Fabrication and Comparative Evaluation of Regenerated Cellulose Films using Pulp Fines and Pith from Corn Stalk in DMAc/LiCl Solvent System

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Cellulose of corn stalk pulp fines and stalk pith were dissolved in an ionic liquid solvent system (DMAc/LiCl) and then regenerated to form films. The mechanical properties and T_{max} of pulp fines/regenerated cellulose (RC) films were higher than that of the corresponding films from stalk pith. As the ratio of small particles was high in the pulp fines, the elongation at breakage of their RC films also increased to 12.9%. Thus, pulp fines were suitable for prepared regenerated cellulosic materials.

Keywords: Corn stalk pith; Soda-AQ pulping; Pulp fines; Regenerated cellulose film

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INTRODUCTION

Cellulose is the most abundant natural resource in the world. It can be derived from trees, algae, cotton, crop residues, and bacteria. Because cellulose has huge advantages with respect to biocompatibility and biodegradability, it has been a competitive resource in many areas. Due to insufficient storage of forest resources, China imports approximately 234,967,000 m^3 timber per year (Chen *et al.* 2015), but China has abundant resources of crop straw reaching as high as 700 million tons per year (Wu *et al.* 2017). Sadly, most agricultural residues including rice straw, corn stover, and wheat straw have been used improperly or burned.

Corn stalk production was 1661 million tons in 2014 (Liu *et al.* 2018), which equates to 750 million tons of potential cellulose resources. The merits of corn stalk cellulose, as a low cost, abundant, and environmentally friendly material are attractive and desired in this country. By contrast, the rind is rich in fibrous cells. The pithy core of the stalk consists mainly of parenchymal cells, and its weight is about 21% in the whole corn stalk.

Soda-AQ pulping is a modified chemical process for non-wood pulp production using anthraquinone as the auxiliary agent; usage of the AQ decreases carbohydrate degradation during pulping. The paper quality deteriorates due to the presence of pith cells among pulp fibers from corn stalk (Jahan and Rahman 2012), as the parenchyma cells from stalk pith can cause high chemical consumption, sheet-opacity reduction, washing difficulties, and dewatering problems during pulping and papermaking manufacturing (Jiang *et al.* 2016). Parenchyma cells are usually treated as a waste in paper-making. Therefore, the key problem for corn stalk utilization is the separation of parenchyma cells from fibers. In this way, the quality of corn stalk pulp can be improved. Meanwhile, the parenchyma cells can be collected and applied as a raw material for fabricating other bio-macromolecules. Several approaches, such as moist, dry, and mechanical depithing, have

been developed for pith-cell elimination (Rainey *et al*. 2013). These methods usually suffer from the limitations of damage to fibers, environmental contamination, and pith-disposal drawbacks.

Most intact cells are isolated and the morphology of the resulting fibers is similar to that in the plant after chemical pulping. In non-wood raw materials, such as straw and wheat, chemical pulp fines are primarily composed of broken, short fibers, and parenchyma cells because the cell types are complex. In contrast, the content of cellulose in corn pith is approximately 26% (Zhang *et al.* 2018a). Parenchyma is a resource for cellulose preparation and modification. Furthermore, on account of their thin cell walls and relatively high specific surface area, the chemical reaction between parenchyma cellulose and a solvent or reagent can be expected to be more rapid (Lamaming *et al.* 2015).

Swelling or dissolution begins in the amorphous region of cellulose because the hydrogen bonds are broken easily. The crystalline zones contribute to the low dissolvability of cellulose. Compared with wood, cotton, or corn stalk rind, the crystalline index of corn stalk pith is much lower, and the structure is looser (Zhang *et al.* 2018b). These factors suggest that cellulose from corn stalk pith is a suitable material for dissolution. Both mechanical and chemical depithing processes cause damage to corn stalk pith cellulose. However, the effect of cellulose by different obtained methods on dissolution and regeneration were of little concern.

In this study, whole corn stalk was chosen for soda-anthraquinone (AQ) pulping, and the pulp was separated by screening to obtain the fines. The celluloses of corn stalk pulp fines and corn stalk pith were dissolved with DMAc/LiCl solutions. The mechanical properties of these regenerated films were assessed. This work showed that cellulosic fines achieved favorable dissolution capacities in DMAc/LiCl systems.

