Nanocellulose Extracted from Waste Polyester/Cotton Fabric by Chemical-Mechanical Separation Technology

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Abstract. The millions of tons of textile waste were produced every year. Most of them were made from polyester/cotton blended fabrics. The key technologies for recycling the waste blended fabrics were how to separate the blended components and then use the materials from the waste. Based on this, this research aimed to develop the technology of sustainable recycling of cotton fiber and polyester. The waste polyester/cotton blended fabric was first cut into small pieces, then treated with acid, and then used a grinder to crush it to separate the polyester from the cotton fiber. After the separation process, the cotton fiber was powder and the polyester fiber was fibrous mass. The components of the separated cotton samples were studied by Fourier transform infrared spectroscopy. The crystallinity of separated cotton fiber was analyzed by X-ray diffraction. The morphology of the separated cotton fiber and polyester were analyzed by scanning electron microscope. The particle size and potential of the cotton fiber after final treatment were analyzed by using the nanoparticle size and zeta potential analyzer. The results showed that the waste polyester/cotton blended fabric was successfully separated and the separated cotton fiber was successfully prepared into cotton nanocellulose for future use.

1. Introduction

The textile and garment industry is very important to the economy of every country and played a vital role in trade, investment, employment, and income. At the beginning of the 21st century, the clothing industry performed well, the cost decreased, the operation simplified, and consumer spending increased, which led to the substantial growth of clothing production from 2000 to 2014. As a result, a large number of waste textiles have been produced. So, recycling of waste textiles has become a major problem that needs to be solved now. There are many disposal methods for waste textiles. Currently, incineration and landfill are the main methods, which has a great impact on the environment[1]. Besides, it can be recycled. There are many ways to recycle waste textiles, including physical methods and chemical methods. However, cellulose extraction from waste textiles has great advantages, which has become a hot research topic all over the world in recent years. Cellulose is often regarded by scholars as one of the most important natural resources, which has excellent properties of degradation and regeneration[2, 3]. There are many sources of cellulose, such as wood, natural fiber (agricultural biomass), and algae[4, 5]. By using different physical, chemical, and physicochemical processes, the components of waste textiles are separated and cellulose is extracted. In recent ten years, the preparation of nanocellulose from renewable
and abundant existing cellulose has attracted the attention of researchers because of its unique properties, such as unique chemical properties, optical properties, as well as large specific surface area, high crystallinity, and high Young's modulus\cite{6-9}.

A large proportion of waste textiles are polyester and cotton blended fabrics, and cotton can even account for 35~40\%\cite{10-12}. At present, there are many methods for separating and recovering polyester/cotton blended fabrics\cite{13}. Some studies used cellulase to dissolve and remove cotton fibers to obtain lightweight polyester fabrics, but the concentration of cellulase required by this method was higher\cite{14}. It is also possible to separate polyester/cotton blends using ionic liquids, but this system has other disadvantages, such as low separation efficiency and high cost\cite{15}. There are many research papers reported the technology of cellulose hydrolysis by strong acid and alkali, but it needs to consume a lot of chemicals, and they are corrosive to equipment and so on\cite{16-20}.

The purpose of this study was to separate the polyester/cotton blended fabric with a highly efficient and environmentally friendly method. The separated products were polyester fiber and cotton fiber, and nanocellulose from the cotton was obtained by further processing. The extracted materials were characterized by scanning electron microscopy (SEM), X-ray diffraction (XRD), Fourier transform infrared spectroscopy (FTIR), nano-particle size, and zeta potential analysis. It could be shown that this method not only had a short separation time but also had a better separation effect. Moreover, the particle size of cotton fiber-based nanocellulose was small, which had more advantages for future applications.

2. Experimental

2.1. Materials and Reagents

Commercially available polyester-cotton blended fabric was used as the source material. The reagents used were sodium hydroxide (NaOH), 98\% sulfuric acid (H\textsubscript{2}SO\textsubscript{4}), all of which were analytical grade, purchased from Tianjin Kailisi Fine Chemical Co., LTD. All the solutions needed in the experiment were prepared with deionized water made in the laboratory.

