Recycling of Waste Cotton Fabrics into Regenerated Cellulose Films through Three Solvent Systems: A Comparison Study

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Abstract

Large amounts of textile waste are generated every year and disposed of through landfill or incineration, leading to numerous environmental and social issues. In this work, the dissolution of three typical waste cotton fabrics (t-shirts, bed sheets and jeans) in NaOH/urea aqueous solution, H_2SO_4 aqueous solution, and LiCl/DMAc solution was investigated. Compared to different types of cotton fabrics, the effects of three solvents on the dissolution of fabrics were more obvious, leading to the significant changes in the structure and properties of regenerated cellulose films. Cotton fabrics (about 2-5%) were rapidly dissolved (8 min) in H_2SO_4 and NaOH/urea solvents after acid pretreatment, while the dissolution in LiCl/DMAc solvent did not need any pretreatment, but a lower cellulose concentration (1%), higher dissolution temperature (80 °C), and longer dissolution time (24 h) were required. The films produced from bed sheets in NaOH/urea solution exhibited the highest tensile strength, thermal stability, and water vapor barrier property. It was because of the stronger cellulose chain entanglement and hydrogen bonds induced by the higher cellulose concentration in NaOH/urea solution. Therefore, this work proves the feasibility to recycle waste cotton fabrics into biodegradable cellulose films, which can be potentially used in various food and agricultural applications.

1. Introduction

Due to the increase in world population, fast changing fashion cycles and higher quality of life, large amount of textile waste are generated every year and have become a global issue.[1] It was estimated that only 15-16 % of textile waste were recycled or reused in the United States in 2015, while the rest were commonly landfilled, discarded or incinerated.[2] It has led to multiple environmental and social problems, including increased production of greenhouse gas emission (1.2 billion tonnes of carbon dioxide each year), ground water contamination, hazardous chemical generation, and limited landfilling space especially in cities.[1,3] Therefore, efforts on recycling textile waste have gained attention such as carbonizing textile waste into biochar, extracting cellulose nanocrystals (CNCs), recovering cotton fibers and polyester, and producing ethanol, biogas and cellulose acetate.[3-7] For example, Yousef et al.[4] recovered cotton and polyester fibers from textile waste using a sustainable technology, achieving $1,629/ton economic returns and a 96% recycling rate. Huang and Wang[5] directly extracted CNCs from textile waste and evaluated their application as reinforcing agents of soybean protein isolate films.

Cotton is one of the most utilized fibers in textile industry, which consists of over 90% cellulose.[8] The dissolution and regeneration of cellulose is a promising way to recycle waste cotton fabrics. However, it is difficult to dissolve cellulose in water and common organic solvents, because cellulose molecules are tightly linked by numerous intramolecular and intermolecular hydrogen bonds.[9] In the last few years, several eco-friendly solvents have been reported to dissolve cellulose, such as lithium chloride (LiCl)/N, N-dimethylacetamide (DMAc), N-methylmorpholine N-oxide (NMMO), ionic liquids, sodium hydroxide (NaOH)/urea aqueous solution, and sulfuric acid aqueous solution.[10-15] Among them, sulfuric acid aqueous solution could directly dissolve wood cellulose with high molecular weight at low temperature in 5 min,[9] while NaOH/urea aqueous solvent proposed by Cai and Zhang[14] is able to rapidly dissolve cellulose with molecular weight less than 1.0×10^5 g mol^{-1} when pre-cooled to -12.6 °C by fast dynamic self-assembly among solvent molecules and cellulose macromolecules. LiCl/DMAc is the most popular solvent that is capable of dissolving different types of cellulose without severely degradation, and the obtained cellulosic suspension can be regenerated easily in water.[10] These three solvent systems are relatively more cost-effective and have been widely studied to dissolve cellulose from different sources such as wastepaper, toilet paper, pulp, bamboo and durian rind.[15-18] Nevertheless, their capacities and applications in recycling waste cotton fabrics are seldom reported.

In order to determine a feasible way to recycle waste cotton fabrics, in this work, t-shirts, bed sheets and jeans made of 100% cotton were selected and their dissolution in sulfuric acid aqueous solution, NaOH/urea aqueous solution, and LiCl/DMAc was investigated and compared. The structure and morphology of regenerated cellulose films were characterized by Fourier transform infrared spectroscopy, X-ray diffraction, and scanning electron microscopy. Additionally, the optical transmittance, mechanical property, thermal stability, and water vapor permeability of regenerated cellulose films were studied via ultraviolet-visible spectroscope, tensile test, thermogravimetric analysis and modified cup method.

2. Materials and methods

2.1. Materials

T-shirts and bed sheets (100% cotton) were collected from the secondhand shop in Montreal, Canada, and jeans (100% cotton) were kindly provided by Renaissance (Montreal, QC Canada). The molecular weights of t-shirts, bed sheets and jeans were determined to be 6.94×10^5, 8.40×10^5, and 7.42×10^5 g mol^{-1}, respectively. Sulfuric acid (95.0-98.0%) and sodium hydroxide were purchased from Sigma-Aldrich (MO, USA). Urea, lithium chloride, acetone and N, N-dimethylacetamide were purchased from Fisher Chemical (Ontario, Canada). Distilled water is used unless specific mentioned.