EXPERIMENTAL

Materials and Chemicals

Corn stalk (CS) was collected during the year 2018 from the Kunming area in Yunnan Province, China. The CS was washed with deionized water and air-dried, providing a weight of almost 50 kg. The stalk was manually cut into blocks with the length of 3 cm to 4 cm and stored for cooking. In addition, the rind and pith of CS were separated and ground using an analytical mill (IKA A11 analytical mill, IKA, Staufen, Germany) with stainless steel grinding blades, and sieved (40 mesh to 60 mesh) manually to obtain the fraction. In the meantime, the powder of pith was separated from CS, then milled and stored for cellulose preparation.

The chemical properties of the CS rind and pith, pulp cooked by whole corn stalk, and CS pulp fines (CSPF) that separated from pulp were determined according to the TAPPI standards T211 om-93 for ash (2000), T204 cm-07 (2007) for solvent extractives, T222 om-83 for lignin (1999), and T201 wd-76 for cellulose (2004).

The N, N-dimethylacetamide (analytical grade, purity ≥ 99%) and lithium chloride (analytical grade, purity ≥ 99.9%) were obtained from Aladdin (Shanghai, China). Sodium hydroxide, glycerol, sodium chlorite, and other chemicals were all purchased from Tianjin Fengchuan Chemical Reagent Technologies Co. Ltd (Tianjin, China) and used without further purification. Deionized water was utilized throughout this work.

Soda-AQ Pulping Process

Soda-AQ cooking was employed in a 15 L laboratory rotating digester, (model ZQS1-15, Shanxi University of Science and Technology Machinery Factory, Xianyang, China). The active alkali charge was 17% on o. d. raw materials. The following parameters were kept constant in the soda process: anthraquinone charge, 0.05% on p. d. raw materials; liquor to fiber ration, 5:1; temperature, 150 °C; time at maximum temperature, 90 min; time to maximum temperature, 150 min. After cooking, the pulp was screened and washed until the pH of the filtrate water was neutral. The properties of CS pulp and fines were determined according to the following standard procedures: T236 om-99 for the kappa number (1999), ASTM D588-42 for the α-cellulose content (2000), ASTM D1795-62 for the degree of polymerization (1984).

Screening and Subfractionation of Corn Stalk Pulp Fines

The pulp fines were separated from CS pulp by laboratory standard screens of 120 meshes (120 μm). After screening, the residue was collected and reserved in a plastic bag.

Pretreatment for Corn Stalk Cellulose

The pulp fines and stalk pith powder were suspended in water under the condition of 3% consistency at 70 ± 0.5 °C. Sodium chlorite and glacial acetic acid were added at the ratio of 2.5:1 (w/v). This mixture was heated at 70 °C ± 0.5 °C for 60 min in a water bath. This procedure was repeated at least 3 times for full delignification (Ruzene *et al.* 2009). Delignified fines were filtered and washed with deionized water until the filtrate pH was neutral. The holocellulose was dried for 48 h in a fume hood and collected.

Subsequently, 20 g of holocellulose was suspended in 500 mL of 10% potassium hydroxide solution for 15 h at room temperature. The mixture was filtered, washed, and air-dried for the next process.

Dissolution and Regeneration of Cellulose

Cellulose specimens were freeze-dried for 24 h to remove their residual water before the dissolution process. Two cellulose specimens were heated and stirred at 110 °C for 120 min in DMAc solution for activation. After that, LiCl (8/100, w/v, compared to DMAc) was added, and the mixtures were heated and stirred at 100 °C for 120 min. The suspension was stored at 4 °C for 6 h, where it was gradually transformed into a gel-like sample. Afterwards, the gel was spread on a polytetrafluoroethylene (PTFE) plate, where it coagulated in the air without heating. The plate with cellulose gel was immersed into 4% glycerol solution for regeneration, and the target film was detached. In the end, the regenerated film was rinsed with running water for 15 min, dried, and stored for characterization.

