<sup>5,6</sup>. However, the dyeing techniques using natural dyes still has some shortcomings, such as poor reproducibility, lower stability of the dyes, and low dyeing fastness. Additionally, the use of metal mordant in natural dyes also leads to environmental pollution in the dyeing process.

In recent years, many researchers exploited the ability of laccase to catalyze oxidation for applications in the field of textile coloration<sup>7</sup>. Laccase from the ascomycete, *Myceliophthora thermophila*, is able to oxidize phenolic compounds such as catechol and catechin and mediate their attachment to denim

surfaces. Laccase-mediated polymerizations gave rise to new coloration states from dark brown to green-yellow<sup>8</sup>. Cotton cellulose is dyed *in situ* with a polymeric dye generated by oxidative coupling of colorless catechol, using laccase catalyst<sup>9</sup>. Laccase is able to polymerize flavonoids, resulting in a strongly colored polymeric solution which can be applied to the coloration of flax fabrics<sup>10</sup>.

As a new concept of textile dyeing, the biological coloration technique makes textiles obtain certain colors through enzymatic oxidation. This new method has the advantage of avoiding the use of synthetic dyes in the industrial dyeing of fabrics, which are harmful pollutants and also help in cutting the use of upstream raw materials for preparing synthetic dyes. However, exogenous substances as colorants must be used in the above laccase-mediated coloration methods. Regarding the dye-free dyeing technique, some processes for wool and silk colorations have been reported. For instance, dyeing of tussah silk using p-hydroxy benzaldehyde is studied without adding any other dyes by Liu *et al.*<sup>11,12</sup>. A benzaldehyde derivative is used to carry out the dye-free process for wool or silk under acidic conditions and is very successful. In these studies, a chemical reagent is used as the color-developing agent<sup>13</sup>.

However, till date, enzymes are rarely used in the dye-free colorations of fabrics. Laccase is a

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phenoloxidase, which mainly plays an important role in the formation of the liquid paint varnish (raw lacquer), and is present in fungi that catalyzes the transformation of a large number of phenolic and non-phenolic aromatic compounds<sup>14-16</sup>.

Wool is one of the main raw materials used in textile industry which contains about 19 amino acids. Amongst them, tyrosine and tryptophan are suitable substrates of laccase<sup>17</sup>. In this paper, a new laccase-mediated dye-free coloration concept of wool fabrics is presented. Based on catalytic oxidation of tyrosine and tryptophan in wool structure by laccase, coloration of wool fabrics is achieved by oxidation coupling reactions in the polypeptide chains, without dyeing them.

## 2 Materials and Methods

### 2.1 Materials and Reagents

Laccase from *Trametes versicolor* (46.6 U/g) were obtained from Novozymes. 100 % wool fabrics (220 g/m<sup>2</sup>, 2/1 twill, 32s, 410 ends/10 cm×250 picks/10 cm) are obtained from Jiangsu Xiexin Wool Textile Co. Ltd. (Wuxi, China). All other chemicals, obtained commercially, were of analytical purity.

### 2.2 Enzymatic Dyeing Process of Wool Fabric

The wool fabrics were pretreated with distilled water for 10 min in order to improve wettability. The fabric was dyed in 50 mL 0.1 M acetate buffer (pH 3-7), containing laccase. The reaction was carried out at 30-70 °C for 1-24 h in a shaking bath. Considering the color of laccase itself, the wool and cotton fabrics were respectively treated with inactivated laccase and laccase. After the completion of reaction, the fabric was washed first with 0.1 M acetate buffer and then with distilled water at 50 °C.

### 2.3 Color Measurement

The color feature is tested using a 650 datacolor (datacolor, USA). The wavelength is in the range of 360-740 nm.

### 2.4 UV-Vis Spectrophotometric Analysis

The samples are analyzed for their colorations using a UV-2550 UV-VIS spectrophotometer (Shimadzu, Japan) with BaSO<sub>4</sub> as the reference. The wavelengths are recorded in the range of 200 - 800 nm.

### 2.5 Determination of Levelness of Coloration

The K/S values are measured using 650 datacolor (datacolor, USA) at eight different points on the dyed wool fabric to determine the levelness of coloration in the fabrics by calculating the mean of K/S value and

relative standard deviation  $S_r(\lambda)$ . The smaller the deviation  $S_r(\lambda)$ , the more is the evenness<sup>18</sup>.

