

# Preparation of Microcrystalline Cellulose by Liquefaction of Eucalyptus Sawdust in Ethylene Glycol Catalyzed by Acidic Ionic Liquid

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Microcrystalline cellulose (MCC) was prepared from the residue of the incomplete liquefaction of eucalyptus sawdust; atmospheric liquefaction was carried out using ethylene glycol as the solvent and 1-(4-sulfobutyl)-3-methylimidazolium hydrosulfate as the catalyst. The highest cellulose content in the residue reached 93.9%. The MCC prepared from liquefaction was characterized by various techniques, which included infrared spectroscopy, X-ray diffraction, thermo-gravimetric analysis, and scanning electron microscopy. The results were compared to those of a commercial MCC (cotton linters). The analyses indicated that hemicelluloses and lignin were removed extensively from the MCC produced from the sawdust. The MCC was a cellulose I polymorph with 79.0% crystallinity. The particles were shaped as elongated rods and had good thermal stability. The particle sizes of the produced MCC ranged from 1  $\mu\text{m}$  to 100  $\mu\text{m}$  with a mean of 38.6  $\mu\text{m}$ .

*Keywords:* Microcrystalline cellulose; Liquefaction; Eucalyptus sawdust; Acidic ionic liquids

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## INTRODUCTION

Cellulose is the most abundant natural polymer on the earth. Commercial microcrystalline cellulose (MCC) is a purified and partially depolymerized cellulose that appears as a white, tasteless, and odorless crystalline powder. It has been widely used as an additive in processed foods, pharmaceuticals, cosmetics, and various chemical products as a thickener, stabilizer, texture modifier, or emulsifier. The MCC is generally regarded as safe when used in moderate quantities (Tuason *et al.* 2009).

Microcrystalline cellulose can be prepared by treating cellulose with mineral acid until the degree of polymerization has been reduced and reaches a floor-level. It can be produced from any material that is high in cellulose, which can range from pure cellulose to commercial grade cellulose to lignocellulosic materials. Wood pulps and cotton are the principal sources for the commercial production of MCC. Other lignocellulosic resources for MCC production, such as rice, bean hulls, jute, and sago seed shells, have been also studied (Adel *et al.* 2011; Jahan *et al.* 2011; Naduparambath and Purushothaman 2016). Moreover, different methods have been employed to isolate MCC from lignocelluloses. Ejikeme (2008) isolated MCC from orange mesocarp by pulping it with sodium hydroxide, which was then bleached with sodium hypochlorite and hydrogen peroxide, and that mixture was subsequently hydrolyzed with hydrochloric acid. Wang *et al.* (2009) isolated MCC from jute fiber by mercerizing it with sodium hydroxide, then steam treating it with

hydrochloric acid, and subsequently treating it with NaOH. Kusumattaqiin and Chonkaew (2015) isolated MCC from cotton wool by using rapid microwave-assisted acid hydrolysis.

Currently, liquefaction has been recognized as an effective method for biomass utilization; various liquid products can be obtained by this process from the use of lignocellulosic materials. Enhanced liquid product yield has been the focus of most studies; however, the use of the liquefaction residue has been largely ignored. In some cases, valuable products can be isolated from the liquefaction residue. Zhang *et al.* (2012) studied the effects of various process conditions on the liquefaction yield from Chinese eucalyptus. The authors used sulfuric acid as the catalyst in the ethylene glycol solvent; then characterized the chemical compositions of liquefied wood residues. The investigations noted that the liquefaction process could be divided into two stages; *i.e.*, a rapid liquefaction stage that corresponded to lignin, hemicelluloses, and cellulose removal in the amorphous regions, and a slow stage that corresponded to cellulose degradation in the relatively stable crystalline regions. Zhang *et al.* (2007) reported that bagasse was liquefied in ethylene glycol that was catalyzed by sulfuric acid at 190 °C under atmospheric conditions. The authors found that the hemicelluloses and lignin were hydrolyzed more quickly than cellulose, and that liquefaction residues were comprised primarily of undissolved cellulose and lignin or lignin derivatives. Jasiukaitytė *et al.* (2009) studied cellulose liquefaction in acidified ethylene glycol. The authors reported that the cellulose in the disordered regions became degraded in the first minute of liquefaction. However, highly ordered cellulose regions remained stable for a longer time. In addition, the investigators noted that prolonging liquefaction time can reduce the degree of polymerization of the cellulose to a certain floor-level value (*i.e.*, leveling-off degree of polymerization). In recent years, ionic liquids have been recognized as effective solvents or liquefying reagents in biomass liquefaction; these solvents can dissolve wood, cellulose, and lignin (Pu *et al.* 2007; Fukaya *et al.* 2008; Long *et al.* 2011). However, ionic liquids are not widely used in liquefaction due to their high costs and their difficulties to separate and recover from the liquefied solute. Furthermore, ionic liquids are not as recyclable as some investigators have expected. Despite these shortcomings, some acidic ionic liquids are highly efficient catalysts in the liquefaction reactions because of their high acid density, low corrosively, uniform active centers, *etc.* (Long *et al.* 2012, 2013; Lu *et al.* 2015).