Chemical and Physical Characterizations of Corn Stalk Pulp Fines and Stalk Pith Cellulose

Analysis fibers from corn pulp fines and corn stalk pith

The CS pulp fines and CS pith lengths, diameters, and fine contents were examined with a fiber quality analyzer (model LDA02, Optest Equipment Inc., Hawkesbury, Canada). The fiber length was determined in the range of 70 μm to 10,000 μm, and the fines were defined as the fiber portion with the fiber length between 70 μm and 200 μm. Morphological features were examined with a Leica DM1000 light microscope (Wetzlar, Germany).

Degree of polymerization analysis

The analyses were performed on CS pulp fines and CS pith according to TAPPI T 230 om-13 (2013). The intrinsic viscosity was measured with a Pinkevitch viscosimeter (Xingao Instruments co., LTD, Shanghai, China). The obtained intrinsic viscosities were converted into the respective values of DP according to Eq. 1,

$$DP^{0.905}=0.75\eta \quad (1)$$

where *DP* is the initial polymerization degree of cellulose, and the η is intrinsic viscosity.

Optical transmittance

The optical transmittance (T_O) of regenerated cellulose films was measured with a TU-1900 UV-vis spectroscope (Pgeneral Co. Ltd., Beijing, China) at the wavelength of 800 nm (Zhao *et al.* 2014). Each sample was tested 5 times, and the average values were recorded.

Scanning election microscopy (SEM)

The SEM observation of the regenerated films was performed using a Nova Nanosem 450 (FEI, Hillsboro, OR, USA) scanning electron microscope at an accelerating voltage of 5.0 kV. The samples were placed onto a carbon disk attached to the stage. The areas of observation were chosen randomly.

X-ray diffraction (XRD)

The crystalline phases of samples were determined by XRD measurement on an X'Pert 3 powder diffractometer (PANalytical Co. Ltd., Almelo, Netherlands). The samples were positioned on a glass sample holder. Radial scans of intensity were recorded in ambient conditions over scattering 2θ angles from 3° to 50° (step size = 0.01313°, scanning rate = 13.77 s/step) using Cu Kα radiation (λ = 1.5406 nm), an operating voltage of 40 kV, and a filament current of 40 mA.

The crystallinity index (CrI) was calculated based on the peak deconvolution results as follows,

$$CrI = \frac{A_{total}-A_{am}}{A_{total}} \times 100\% \quad (2)$$

where A_{total} and A_{am} represent total area of the diffractogram and the area of amorphous peak, respectively. The total intensity of the (200) peak for cellulose I is taken near 23°. All CrI values in this study were calculated from the original data without subtracting background. The XRD spectra were analyzed by Jade 6.0 software (Kreze and Malej 2003; Xing *et al.* 2018).

Thermogravimetric analysis (TGA)

The TGA of CS pulp fines and CS pith regenerated cellulose films was performed using the Netzsch STA 449 F3 (Selb, Germany). Thermograms of samples were recorded between 50 °C and 600 °C with a heating rate of 10 °C /min in an inert atmosphere maintained by a nitrogen flow of 50 mL/min. Proteus software (Version 5.0.0, Selb, Germany) was used for calculating the first derivatives of the thermograms (DTG), the percentage weight loss, and the decomposition temperatures.

Mechanical properties

The mechanical properties of films were measured using a tensile testing machine (model DRK 101, Jinan Drick Insrument Co. Ltd, city, China). The film was tested in accordance with the ASTM standard method D882-97 (1999). The testing machine was fitted with a 200 N load cell with a crosshead speed of 20 mm/min, and the initial distance between the grips was 50 mm. The film was cut into 15 mm wide strips, and measurements were performed at room temperature. The average of three measurements was taken for each film.

RESULTS AND DISCUSSION

Cell Morphology and Chemical Characteristics of Corn Stalk Pulp and Fines

The properties of the pulp and black liquor are given in Table 1. The random occurrence of alkaline scission among glycosidic linkages leads to considerable weight loss and a decrease in the degree of polymerization when cellulose is heated over 170 °C (Knill and Kennedy 2003). Thus, the maximum temperature of cooking was set at 150 °C to reduce the damage on carbohydrates. The rejects and yields of CS pulp were 0.36% and 46.8%, respectively. The Kappa number was 13, and the DP was 454. These results indicated that the corn stalks were delignified and produced a high pulp yield under mild conditions.