### 2.6 SEM Analysis

The wool fabric samples are scanned using a SU1510 scanning electron microscope (SEM) (Hitachi, Japan) that operated at 5.00 kV with a magnification of × 2.00 k.

### 2.7 FTIR Spectroscopy

The FTIR spectroscopic analysis is carried out on a Nicolet iS10 FTIR spectrometer (Thermo Fisher Scientific, USA). The spectra are recorded in the range of 4000-500 cm<sup>-1</sup> with a resolution of 4 cm<sup>-1</sup> and 16 scans per sample.

### 2.8 Color Fastness Test

The color fastness of the dyed fabrics is evaluated by the test procedures as follows: ISO 105-CO6:1998—Color fastness to domestic and commercial washing (grey scale 1-5); and ISO/DIS 105-X12:2001—Color fastness to rubbing (grey scale 1-5).

### 2.9 UV Protection Factor of Colored Wool Fabric

Ultraviolet transmittance spectra are obtained using a UV-1000F ultraviolet transmittance analyzer (Labsphere, USA). Each spectrum is an average result of 10 measurements.

## 3 Results and Discussion

### 3.1 Process Optimization for Enzymatic Dyeing of Wool Fabric

#### 3.1.1 Effect of Time on Dyeing Process

In order to study in detail, the effect of the reaction time on coloration process, the coloration times chosen for the process are kept between 1h and 24 h at 50 °C.

Figure 1(a) shows the effect of time on the coloration of wool fabrics. It is apparent that the highest K/S value is obtained, when the fabric is incubated with laccase for 8 h. Beyond this time, the change in K/S value with increase in reaction time is reduced. The results also demonstrate the laccase catalyzed oxidation of certain amino acids during the coloration of wool fabrics<sup>19,20</sup>. Oxidation of phenolic hydroxyl group of tyrosine and indole ring of tryptophan produces free radicals that initiates the coupling reaction. Moreover, enzymatic oxidation usually results in coloring of the substance, due to formation of long conjugated structures along the polypeptide chains in the wool.

The number of conjugated bonds increases as the degree of oxidation is increased. This means

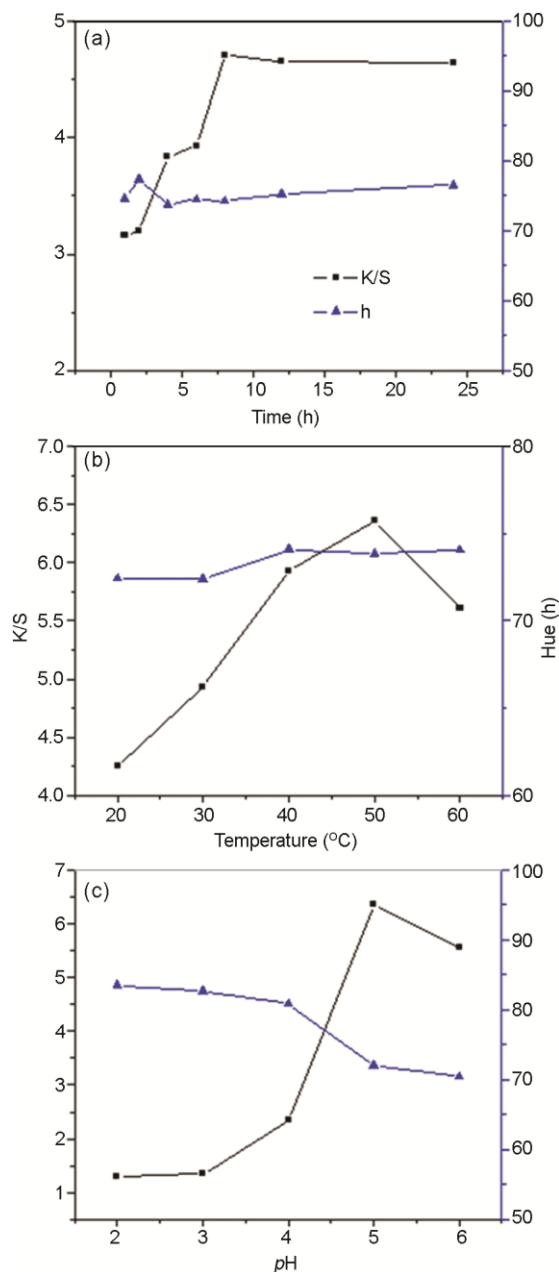


Fig. 1 — Effect of time, temperature and  $pH$  on coloration of wool fabrics with laccase

that a higher degree of oxidation shows a deeper coloration<sup>21,22</sup>. However, wool fabric has an essentially fixed total number of each amino acid and so oxidative coupling reactions reach a saturation point after a certain time.