2.2. Cotton fabric dissolution and regeneration

2.2.1. H_2SO_4 aqueous solvent

A grinding machine (KRUPS, Ontario, Canada) was applied to break cotton fabrics until no obvious fabrics were observed, and the pretreatment conditions were optimized in our preliminary experiment. The fabrics (5 g) were pretreated in 250 mL 20% (w/v) H_2SO_4 at 25 °C, where t-shirts were soaked for 48 h, and bed sheets and jeans were soaked for 72 h. They were then washed thoroughly with water and dried in the oven. The desired amounts of fabrics (listed in Table 1) were added into 50 ml 64% (w/v) H_2SO_4 aqueous solutions that were pre-cooled to

-20 °C, and then stirred at 750 rpm for 8 min using IKA® Eurostar 60 digital mixer in ice bath.[9] The obtained solutions were centrifuged at 6500 rpm for 5 min by Centrifuge 5430 (Eppendorf, USA) to remove bubbles. After centrifugation, the transparent and homogenous solutions were immediately poured onto glass plate and immersed in a coagulation bath with 10% (w/v) NaOH for 15 min. The regenerated films were then soaked in water for 24 h and finally dried in air at room temperature. Cellulose films prepared from t-shirts, bed sheets and jeans in H_2SO_4 aqueous solution were coded as T-H, B-H and J-H, respectively.

2.2.2. NaOH/urea aqueous solvent

After grinded into cotton wool, t-shirts (5 g) were soaked in 250 mL 20% (w/v) H_2SO_4 at 25 °C for 6 days, while bed sheets and jeans were soaked for 7 days. The pretreated fabrics (2 g) were added into 7% NaOH / 12% urea aqueous solutions and kept at -20 °C for 12 h. After thawing, 5% (w/w) transparent cellulose solutions were obtained through vigorously stirring at 2000 rpm for 8 min,[14] followed by centrifugation at 6500 rpm for 5 min. Then, the solutions were quickly spread on a glass plate and immersed in a coagulation bath with 5% (w/v) H_2SO_4 for 15 min. After washing with water, the regenerated films were dried in air at room temperature. Cellulose films prepared from t-shirts, bed sheets and jeans in NaOH aqueous solution were coded as T-N, B-N and J-N, respectively.

2.2.3. LiCl/DMAc solvent

Cotton fabrics were grinded into cotton wools, and washed with water, acetone, and finally DMAc for activation to open up the chains. The fabrics (0.25 g) were added in mixed solutions of DMAc (23 g) and LiCl (2 g), and stirred for 6 h at 80 °C and then for 18 h at room temperature.[19] After dissolution, cellulose solutions were centrifuged at 6500 rpm for 5 min and spread on a glass plate, followed by thoroughly water washing. Finally, the regenerated films were dried in air at room temperature. Cellulose films prepared from t-shirts, bed sheets and jeans in LiCl/DMAc were coded as T-D, B-D and J-D, respectively.

2.2.4. The recovery (%) of cotton fabrics in three solvent systems

The recovery rate of cotton fabrics was calculated using a gravimetric method by the equation (1):[5]

$$Recovery\ (\%) = \frac{w_2}{w_1} \times 100 \qquad (1)$$

Where w_1 is the weight of waste cotton fabrics, and w_2 is the weight of dried regenerated cellulose films.

2.2.5. The molecular weight (Mw) of cotton fabrics and regenerated cellulose films

The Mw of original and cellulose films was tested by viscosity. The intrinsic viscosity [η] of the sample in 0.5 M cupriethylenediamine (CED) solution was measured using a viscometer at 25±0.1 °C. The Mw was calculated by the Mark-Houwink equation:[9]

$$[\eta] = 3.72 \times 10^{-2} Mw^{0.77} \tag{2}$$

2.3. Regenerated cellulose film characterization

2.3.1. Fourier transform infrared spectroscopy (FT-IR)

The structures of cotton fabrics and regenerated films were investigated by Nicolet 6700 spectrophotometer (Thermo Fisher Scientific Inc., MA, USA) equipped with an attenuated total reflectance (ATR) accessory. The FT-IR spectra were recorded as the average of 64 scans with 4 cm^{-1} resolution from 400-4000 cm^{-1} at 25 °C, using the empty accessory as background.

2.3.2. X-ray diffraction (XRD)

XRD patterns of cotton fabrics and regenerated films were collected through a Bruker D8 Discovery diffractometer (Bruker, Billerica, MA, USA), using Cu Kα radiation (λ=0.1542 nm) generated at 40 kV and 44 mA with the speed of 2°/min (2θ was from 4° to 45°).

2.3.3. Scanning electron microscopy (SEM)

The surface and cross-section morphology of cotton fabrics and regenerated films were observed by a Hitachi TM1000 SEM (New Jersey, USA) with an acceleration voltage of 4 kV. Samples were sputtered with gold with the thickness of 5 nm by a Leica EM ACE200 low vacuum coater (Ontario, Canada) prior to observation.

2.3.4. Optical transmittance

The optical transmittance of regenerated films at a wavelength of 800 nm (the thickness was around 0.03 mm) was measured by a DU 800 UV/vis spectrophotometer (Beckman Coulter, Brea, CA, USA), using air as background.

2.3.5. Mechanical property

The tensile strength, elongation at break, and Young's modulus of regenerated films were tested on an Instron 5967 universal testing machine (Instron Corp., MA, USA) at the crosshead speed of 5 mm/min under room temperature (50% RH). The initial grip separation distance was set as 20 mm. Five strips were cut from films with the dimension of 6 cm×1 cm (length × width). The thickness of each strip was measured using a digital micrometer with a precision of 1 μm. All the films were vacuum-dried for 24 h before test.

2.3.6. Thermogravimetric analysis (TGA)

TGA of regenerated films was performed using the thermogravimetric analyzer Q50 (TA instruments, DE, USA). Thermograms of samples were collected from 50 °C to 600 °C with a heating rate of 10 °C/min in nitrogen (40 mL/min).[10] Universal analysis 2000 software was used to calculate the first derivatives of thermograms (DTG) and the percentage of weight loss.