Table 1. The Results of Soda-AQ Cooking of Corn Stalk

Sample	Black liquor		Pulp			
	pH	Residual alkali (g/L)	Yield (%)	Rejects (%)	Kappa number	Degree of polymerization
Corn stalk	11.8	3.99	46.83	0.36	13	454

Cell morphologies of the corn stalk pulp and pith are shown in Fig. 1. Many parenchyma cells were present in unscreened CS pulp (Fig. 1a). Numerous parenchyma cells can plug paper sheets, reduce paper machine drainage, and increase paper density, which are the main reasons that corn stalk has not been used widely in papermaking (Li *et al.* 2012). There are four kinds of parenchyma cells shown in Fig. 1a, including claviform, round, pillow-like, and spongy. The average fiber length and width was 0.250 mm and 21.5 μm, respectively, and the fines percent was 56.0%.

After the screening, the number of parenchyma cells was clearly decreased in the residue, as shown in Fig. 1b. Meanwhile, it can be observed that the width parameters of parenchyma cells were much larger than that of fiber, suggesting that the complete separation of parenchyma cells from fibers was hard. In the course of the separation process, the percent of fines was decreased from 58.1% to 30.8%, fiber length was increased to 0.496 mm, and the width was decreased to 18.6 μm. The average length of the screened pulp was closer to that of the corn stalk rind (0.610 mm), which suggested that the screening had a useful effect on the separation of parenchyma cells. In the CS pulp fines picture (Fig. 1c), parenchyma cells, short fibers, and broken fibers can be found. It shows that the number of fibers decreased after the primary screening. The results of fiber length and width was 0.235 mm and 21.06 μm, respectively. More importantly, the fines percent was increased to 70.4%, and the lengths of majority cells were shorter than 0.2 mm.

The specific surface area decreased from 4.02 m^2/g to 1.13 m^2/g after the screening, as exhibited in Table 1. The main reason of this change was the fines were separated out from the pulp, thus the pulp was almost composed by fibers. Meanwhile, the specific surface area of fines has been increased to 9.067 m^2/g. The phenomenon showed that the specific surface area of fines was far larger than fibers, which indicated that the effect of the contact with chemical solvent was much better.

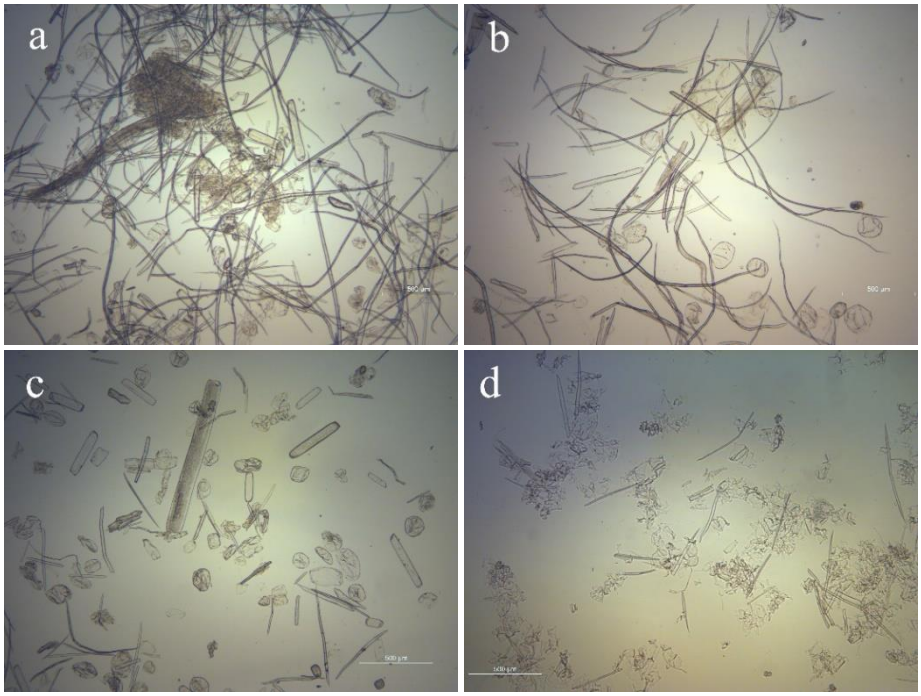

Fig. 1. Cell morphology of different samples from corn stalk by optical microscopy: (a) unscreened pulp; (b) screened pulp; (c) pulp fines; and (d) stalk pith