As shown in Fig. 1(a), the change in hue angle  $h$  is less as the incubation time progresses. This indicates that the enzymatic time has little effect on the coloration of wool fabric. This can be explained based on the fact that the degree of oxidation of wool fibre

increases with time; however, there is no change in the mechanism of oxidation. Therefore, the hue of the color in the wool fabric does not change as the reaction progresses.

### 3.1.2 Effect of Temperature on Dyeing

Temperature is the primary factor influencing the efficiency of catalytic oxidation using laccase. Considering the thermal stability of laccase, a preliminary study has been performed to evaluate catalytic oxidation of laccase at different temperatures.

Figure 1(b) shows the results of coloration of wool fabrics at different temperatures. By increasing the reaction temperature a deeper coloration of the samples could be obtained, keeping the enzyme concentration constant, due to the activation of microbial enzyme at high temperature. It is found that the highest  $K/S$  is reached when the coloring of the woolen fabric with laccase is carried out at 50°C. Beyond this temperature, the  $K/S$  decreases with the increase in temperature. This indicates that the most suitable temperature of laccase activity is 50°C. Laccase shows good performance in the range of high temperature, but at excessively high or low temperatures it loses its activity due to denaturation. When the temperature reaches 50 °C, laccase has much higher activity. Thus, the coloration of wool fabric attains its highest level of effectiveness.

The effect of temperature on the hue angle ( $h$ ) is consistent with reaction time. Temperature has greater influence on the laccase activity without impacting the mechanism of catalytic oxidation.

### 3.1.3 Effect of $pH$ on Dyeing

Figure 1(c) shows the results of coloration of wool fabrics at different  $pH$  values. It is found that the highest  $K/S$  value is reached when the fabric is treated with laccase at  $pH$  5. With further increase in  $pH$  value, the  $K/S$  value decreases. The results show that the  $pH$  of solution could have a strong impact on the activity. In acidic solution, specifically at  $pH$  2-3, the laccase activity is low. The highest activity is reached when  $pH$  of the solution reaches 5. This is because of the fact that at this  $pH$  oxidative coupling reactions of phenol hydroxyl of tyrosine and indole of tryptophan reaches maximum efficiency.

However, the change in hue angle range displays the little effect of  $pH$  on the coloration of wool fabric. On one hand, characteristic and biological functions of laccase require specific conditions; the catalytic oxidation of laccase on wool fabrics is highly efficient

at  $pH$  5. On the other hand, the types of the amino acids which could be catalyzed and oxidized by laccase are not changed<sup>23,24</sup>. Therefore, the color of the wool fabrics shows little changes.

### 3.2 Evaluation of Levelness Parameters

According to catalog 2.5, the uniformity of the colored wool fabric, treated with laccase is computed and  $S_r(\lambda)$  is 0.0488. Lower numbers indicate a better leveling performance, so the color uniformity of the wool fabric treated by laccase is very good. The dye-free coloration of wool fabrics is different from the traditional dyeing method, since in the dye-free coloration technique, tyrosine residues of wool fabrics are catalytically oxidized to form conjugate structure and then display color. As the tyrosine residues are evenly distributed in the structures of the woolen fabrics, the uniformity in coloration is excellent<sup>25</sup>.

### 3.3 Color Fastness Test

Color fastness is very important in textile dyeing, especially the color fastness on rubbing and washing. The results show that the colored woollen fabrics has 4-5 grade of dry and wet rub fastness and 4-5 grade of wash fastness.

This is attributed to the formation of chromophores from the peptides by the coupling reactions of tyrosine residues, present on different peptides. The peptide links are immobilized in the woolen structure by covalent bonds. However, in the traditional dyeing of wool fibre with acid dyes, the binding force of dyes and fibres are mostly ionic bonds, van der Waals forces and hydrogen bonding. Therefore, the firm covalent structure makes colored substance stable<sup>26</sup>.