In this work, the authors examined MCC, with the goal to obtain a more economically beneficial product from the incomplete liquefaction of eucalyptus sawdust. An acidic ionic liquid, 1-(4-sulfobutyl)-3-methylimidazolium hydrosulfate, was used as the catalyst in the liquefaction process. The effects of liquefaction reaction time, temperature, and acid concentration on liquefaction yield and cellulose content in the residues were studied. The aim of this work is to investigate the possibility of preparing microcrystalline cellulose (MCC) from sawdust *via* the liquefaction process.

## EXPERIMENTAL

### Materials

*Eucalyptus grandis* sawdust was obtained from a wood processing factory in Leizhou, China. The sawdust was sieved to a particle size between 300 µm to 400 µm, and it was oven-dried at 105 °C to a constant mass value prior to its use. The sawdust's composition was 47.9% cellulose (*via* the nitric acid-ethanol method of Shi and He (2003)), 71.2% holocellulose (GB/T 2677.10 (1995), 27.1% Klason lignin (TAPPI T222 om-07

(2007)) and 0.29% ash (TAPPI T222 om-11 (2011)). The acidic ionic liquid, 1-(4-sulfobutyl)-3-methylimidazolium hydrosulfate ( $[\text{C}_4\text{H}_8\text{SO}_3\text{Hmim}] \text{HSO}_4$ ), was purchased from the Lanzhou Institute of Chemical Physics, Chinese Academy of Sciences (Lanzhou, China). Aqueous cupriethylenediamine solution (1.0 M) was purchased from Sigma-Aldrich (Shanghai, China). Commercial microcrystalline cellulose (cotton linters, 25  $\mu\text{m}$ ; Aladdin (Shanghai, China)) was used as a standard for comparisons. All other chemicals used in this study were of reagent grade and were used without further purification (Guangdong Guanghua Sci-Tech Co., Ltd., Shantou, China).

### *Liquefaction*

Air-dried sawdust (10 g), ethylene glycol (80 g), and a specified amount of the  $[\text{C}_4\text{H}_8\text{SO}_3\text{Hmim}] \text{HSO}_4$  were premixed thoroughly. Next, the mixture was transferred to a 100-mL three-necked flask equipped with a mechanical stirrer, a reflux condenser, and a thermometer. The flask was heated to a preset temperature (140 °C, 160 °C, and 180 °C) for the prescribed reaction time (15 min, 30 min, 60 min, and 90 min). Afterwards, the reaction was quenched with cold water. The mixture was diluted with 4:1 (v/v) 1,4-dioxane-water and filtered to obtain the liquefaction residue. The residue was thoroughly rinsed with 1,4-dioxane-water solution and then rinsed with acetone to remove traces of 1,4-dioxane-water. The washed residue was vacuum-dried at 50 °C to a constant mass and then weighed to determine the mass of the residue. The liquefaction yield of the sawdust ( $Y_L$ ) was calculated according to Eq. 1,

$$Y_L = (1 - \text{weight of residue} / \text{weight of sawdust}) \times 100\% \quad (1)$$

The major component in the liquefaction residue was cellulose.