Comparing the fiber lengths of CS pith and screening pulp fines, the lengths of the pulp fines were lower than stalk pith. Combining the Fig. 1d, the main reason of this result was that numerous small cells were broken during the milling process. This means whether moist or dry, mechanical depithing process caused remarkable damage on the structure of the cells. In this case, whole stalk pulping would be a good method to protect the integrity of plant cells.

The influence of pulping and screening on chemical compositions is presented in Table 2. The pulping process played a key role in removing the lignin and hemicellulose, which greatly increased the cellulose content from about 38% for CS raw material to 78.92% for pulp. Consequently, the content of Klason lignin reduced from approximately 19% for the raw material to 1.90% for the pulp. The change of ash and benzene ethanol soluble extractives followed the same trend. Because of the decrease of parenchyma cells after the screening process, the DP value of pulp increased from 454 to 493, which was close to that of rind.

Table 2. Morphology and Chemical Components of Corn Stalk Pith, Rind, CS Pulp, and Fines

Sample		Morphology				Chemical Components					
		Fiber length (mm)	Fiber width (μm)	Fines percent (%)	BET specific surface area (m^2/g)	Ash (%)	Benzene-ethanol soluble extractives (%)	Cellulose (%)	Klason lignin (%)	Pentosane (%)	DP
Raw material	Pith	0.372	18.30	56.0	nd	5.02	7.50	37.45	12.56	23.65	330
	Rind	0.610	19.60	42.2	nd	6.05	5.01	38.13	19.10	17.93	546
Unscreened CS pulp		0.250	21.50	58.1	4.015	4.01	4.89	78.92	1.90	18.66	454
Screening	Pulp	0.496	18.60	30.8	1.126	2.20	1.86	80.00	1.21	13.48	493
	Fines	0.235	21.06	70.4	9.067	3.07	1.96	76.51	0.85	8.62	348

Chemical and Physical Characterization of Regenerated Cellulose Films

Corn stalk pulp fines and pith cellulose were dissolved in the DMAc/LiCl solvent and subsequently regenerated in water to form a membrane. As shown in Fig. 2, the mixed cellulose solutions were very stable after storage under a temperature of 3 °C for over a month. The surface of both regenerated films appeared to be smooth and homogeneous. The background was observed clearly through the films in the digital photograph, indicating that the transmittance of RC films was high. The optical transmittance (Tr) at the wavelength of 800 nm for these films is shown in Table 3. The Tr values were 79.3% for pulp fines RC film and 75.2% for pith RC film. These results suggest that larger fibrils increase light scattering and thus reduce the transmittance (Borch and Marchessault 1969; Zhao *et al.* 2017).

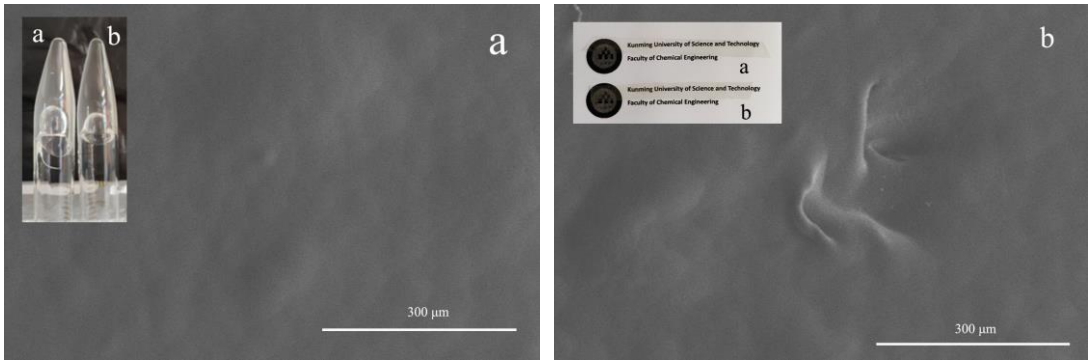

Fig. 2. SEM image of regenerated cellulose film from a) corn pith cellulose and b) corn stalk pulp fines; the mixed cellulose solutions and appearance of films are shown in the inset