### 3.4 Effect Contrast of Wool fabric with Inactivated Laccase and Laccase

In order to eliminate interruptions in the color of laccase itself on wool fabric, the wool and cotton fabrics are respectively treated with inactivated laccase and laccase. The  $K/S$  values of wool and cotton fabrics are listed in Table 1. The wool fabric treated by laccase shows yellow with a little red light, but the color of wool fabric treated by inactivated laccase is close to the untreated wool fabric. Because inactivated laccase has the same color with laccase, the contrasting results show that the coloration of

Table 1 —  $K/S$  of wool fabric with inactivated laccase and laccase

| Sample        | Untreated | Treated with laccase | Treated with inactivated laccase |
|---------------|-----------|----------------------|----------------------------------|
| Wool fabric   | 1.109     | 7.468                | 1.141                            |
| Cotton fabric | 0.164     | 0.172                | 0.169                            |

wool fabric is independent of the color of laccase itself. Cotton fabric has no groups which can be catalyzed by laccase. The cotton fabrics with laccase and inactivated laccase also show no coloration. This can explain that the coloration of wool fabric has some correlativity with amino acid catalyzed by laccase.

### 3.5 UV-Protection Factor of Colored Wool Fabric

Over the past years, there has been a growing need for using textile products, which provide effective protection against UV radiation, which is responsible for skin damages such as skin cancer, sunburn, and photo-aging. In case of wool, although it is endowed with a good inherent UV protection property, a serious consequence of photo-radiation, that is photo-yellowing, still exists<sup>27</sup>. This is probably due to tryptophan and tyrosine of wool absorbing light energy to form a colored substance by photo-oxidation. The coloration of wool fabric catalyzed by laccase shows not only coloration but also improved the UV resistance of wool fabric.

Figure 2 shows resistance of uncolored and colored woolen fabrics to ultraviolet radiations. In 280 - 380 nm range, the ultraviolet transmittance of uncolored wool fabric increases gradually, while the UV transmittance ranging from 280 nm to 370 nm of colored wool fabric shows a straight line and the values are close to zero. It proves that the colored wool fabric has good ultraviolet resistance.

Colored and uncolored wool fabrics show different UV protection with UPF of 100+ and 50+ (Table 2). The reason could be that the colored wool fabric

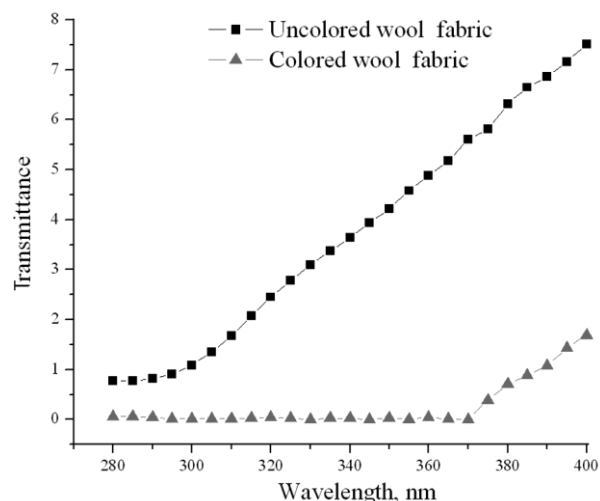


Fig. 2 — UV-protection of colored wool and uncolored wool fabrics

has a more conjugated structure which could absorb UV light as compared to uncolored wool fabric<sup>28,29</sup>.

### 3.6 SEM Analysis

To demonstrate the effect of laccase treatment on the wool surface, wool fabric is washed with distilled water at room temperature. The surfaces of uncolored and colored wool fabrics are studied by SEM. As seen in Fig. 3, the surfaces of the wool fibres show slight changes after the enzymatic process. The squama of colored wool fibre remains intact and also color coatings and sediments are not observed on the fibre surfaces. This suggests that the coloration of the wool fabric is not caused by pigment deposition on woolen fabrics, but it is a result of formation of conjugated structures inside the woolen fibre.

### 3.7 Ultraviolet-visible Spectral Analysis

For comparison purposes, ultraviolet-visible spectra of both uncolored wool and colored woolen fabrics are tested. The ultraviolet absorption spectrum of the woolen fabrics is shown in Fig. 4.