2.2. Preparation method

Sulfuric acid treatment: firstly, the waste polyester/cotton blended fabric was cut into small pieces with a size of 2×2 cm, and treated with a different concentration of sulfuric acid for different times. The treated polyester/cotton blended fabric was washed to neutral and dried. The cotton fiber powder and the polyester fiber ball stained with the cotton fiber powder were obtained by grinding with a grinder. After that, a certain amount of cotton fiber powder was weighed and swelled with NaOH solution, and then the pH value was adjusted to neutral. Then, 64\% of sulfuric acid was used for a certain time, and 10 times of deionized water was added to finish the reaction. The diluted solution was centrifuged and dialyzed to neutral. The treated solution was homogenized with a high-pressure homogenizer for a certain number of times, and its concentration was mixed to 0.5\%, and then freeze-dried.

2.3. Characterization

2.3.1. Fourier transform infrared (FTIR) spectroscopy. The FTIR was tested with a spectrometer (Nicolet iS50, Seymour Fisher Technologies) and the analysis range was 4000~400 cm\textsuperscript{-1}. After KBr drying, grinding, and granulating, the infrared spectrum of the samples was measured to clarify the changes of functional groups.

2.3.2. X-ray diffraction (XRD). The crystallization index of the sample was measured using a Bruker D8 Advance X-ray Diffractometer. The parameters were D8 advanced powder X-ray Cu-K\textalpha\ radiation diffractometer with a wavelength of 0.15406 nm, voltage of 40 kV, and a current of 40 mA. The diffraction angle range was 2θ =10~80° and the scanning speed was 5 °/min. The crystallinity index (CrI) was calculated by the formula: CrI = [I(002)-I(am)] / I (002) * 100.Where I (002) referred to the peak strength of the crystalline materials; I(AM) referred to the peak strength of the amorphous materials.
2.3.3. **Scanning electron microscope (SEM).** After the sample was sprayed with gold, a 10 kV scanning electron microscope (SEM, Hitachi S4800, Japan) was used to characterize the morphology of the sample.

2.3.4. **Nanoparticle size and Zeta potential.** The particle size distribution and potential of the samples were measured with a nanometer size and zeta potential analyzer (Marvin ZS90).

3. **Results and discussion**

3.1. **Fourier transform infrared spectroscopy (FTIR)**

The Fourier infrared spectra of the cotton fibers separated were shown in Figure 1.

![Figure 1](image-url)

**Figure 1.** The infrared spectrum of the separated cotton fiber. The e was white cotton fiber, and the treatment time was 40min; a~d was red cotton fiber, and the treatment time was 20min, 40min, 40min, 60min (c was 30% sulfuric acid treatment, the rest was 10% sulfuric acid treatment)

Figure 1 showed two major absorbance regions, 2700 cm$^{-1}$~3500 cm$^{-1}$ and 500 cm$^{-1}$~1800 cm$^{-1}$, respectively. The absorption peak near 3335 cm$^{-1}$ was the stretching vibration of O-H in cellulose structure$^{[21]}$. The stretching vibration of C-H was near 2902 cm$^{-1}$ because CH$_2$ groups were present in the glucosyl side ring in cellulose$^{[22]}$. The absorption peaks related to lignin or hemicellulose range from 1720 cm$^{-1}$ to 1733 cm$^{-1}$, which was the stretching vibration of carbonyl C=O$^{[23]}$. In the treated samples, the peak was weakened, and even disappeared in some samples. It showed that lignin or hemicellulose in the sample had been removed after treatment. The C-H bending vibration at the wavenumber of 1431 cm$^{-1}$ and the C-O-C absorption band with the wavenumber of 1158 cm$^{-1}$ were cellulose structures$^{[24]}$. The absorption band around 1055 cm$^{-1}$ was the carbon-oxygen pyranose ring$^{[21]}$. It could be proved that the separated cotton fiber was still a cellulose structure.

3.2. **X-ray diffraction (XRD)**

The cotton fibers separated were measured by XRD, as shown in Figure 2.
Figure 2 (2θ =14.75° and 16.39°) showed additional bimodal signals that were generated, representing amorphous materials. A high-intensity diffraction peak was observed at about 2θ = 22.69 °, indicating that the crystallinity of cellulose fiber was very high, and it was the structure of cellulose type I. The results showed that this method would not change the crystal structure of the cotton fiber, and the results were consistent with the results of infrared analysis [25-27]. After calculation, the crystallinity of red polyester/cotton blended fabric was 79.18 %, 79.39 %, 75.84 %, and 76.43 % respectively, and the crystallinity of white polyester/cotton blended fabric was 76.67. The treated cotton fibers had an increased crystallinity compared to raw and other fibers, such as straw and wood fibers [27]. This was because lignin and hemicellulose had been removed [28].