The Segal method for calculating the crystallinity of cellulose was used because it is simple and convenient. The areas of the crystalline and amorphous regions in the XRD spectra were used, and the crystallinity based on the ratio of the crystalline area to the total area was employed for convenient comparison in this study. The macromolecules of the dissolved cellulose were very hard to recrystallize in the regenerated process without any alignment. Therefore, the patterns of RC samples were all essentially amorphous, which is similar to cellophane (Zhang *et al.* 2015).

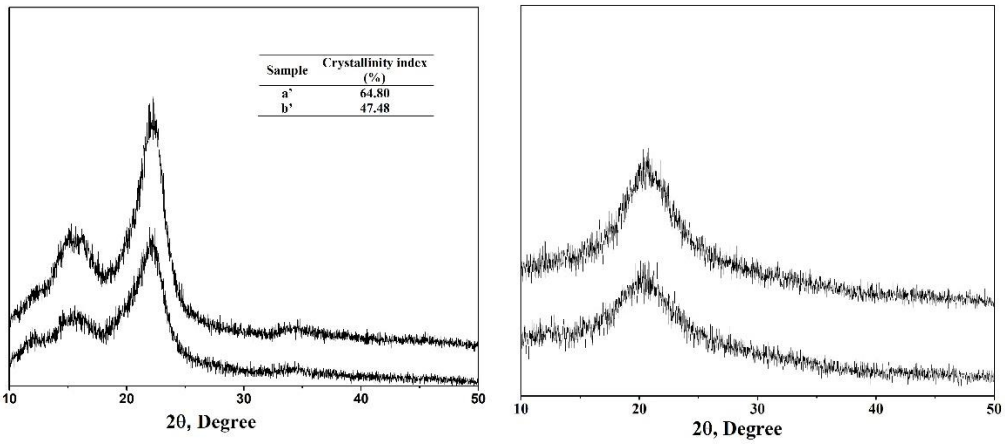

Fig. 3. XRD patterns of a` corn stalk pith cellulose, and b` corn stalk pulp fines cellulose; regenerated cellulose from a corn stalk pith, b` pulp fines

Figure 3 shows the wide-angle X-ray diffraction (WAXD) of cellulose samples and their RC films from DMAc/LiCl solutions. The diffractogram exposed changes in the intensities of the characteristic peaks and the transformation of crystal forms from cellulose I to cellulose II with the decrease of fine sizes. In the curve of cellulosic fines (Figs. 3a', b'), the diffraction peak at $2\theta = 15.4°$ was the superposition of $(1\bar{1}0)$ and (110) planes, and the peak at $2\theta = 22.7°$ was for (200) plane, which represents the characteristics of cellulose I crystal (Panaitescu *et al.* 2015; Sadeghifar *et al.* 2017). These diffraction intensities of pulp fines cellulose samples were lower compared with the pith cellulose. The crystallinity index of cellulose can be calculated as 64.80% for pith and 47.48% for pulp fines. This indicated that the structure of cellulose crystallinity was damaged during the pulp process. Furthermore, the low crystallinity of cellulose means more amorphous regions, resulting in easier swelling and dissolution.

After dissolution and regeneration, the crystalline structure of cellulose has been changed, and the main diffraction peaks of RC films were shifted to 20.1° $(10\bar{1})$. The highest scattering intensity of RC films presented the crystal type of cellulose II (Zhao *et al.* 2014). These cellulose samples were dissolved at a highly amorphous state. When they were regenerated from deionized water, their diffraction peaks became broader and gentler, indicating the increase of amorphous regions in cellulose molecules (Teng *et al.* 2018). The pulp fines cellulose exhibited an excellent solubility in the DMAc/LiCl system, and consequently, the regenerated cellulose showed the low diffraction intensity in the XRD spectrum.