Table 2 — Parameters related to the UPF of wool fabric samples

| Sample                | Transmittance |      | UPF              |
|-----------------------|---------------|------|------------------|
|                       | UVA           | UVB  |                  |
| Uncolored wool fabric | 4.78          | 1.18 | 50 <sup>+</sup>  |
| Colored wool fabric   | 0.35          | 0.02 | 100 <sup>+</sup> |

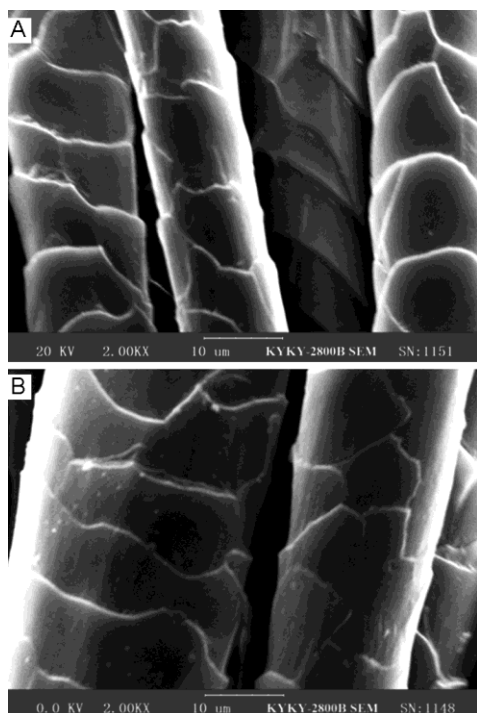


Fig. 3 — SEM analysis of wool fabrics (A) uncolored wool, (B) colored wool

A significant absorption band appears at 285 nm due to the presence of a phenolic group<sup>30</sup>. The samples of untreated wool fabric show bands between 219 nm and 243 nm due to  $\pi$ - $\pi^*$  transitions of the aromatic groups, present in the peptide chains of wool. The absorption peaks at 219 nm and 243 nm could be assigned to C=C of the benzene ring. After 24 h of incubation with laccase, the wool fabric shows a shift of the band from 285 nm to 287 nm. Meanwhile, the band at 219 nm disappears and the intensity of the peak at 243 nm decreases. This phenomenon is probably due to the oxidation coupling of tyrosine and tryptophan residues of wool fabric, which results in increased conjugation<sup>31</sup>.

In the macromolecular polypeptide chain of wool, hydrogen atoms of the phenol group of tyrosine and the amino group of indole moiety of tryptophan form free radicals which are active and labile. This results in an intercoupling reaction between macromolecular peptide radicals to form new conjugated bonds. Consequently, dye-free coloration of the protein fibre by laccase-mediated coupling reaction is achieved under mild conditions.

As seen in Fig. 4, an increased area of absorption band after laccase oxidation is due to the dark coloring effect of the woolen fabric by enzymatic oxidation. In the range of 350-500 nm, the colored woolen fabric shows absorption, whereas the uncolored woolen fabric has little absorption. Based on the literature<sup>32</sup>, this is attributed to the changes occurring in the peptide chain structures of wool. Covalent coupling reactions between the different peptides result in the formation of more conjugated

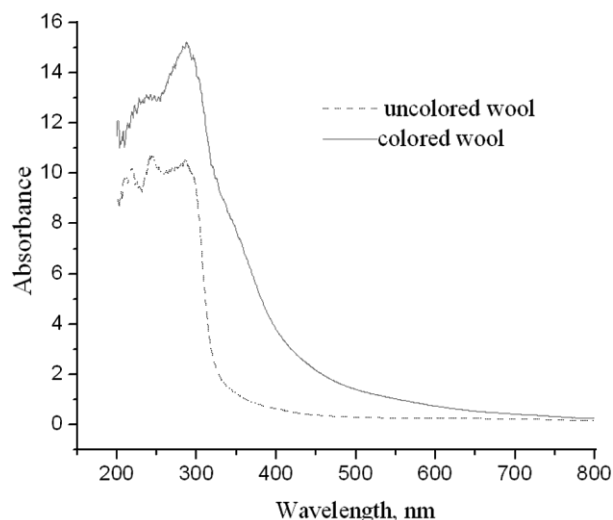


Fig. 4 — UV-visible spectra of wool fabric incubated with laccase

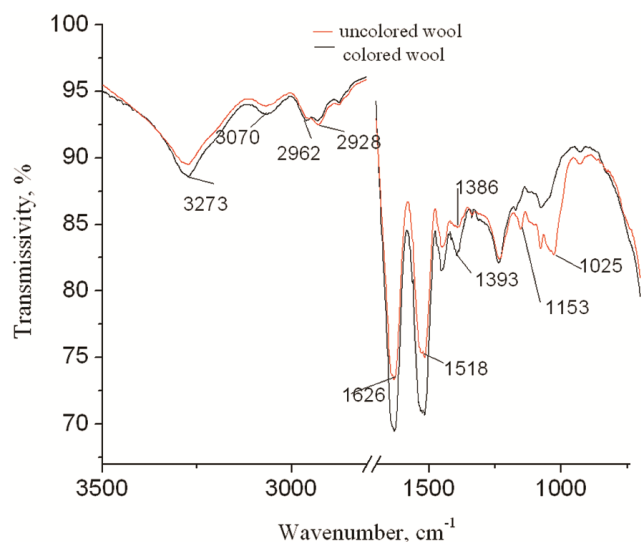


Fig. 5 — FTIR spectra of uncolored and colored wool fabrics

linkages, which, in turn, results in the coloration of woolen fabrics<sup>32</sup>.