2.3.7. Water vapor permeability (WVP)

A modified cup method was performed to measure the WVP of regenerated films according to ASTM E96-92 standard.[20] A dried film was taped on the top of a glass cup containing anhydrous calcium chloride. The sealed glass cup was then placed in a desiccator that contained saturated sodium chloride solution (75% RH). The weight change of the cup was recorded periodically at 25 °C. The WVP (g m^{-1} h^{-1} Pa^{-1}) of films was calculated by the following equation:[20]

$$WVP = \frac{\Delta m \times k}{A \times \Delta T \times \Delta P} \tag{3}$$

Where Δm is the weight change of the cup (g) during time ΔT (h), k is the thickness of each film (m), A is the exposed area of the film (7.85×10^{-5} m^2), and ΔP is the partial water vapor pressure difference between two sides of the film (Pa).

2.4. Statistical analysis

The experimental results were presented as the mean of three batches ± SD (standard deviation). Statistical evaluation was performed by analysis of variance (ANOVA) followed by multiple comparison tests by Duncan's multiple range test. All of analyses were carried out through SPSS statistical software (IBM, New York, NY, USA) with significant differences within samples at $p<0.05$.

3. Results and discussion

3.1. Dissolution of cotton fabrics in three solvent systems

Three solvent systems were used to dissolve cotton fabrics. Due to their different dissolution capacities, chemical pretreatment (sulfuric acid hydrolysis) was applied to reduce the molecular weight and increase the internal surface area of cellulose, resulting in easier and faster dissolution.[21] The dissolution conditions were optimized by changing the hydrolysis time and cotton fabric concentration in our preliminary experiment and are summarized in **Table 1**.

Table 1. Optimized dissolution conditions of cotton fabrics in three solvent systems

Regenerated films	Pretreatments	Fabric concentrations	Dissolution Time	Dissolution Temperature	Recovery rate (%)	Mw (g mol^{-1})
T-H	48 h in 20% (w/v) H_2SO_4	3%	8 min	0 °C	90.41±2.82	8.65×10^3
B-H	72 h in 20% (w/v) H_2SO_4	2%	8 min	0 °C	90.65±2.02	6.06×10^4
J-H	72 h in 20% (w/v) H_2SO_4	2%	8 min	0 °C	82.40±1.43	2.66×10^4
T-N	6 days in 20% (w/v) H_2SO_4	5%	8 min	0 °C	86.61±2.27	9.01×10^4
B-N	7 days in 20% (w/v) H_2SO_4	5%	8 min	0 °C	96.59±0.58	9.10×10^4
J-N	7 days in 20% (w/v) H_2SO_4	5%	8 min	0 °C	93.17±2.49	6.68×10^4
T-D	/	1%	~1 day	80 °C	99.83±0.25	5.73×10^5
B-D	/	1%	~1 day	80 °C	99.09±0.52	5.80×10^5
J-D	/	1%	~1 day	80 °C	99.52±0.23	4.39×10^5

The pretreatments were required for all the samples dissolved in H_2SO_4 and NaOH/urea aqueous solvent systems. However, an obviously longer acid hydrolysis time was needed for NaOH/urea aqueous solution. It indicated that its dissolution capacity is not as strong as that of concentrated H_2SO_4 aqueous solution. Similar phenomena were observed when spruce cellulose was dissolved in these two solvent systems.[22] Compared to them, LiCl/DMAc solvent was able to directly dissolve cotton fabrics without sulfuric acid pretreatment, but the fabrics required multiple steps of activation (washed with water, acetone and DMAc) before dissolution. In addition, only 1% fabrics could be dissolved in LiCl/DMAc solvent, and the considerably longer dissolution time and higher dissolution temperature were needed. For both H_2SO_4 and NaOH/urea aqueous solvent systems, viscous and transparent cellulose solutions were rapidly obtained in only 8 min. However, cellulose molecules in H_2SO_4 solution were not as stable as in the other two solvent systems. The cotton fabrics dissolved in NaOH/urea solvent underwent the longest acid hydrolysis pretreatments, which resulted in the lowest molecular weight and solution viscosity at the same concentration.[10] Therefore, a relatively higher fabric content was necessary to obtain the solutions with suitable viscosity for regeneration of cellulose films. It was worth noting that the pretreatment durations for bed sheets and jeans were longer than those for t-shirts, but all the samples were successfully dissolved in three solvent systems at the optimized conditions.

The recovery rates of cotton fabrics in three solvents ranged from 82.40±1.43% to 99.83±0.25%, indicating that an efficient recycling approach for cotton fabrics was achieved. Among them, cotton fabrics dissolved in LiCl/DMAc solvent displayed the highest recovery rate with ~99% because the cellulose degradation was the slightest in this solvent. Cellulose was hydrolyzed to some degree in both of the other solvents, so the recovery rates were lower.[9,23]

The Mw of t-shirts, bed sheets and jeans were determined to be 6.94×10^5, 8.40×10^5, and 7.42×10^5 g mol^{-1}, respectively. After dissolution and regeneration, the Mw values of all regenerated cellulose films decreased, especially the films prepared in H_2SO_4 and NaOH/urea solvents. For LiCl/DMAc solvent, it was confirmed that cellulose degradation still occurred at the elevated dissolution temperature even below 85 °C.[24] However, the recovery rates of cotton fabrics in LiCl/DMAc solvent were high, indicating that cellulose hydrolysis was not severe, and cotton fabrics could be recycled efficiently. For H_2SO_4 and NaOH/urea solvents, the sulfuric acid hydrolysis was applied before the dissolution, leading to the partial acid degradation.[22] In addition, the Mw decreased during the dissolution of these two solvents, but the effect of NaOH/urea solvent on Mw was less than that of H_2SO_4 solvent.[9,23] The Mw values of cellulose films prepared in H_2SO_4 solvent were the lowest, which also revealed that this solvent had the greatest impact on the Mw.