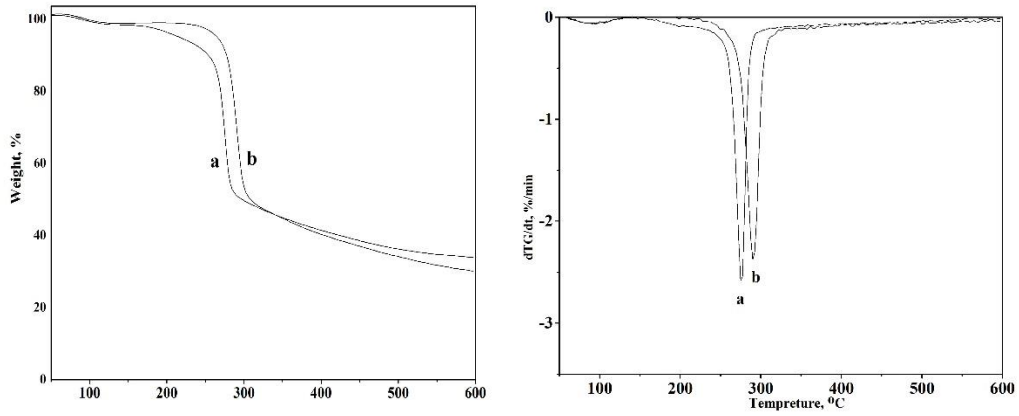

Fig. 4. TGA and DTG cures of RC films by a) corn stalk pith and b) corn stalk pulp fines

The slight mass loss before 150 °C was due to the evaporation of water from the regenerated cellulose (Zhao *et al.* 2014). The losses of both RC films were almost the same in this stage as shown in Fig. 4. Qualities of the biopolymers decreased rapidly, and all the samples started to decompose with charring and volatilization in the range of 220 °C to 330 °C. The T_{max} of pulp fines RC film was 268.6 °C, which was a little higher than that of the stalk pith RC film. This phenomenon was due to the cellulose of the pulp fines that owned lower crystallinity, which can be dissolved much better in chemical solvent. Therefore, the regenerated cellulose probably has a good ordered arrangement of macromolecular. Thus, the thermostability of pulp fines RC film was higher compared that of the stalk pith.

Table 3. Results of Tensile Testing, Optical Transmittance Measurements, and TGA Analysis of RC Films

Samples	σb (MPa)	E (GPa)	ε (%)	Tr (%, 800 nm)	T_d (°C)	T_{max} (°C)	DP
Stalk pith	30.65	16.8	8.26	75.2	253.9	273.7	180
Pulp fines	48.34	27.2	12.94	79.3	268.6	289.8	195

Note: σb, stress at failure; E, Young's modulus; ε, strain at failure; Tr, optical transmittance; CA, water contact angle; T_d, initial decomposition temperature; and T_{max}, maximum decomposition temperature.

The effects of different RC films on the mechanical properties of their regenerated films are shown in Table 3. The physical strengths of pulp fines RC films were much higher than that of stalk pith RC films. A similar trend was observed in the change of Young's modulus (E), with E values from 27.2 GPa to 16.8 GPa for the RC films of pulp fines and stalk pith, respectively. According to Duan *et al.* (2016), low molecular weight compounds act as plasticizers in RC films, leading to increased elongation at break. In this case, the elongation at break (ε) of both RC films exhibited a high value. Compared with fibers, the molecular weights of CS pulp fines and the parenchymal cells were much lower, indicating that the small size part in these were acted as a plasticizer in the RC films. The ratio of fines in pulp fines was higher when compared with stalk pith, exhibited that the content of lower-mass monomers was higher. Thus, the improved plasticizing performance of RC films led to higher elongation at break. The DP value for RC films were almost at the same level. Compared with the DP value shown in Table 2, this result suggested that the cellulose macromolecules of the RC films were degraded during the dissolution process.

CONCLUSIONS

1. The cellulose of corn stalk pulp fines and stalk pith was dissolved in DMAc/LiCl solvent and regenerated in deionized water. The cells of pulp fines had better integrity than that of pith that was processed by manual separation.
2. The physical characterizations of RC films had a close relationship with the ratio of fines in raw cellulose. Comparing with stalk pith RC films, the mechanical strength and thermostability of pulp fines RC films had the advantage. Thus, corn stalk pulp fines could be an alternative and valuable resource for widening applications.

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