### 3.8 FTIR Spectroscopic Analysis

The enzymatic coloration of woolen fabrics is studied using FTIR spectroscopy. The FTIR spectra in Fig. 5 shows subtle differences in the peak at 3273  $\text{cm}^{-1}$ , which is due to the hydroxyl and amino groups present in the wool structure, between colored and uncolored wool fabrics. The absorption peaks at 3070  $\text{cm}^{-1}$ , 2962  $\text{cm}^{-1}$ , and 2928  $\text{cm}^{-1}$  could be assigned to C-H stretching vibrations of the benzene ring and the saturated C-H bonds<sup>33,34</sup>. The absorption peaks at 1626  $\text{cm}^{-1}$ , 1518  $\text{cm}^{-1}$ , and 1078  $\text{cm}^{-1}$  could be attributed to N-H, C-H flexural vibration, and S-O stretching vibration, respectively<sup>35</sup>. However, the FTIR spectra in Fig. 5 shows significant differences in the peaks for colored and uncolored wool fabric at 1393  $\text{cm}^{-1}$  and 1386  $\text{cm}^{-1}$  which are as a result of C-H bending vibrations. The peak intensities decrease and result in a blue shift which might be attributed to the coupling reactions between the amino acid residues in wool.

In addition to this, Fig. 5 also shows a significant difference between the peaks for colored and uncolored wool fabric at 1153  $\text{cm}^{-1}$  and 1025  $\text{cm}^{-1}$ , due to the ether bonds in the structure of wool. These ether bonds are formed due to the coupling reactions between the tyrosine residues in wool generating Ph-O-Ph products<sup>36,37</sup>.

All of the above analytical results indicate the formation of Ph-Ph and Ph-O-Ph linkages, due to the inter-coupling of macromolecular radicals of wool via

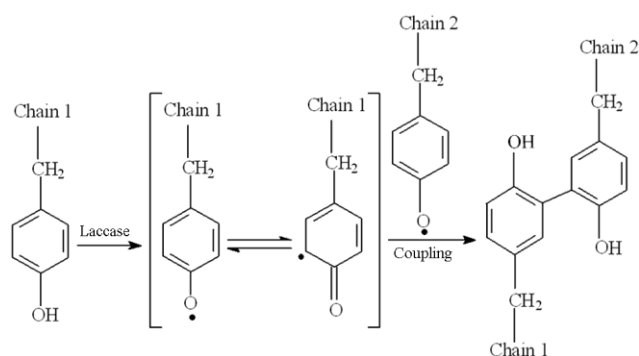


Fig. 6 — Proposed coupling linkages in the peptide chains of wool

laccase treatment. However, the coloration of wool fabric using the dye-free approach mainly forms the Ph-Ph linkages. Proposed pathways for the reaction mechanism of amino acid residues are represented in Fig 6.

## 4 Conclusion

**4.1** The dye-free coloration of wool fabrics has been brought about by laccase-mediated oxidation in acetate-sodium acetate buffer solution. Optimization studies indicate that the enzymatic coloration process is highly efficient in a pH 5 (acetate-sodium acetate buffer solution) at 50°C and a treatment period of 24 h. Ultraviolet-visible spectra of uncolored and colored wool fabrics prove that laccase is able to catalyze the oxidative coupling of tryptophan and tyrosine residues of wool fabrics.

**4.2** The covalent coupling between macromolecular chains of wool through enzymatic oxidation, mediated by laccase, is confirmed by an increase in the intensities of the peaks due to carbon hydrogen bonds and ether linkages in the FTIR spectra of the colored fabric. Surface morphologies of uncolored and colored wool fibre are studied by SEM.

**4.3** Analysis of levelness and color fastness shows that the coloration of wool fabric, catalyzed by laccase, is good and stable. Results of UV-protection experiments show that the colored wool fabric has excellent anti-ultraviolet properties. Laccase-mediated dyeing opens up new strategies for dye-free coloration.

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