3.2. Structure of regenerated films

To investigate the structure change of cotton fabrics before and after dissolution and regeneration in three solvent systems, FT-IR spectra were collected. As shown in **Figure 1**, all samples displayed similar characteristic IR spectra, and no new peaks appeared after dissolution and regeneration, indicating that the processes were a direct physical dissolution and no chemical reaction happened.[18] However, minor differences between the spectra of cotton fabrics and regenerated cellulose films were observed. For example, the absorption peak at 1427 cm^{-1} was attributed to whiskers of cellulose crystalline I,[9] while the peak at 898 cm^{-1} represented amorphous regions.[25] The ratio of these two bands (I_{1430}/I_{900}) that was established as “crystallinity index” and closely corresponded to the portion of cellulose I structure decreased after dissolution and regeneration.[25] Moreover, cotton fabrics exhibited two peaks at 3330 and 3269 cm^{-1} and the former was related to the O(3)H-O(5) intramolecular hydrogen bonds in cellulose I structure.[26] In the regenerated films, these two peaks were flattened and broadened, which suggested the loss of cellulose crystallinity and the change from cellulose I crystals in cotton fabrics to cellulose II crystals after dissolution and regeneration.[15] The change of cellulose polymorph was demonstrated by XRD (**Figure 2**). Cotton fabrics displayed diffraction peaks at ca. 14.8° ($1\bar{1}0$), 16.4° (110), 22.5° (200), and 34° (040), which were typical for cellulose I crystals.[27] However, some of these peaks disappeared in the regenerated films, which exhibited a new broad diffraction at about 20.6°. Especially, another peak at 12° was observed in the films produced from NaOH/urea solutions. It indicated that the cellulose I crystals transformed to cellulose II crystals during the process.[9,28] It was worth noting that there were some other diffraction peaks, for example at 20°, 25.7° and 36°, appeared in the XRD patterns of jeans, which might be attributed to the additives or dyes in textiles (**Figure 2c**). Some of them were removed from the regenerated films (e.g. the considerable decreases of these diffraction peaks in the films prepared from NaOH/urea and LiCl/DMAc solvent systems), while some constituents might be wrapped in cellulose matrix during regeneration.[15]

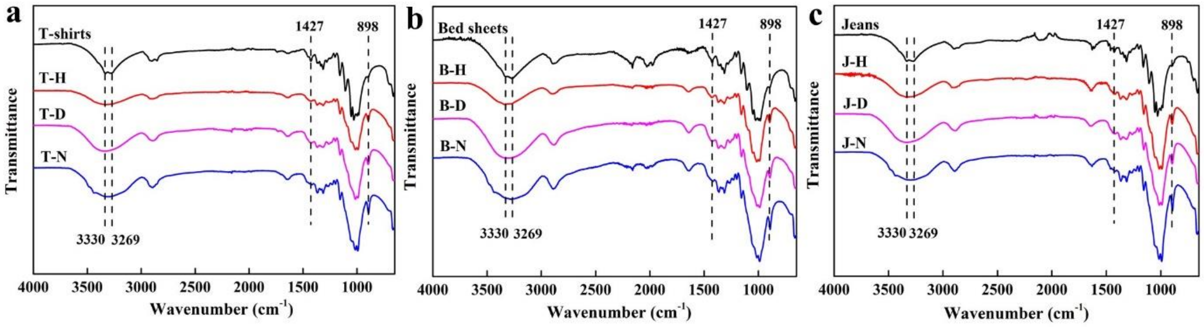

Figure 1. FT-IR spectra of cotton fabrics and regenerated cellulose films prepared in three solvent systems

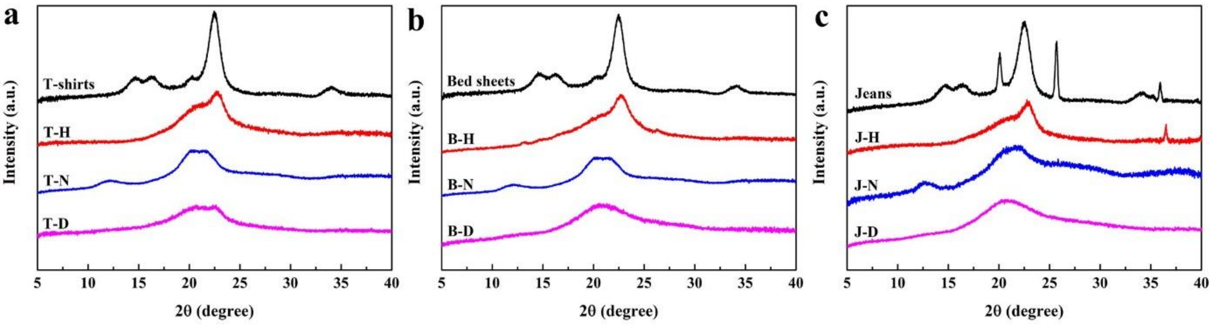

Figure 2. X-ray diffraction patterns of cotton fabrics and regenerated cellulose films prepared in three solvent systems

The surface and cross-sectional morphology of regenerated cellulose films prepared in three solvent systems were observed by SEM. As shown in **Figure 3**, no cotton fibers existed in all the films, which indicated the successful dissolution at our optimized conditions. Similar dense structures were also reported when cotton linter was dissolved in 6 wt% NaOH/4 wt% urea aqueous solution and regenerated in 2 M acetic acid and 2% H_2SO_4 aqueous solution,[29] and would contribute to the mechanical and barrier properties. No obvious difference could be distinguished among the films from t-shirts, bed sheets and jeans. However, the surfaces of regenerated films prepared in NaOH/urea solvent were relatively rougher than the others, which might be due to the rapid and uneven shrinkage during regeneration triggered by high cellulose concentration. A few particles were found in the films derived from LiCl/DMAc solvent, but it should be reminded that the raw materials were dissolved without any chemical pretreatment. As shown in **Figure 4**, no cotton fibers were observed in the cross-section of regenerated cellulose films. Among them, cellulose films prepared in H_2SO_4 solvent displayed the most uniform and homogenous structures. The general trend was consistent with the surface morphology.