3.3. Scanning electron microscope (SEM)
The scanning electron microscope (SEM) was used to observe and analyze the morphology of the separated polyester and cotton fibers and freeze-dried aerogels. The results were shown in Figure 3.

Figure 2. XRD patterns of the separated cotton fibers. The a was white cotton fiber, the treatment time was 40min; b-e was red cotton fiber, the treatment time was 20min, 40min, 60min, 40min(d was 30% sulfuric acid treatment, the rest was 10% sulfuric acid treatment)

Figure 3. Morphology of cotton fiber and polyester fiber after separation. The processing time of a~d diagram was 20min, 40min, 60min, 40min (d diagram was 30% sulfuric acid treatment, the rest was 10% sulfuric acid treatment), e and f diagram were aerogel

It could be observed from figure 3 that the cotton fiber after acid hydrolysis and mechanical agitation was fragmentary, with a significantly reduced length, cracks, and surface damage. The higher the acid concentration and the longer the treatment time, the more serious the damage. However, the recovery
rate should be considered, so it should be controlled at a certain time and concentration. The treated polyester fiber was stained with some cotton fiber powder, the fiber was still continuous, and the surface was not damaged. The aerogels obtained after freeze-drying had a cross-linked and dense pore structure.

3.4. Nanoparticle size and Zeta potential

The nanometer size and zeta potential of the cotton fiber solution treated in this experiment were measured by nanoparticle size and zeta potential meter. The dispersant was water and each sample was measured three times in parallel. The nanoparticle size was shown in Figure 4A. The a–d represented unhomogenized, homogeneous for 5 times, 10 times, and 15 times. The measurement results showed that most of the particle sizes after homogenization were in the nanoscale range. Moreover, in this experiment, the particle size distribution of the samples with more homogenization times was more concentrated than that of the samples with fewer homogenization times. The particle size distribution range of treated cotton fiber was wide, ranging from 40 nm to 400 nm, centrally diameter distributed in 40–100 nm.

Figure 4. A was the Nano particle size, B was Zeta potential.

The stability of suspension can be characterized by the magnitude of zeta potential. When zeta potential was relatively high, the repulsive force between particles was greater than the attractive force, and particles were more inclined to disperse. Therefore, the suspension was in a relatively stable state. When zeta potential was low, the attraction between particles was larger than the repulsion force. At this time, the particles tended to agglomerate, so the system was unstable, and the stability of the system was destroyed. In general, when the zeta potential was less than -15 mV, it indicated that agglomeration began to occur in the suspension system. When the zeta potential was greater than -30 mV, it indicated that the repulsive force of the particles in the solution was relatively strong, which was colloidal and uniformly stable[29]. The a–c of Figure 4B were homogenized for 5 times, 10 times, and 15 times respectively. The zeta potential of the treated cotton fiber solution was between -14 mV and -16 mV. The more homogenization times, the greater the absolute value of potential and the better the dispersion of the solution. Therefore, the repulsion force of particles in the solution was relatively large. However, with the increase of homogenization times, the solution gradually agglomerates.

4. Conclusion

In this study, polyester/cotton blended fabric was successfully separated, and nanocellulose was extracted from the separated cotton fiber. The separated polyester fiber and cotton fiber were characterized by Fourier transform infrared spectroscopy (FTIR), X-ray diffraction (XRD), and scanning electron microscope (SEM). The results showed that the separated cotton fibers still had
cellulose structure. The crystallinity of the separated cotton fiber was high, reaching 79.39 %. The existence of nanocellulose was proved by nanoparticle size analysis, and within a certain homogeneous range, the more homogeneous the number of times, the smaller the particle size. Zeta potential analysis showed that the dispersion effect of the solution did not reach the ideal state, so we could study how to make the dispersion effect better. Nanocellulose has many important properties and application fields, and the separated polyester fiber can be used in the next step.

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