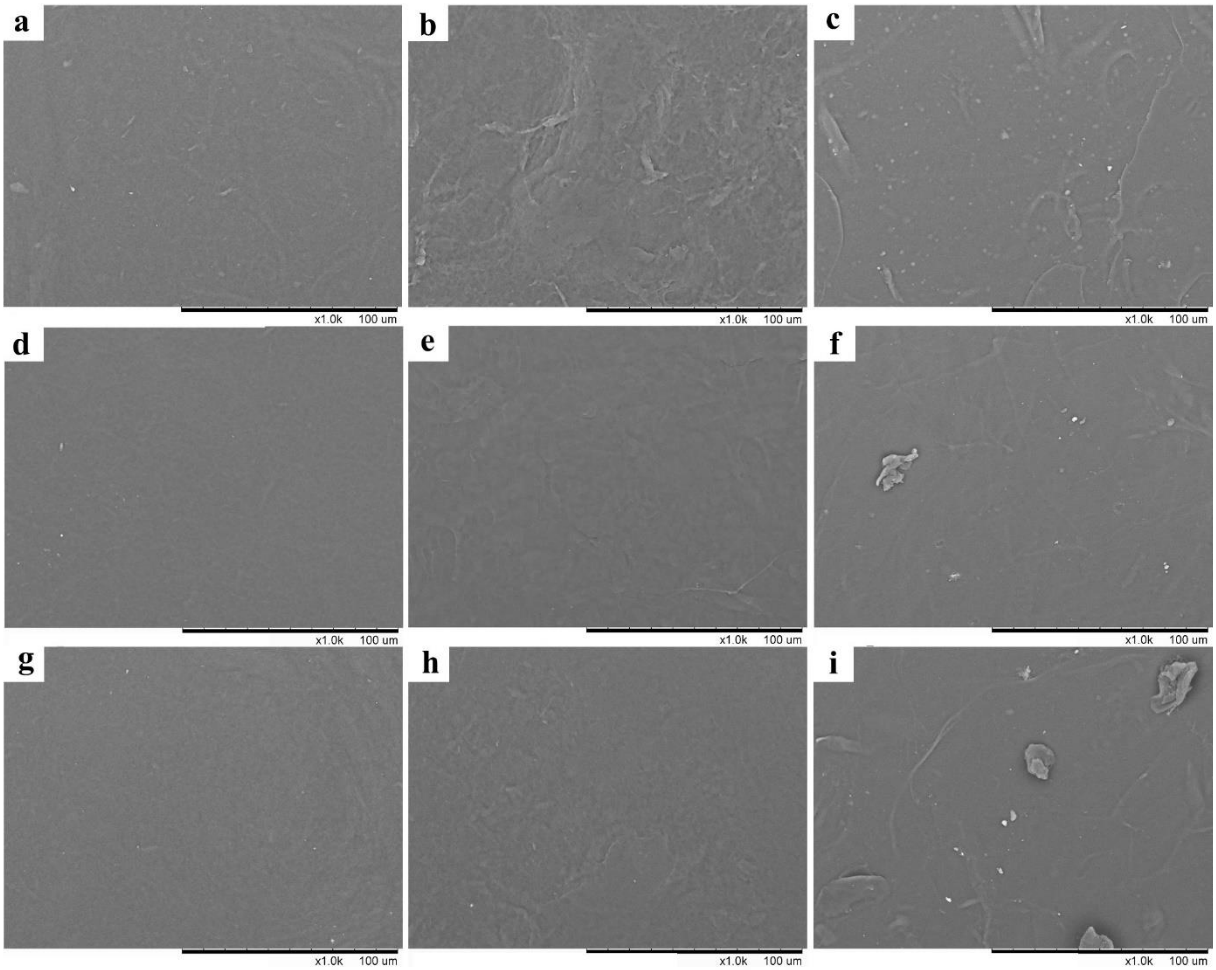

Figure 3. SEM surface images of regenerated cellulose films prepared in three solvent systems: (a) T-H, (b) T-N, (c) T-D, (d) B-H, (e) B-N, (f) B-D, (g) J-H, (h) J-N, and (i) J-D

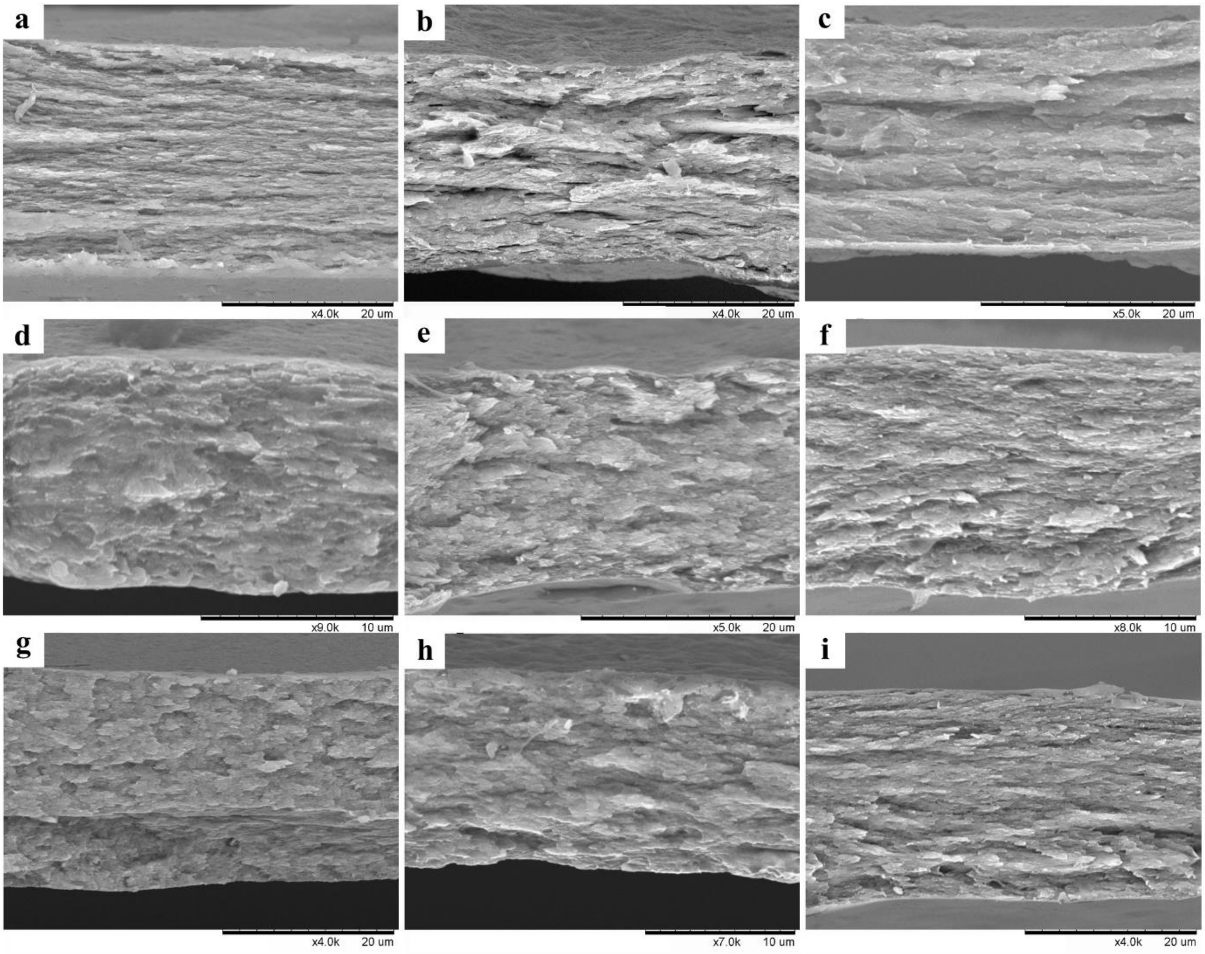

Figure 4. SEM cross-section images of regenerated cellulose films prepared in three solvent systems: (a) T-H, (b) T-N, (c) T-D, (d) B-H, (e) B-N, (f) B-D, (g) J-H, (h) J-N, and (i) J-D

3.3. Properties of regenerated films

The optical transmittance (Tr) of regenerated cellulose films at a wavelength of 800 nm is shown in **Figure 5a**. All the films were translucent (**Figure 5b**), and the Tr values depended on the types of cotton fabrics and solvent systems. Generally speaking, the regenerated films prepared in H_2SO_4 aqueous solution displayed the highest transmittance at the wavelength of 800 nm, followed by the films from LiCl/DMAc and NaOH/urea solvents. The Tr value of regenerated cellulose films could be affected by the following factors: firstly, it decreases with the increasing content of cellulose because higher cellulose content leads to more compact structure;[30] secondly, the presence of undissolved cellulose particles causes optical scattering and internal reflection and result in the decrease of Tr value;[26] and thirdly, the additives in cotton fabrics such as dyes may have significant absorption of visible light.[31] The colors of three cotton fabrics (bed sheets, t-shirt and jeans) are white, light blue and dark blue, respectively. Tr values of cellulose films prepared in three solvents from bed sheets were the highest among all cellulose films due to the light color of bed sheets.

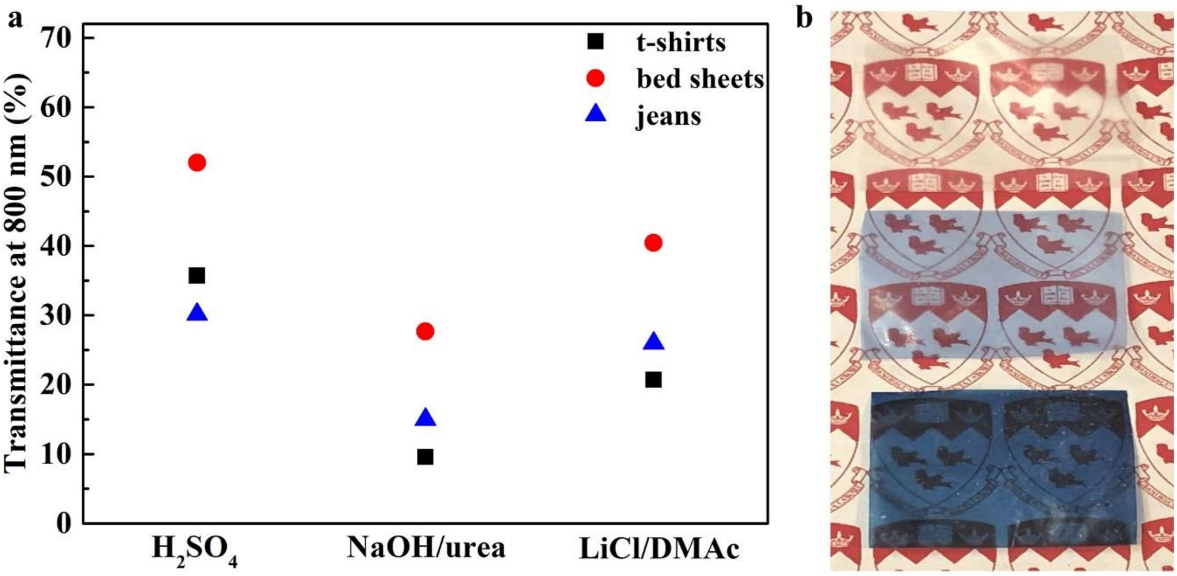

Figure 5. (a) Tr of regenerated cellulose films prepared in three solvent systems at the wavelength of 800 nm; and (b) transparencies of B-H, T-H, and J-H (from top to bottom)

Mechanical properties of regenerated cellulose films were investigated by tensile test (**Figure 6**). It was found that the films from different raw materials using the same solvent displayed similar mechanical properties. However, the tensile strength and Young's modulus of regenerated films prepared in H_2SO_4 and NaOH/urea solvent systems were higher than those of the films obtained from LiCl/DMAc solvent. Cellulose chain entanglement and hydrogen bonds are generated during regeneration to stabilize the cellulose films, and both molecular weight and concentration of cellulose have great impacts on the mechanical properties of regenerated cellulose films. Therefore, the films generated from H_2SO_4 and NaOH/urea aqueous solutions had similar strength, while the cellulose concentration in LiCl/DMAc solvent was lowest, and the presence of undissolved particles in the films acted as the structural defects, leading to inferior mechanical properties.[15] It was worth noting that the strengths of most regenerated cellulose films from cotton fabrics reported in this work (including J-H, J-N, B-H, B-N, and T-H) were higher to those of cellulose films prepared from spruce cellulose and oil palm biomass, corn starch films, and soy protein films.[5,9,12,32]

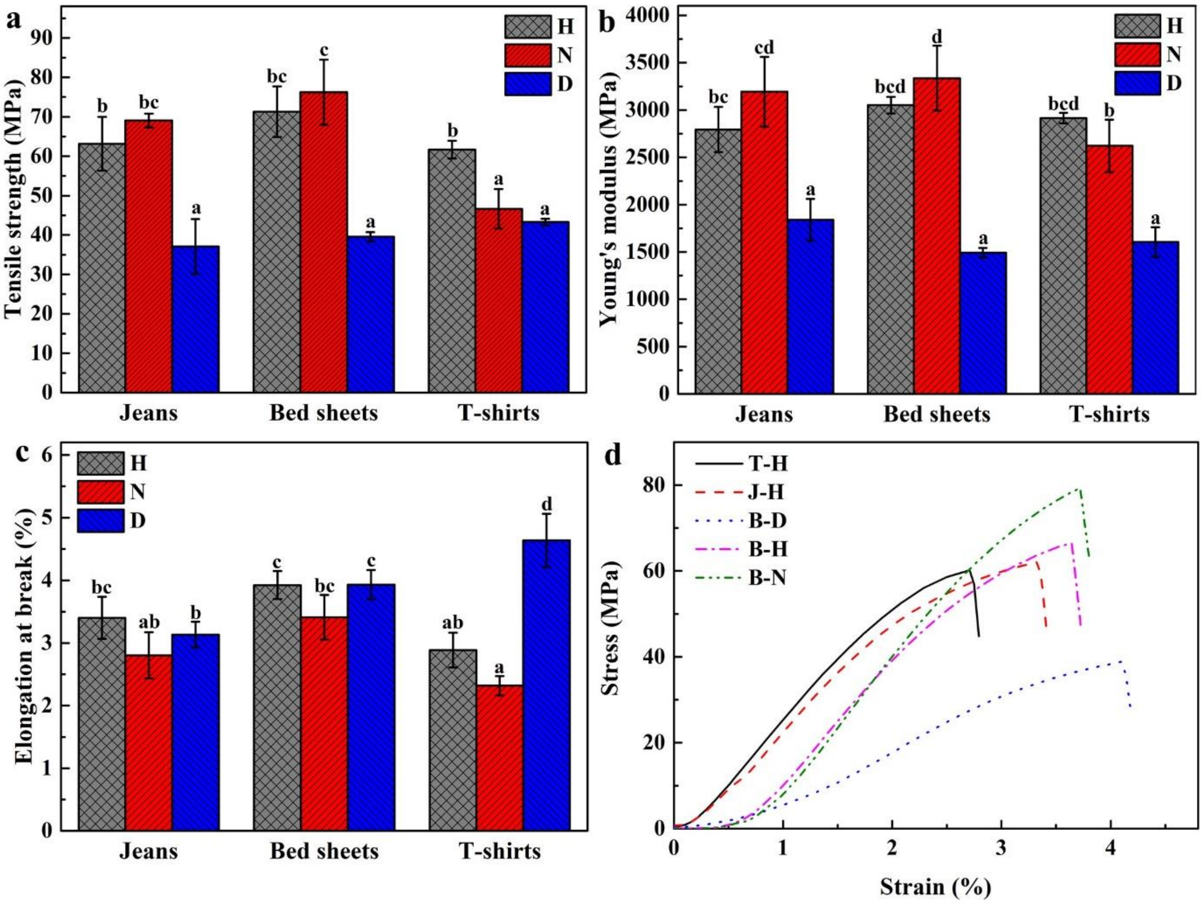

Figure 6. Mechanical properties of regenerated cellulose films prepared in three solvent systems (different letters on the tops of columns represented the significant difference (p<0.05))

Considering the mechanical properties and optical transmittance, five regenerated cellulose films (T-H, J-H, B-H, B-N, and B-D) were selected to study the effects of different types of cotton fabrics and solvents on the thermal stability and water vapor barrier property. TGA and DTG curves are shown in **Figure 7**. The slight weight loss before 150 °C was due to the evaporation of moisture content in the films.[33] All the samples had the major loss of weight in the range of 265 °C to 370 °C, which corresponded to thermal decomposition and carbonization of cellulose.[10] The maximum decomposition temperatures (T_{max}) of T-H, J-H, B-H, B-N, and B-D were 302.09 °C, 295.05 °C, 301.47 °C, 325.66 °C, and 287.51 °C, respectively. The regenerated films prepared in H_2SO_4 aqueous solution showed similar decomposition curves, which might be due to the similar chemical pretreatment and fabric concentration in solutions. However, the sample B-N prepared with the longest acid hydrolysis exhibited the highest T_{max}, while the T_{max} value of B-D film without any pretreatment was lowest. It indicated that, if the fabrics were not excessively hydrolyzed, the concentration of cellulose in solution played a more important role in the thermal stability of regenerated cellulose films. These results were in accordance with their mechanical properties.

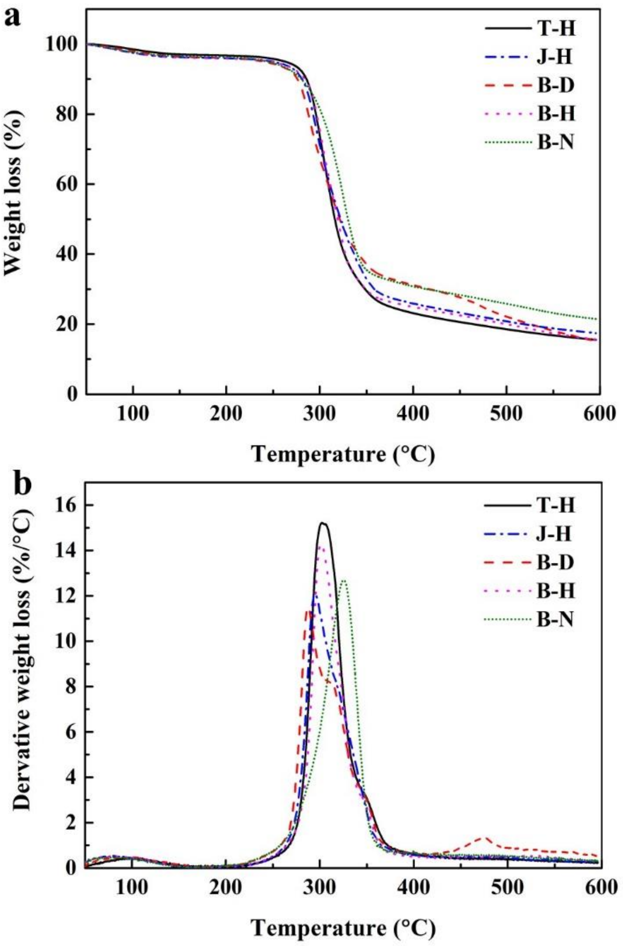

Figure 7. TGA and DTG curves of regenerated cellulose films prepared in three solvent systems

WVP of regenerated cellulose films is shown in **Figure 8**. The film prepared from bed sheets in NaOH/urea aqueous solution exhibited the lowest WVP of $0.81\pm0.10\times10^{-7}$ g m^{-1} h^{-1} Pa^{-1}, while the other films displayed similar WVP values ($p>0.05$). It indicated that different cotton fabrics did not have a significant impact on water vapor diffusion. The increase of cellulose concentration in solution resulted in the formation of regenerated cellulose film with a denser structure, which increased the tortuosity and led to a slower water vapor diffusion process.[34] Although the concentration of B-D was lowest, the B-D and B-H samples had the similar WVP values. It might be due to the cellulose molecules without chemical pretreatment. The WVP values of all regenerated films prepared from waste cotton fabrics were lower than those of cellulose films prepared from oil palm biomass, soybean protein isolate films, and corn starch films.[5,12,32]

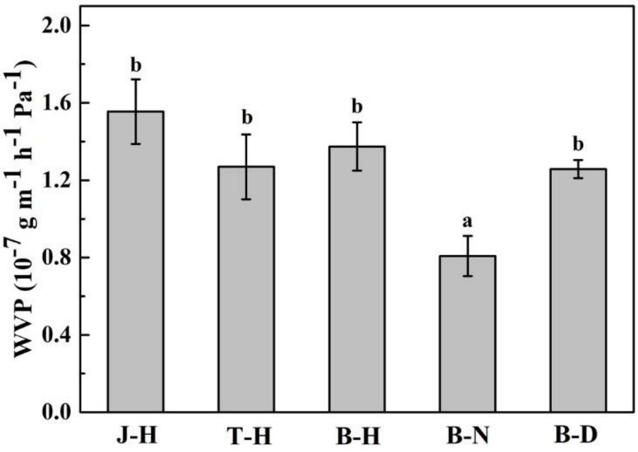

Figure 8. WVP of regenerated cellulose films prepared in three solvent systems (different letters on the tops of columns represented the significant difference (*p*<0.05))

4. Conclusion

T-shirts, bed sheets, and jeans could be directly dissolved in LiCl/DMAc solvent without severe cellulose degradation, but the dissolution required multiple activation steps, low cellulose concentration, high dissolution temperature, and long dissolution time. After the appropriate chemical pretreatments, these cotton fabrics could be rapidly dissolved in H_2SO_4 and NaOH/urea aqueous solutions, but cellulose was less stable in H_2SO_4 and a fairly long acid hydrolysis was needed for the dissolution in NaOH/urea solvent. In addition, acid hydrolysis and H_2SO_4 and NaOH/urea solvents affected the molecular weight of cellulose films obviously. All the solutions were successfully regenerated into translucent cellulose films with smooth surfaces and homogeneous structures. The types of cotton fabrics did not have a significant impact on the mechanical, thermal and water vapor barrier properties of the resultant cellulose films, which would allow the recycle of mixed cotton fabrics through dissolution and regeneration. However, the increase of cellulose concentration in solution resulted in the films with denser structure and obviously improved properties. The films prepared from bed sheets in NaOH/urea solution exhibited the tensile strength of 76.21±8.26 MPa, maximum decomposition temperature of 325.66 °C, and water vapor permeability of $0.81 \pm 0.10 \times 10^{-7}$ g m^{-1} h^{-1} Pa^{-1}, which were comparable to the samples from corn stalk pulp fines, spruce cellulose and oil palm biomass. Therefore, this study provides promising approaches to recycle waste cotton fabrics into biodegradable cellulose films, which have potential applications in food packaging and agriculture.

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Conflict of Interest

The authors declare that they have no conflict of interest.

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