### *Analysis of cellulose content of the residue*

The cellulose content of the residue was determined by the nitrate method. A solution of 25 mL of mixture of ethanol and nitric acid (4 : 1, v/v) was used as the solvent for 1 g of the liquefaction residue, refluxing and extracting for four times at 100 °C and 1 h of each times. Then the residue was filtered and washed two times with 25 mL of mixture of ethanol and nitric acid. Finally, the residue was washed with deionized water until the filtrate reached pH 7. The washed residue was dried at 105 °C to a constant mass and then weighed to determine the cellulose content of liquefaction residue.

$$\text{The cellulose content of the residue} = (\text{weight of residue treated with the nitrate method}) \times 100\% \quad (2)$$

### *Purification of cellulose*

The residue was treated with 0.7% aqueous  $\text{NaClO}_2$  solution (pH = 5, adjusted with acetic acid) at 75 °C for 3 h. During the procedure, the pH was adjusted to 4 to 5 with acetic acid once per hour. Then the residue was filtered and washed several times with deionized water. Subsequently, the chlorite-treated residue was extracted with 5% aqueous sodium hydroxide solution at 50 °C for 1 h. Finally, the residue was filtered and washed with deionized water until the filtrate reached pH 7; the solid was vacuum-dried at 50 °C until constant mass was obtained. The purpose of this procedure was to remove any residual lignin and hemicelluloses in the liquefaction residue as to obtain pure cellulose.

### *Identification of microcrystalline cellulose*

The microcrystalline cellulose was identified according to Chinese Pharmacopoeia (Part IV). Iodine (1 g) was added into a solution made by dissolving 20 g of zinc chloride ( $ZnCl_2$ ) and 2 g of potassium iodide into 10 mL of water. To this solution the cellulose sample (0.01 g) was added that was obtained from the liquefaction residue. The resulting mixture was placed onto a watch glass and the color change of the slurry was recorded. The violet-blue color of the cellulose sample indicated the existence of MCC. The identification of MCC was also verified with Fourier transform infrared (FT-IR) spectra, degree of polymerization, and crystallinity measurements.

## **Methods**

### *Degree of polymerization*

The degree of polymerization (DP) of the cellulose was determined by measuring the intrinsic viscosity of cellulose dissolved in the cupriethylenediamine solution. The DP was calculated based on the following equation (ASTM D1795-62 (1974)),

$$DP = 190[\eta] \quad (3)$$

### *FT-IR analysis*

The FT-IR spectra of samples were recorded with a Vector 33 Fourier transform infrared spectrophotometer (Bruker BioSpin GmbH, Rheinstetten, Germany). The samples were pressed with KBr into pellets. Spectra of the pellets were recorded over the wavenumber range of  $4000\text{ cm}^{-1}$  to  $400\text{ cm}^{-1}$ .

### *Thermal stability*

The thermal stabilities of the samples were investigated using a Q200 thermogravimetric analyzer (TA Instruments, New Castle, DE, USA). Analyses were performed in a  $N_2$  atmosphere with a gas flow rate of 25 mL/min. The tests were conducted from  $30\text{ }^\circ\text{C}$  to  $600\text{ }^\circ\text{C}$  with a heating rate of  $10\text{ }^\circ\text{C}/\text{min}$ .

### *X-ray diffraction studies*

X-ray diffraction (XRD) measurements were conducted over a  $5^\circ$  to  $40^\circ$  range on a Bruker D8 diffractometer (Bruker BioSpin GmbH, Rheinstetten, Germany), using  $CuK\alpha$  radiation generated at 0.154 nm, 40 mA, and 40 kV. The crystallinity index ( $CrI$ ) was calculated according to Eq. 2,

$$CrI = (I_{002} - I_{am}) / I_{200} \times 100\% \quad (4)$$

where  $I_{002}$  is the diffraction intensity from the [002] crystalline lattice planes at  $2\theta = 22.5^\circ$ , and  $I_{am}$  is the minimum diffraction intensity from the amorphous region at  $2\theta = 18^\circ$ .

### *Morphological analysis and particle size distribution*

The morphology of the sawdust and microcrystalline cellulose was observed by scanning electron microscopy (SEM) (Carl Zeiss Jena GmbH-EVO 18, Jena, Germany). The particle size distributions of samples were measured using a laser diffraction particle size analyzer (Malver Mastersizer S3000, Malvern Instruments Ltd., Malvern, UK).

## RESULTS AND DISCUSSION

Typically, biomass liquefaction is expected to achieve a high conversion yield at a rapid reaction rate. However, in this work the authors did not pursue a high conversion yield to preserve the cellulose in its crystalline state as much as possible. Lignin and hemicelluloses are amorphous polymers; this feature makes them easier to degrade the intra- and inter-molecular linkages connecting these components than the linkages in the partially crystalline cellulose. Under relatively mild conditions, lignin and hemicelluloses in the wood can be solubilized into liquid products, while the crystalline cellulose can be retained. The challenge was to enhance the selectivity of the liquefaction process.

### The Influence of Reaction Conditions on the Liquefaction Yield and Properties of Cellulose

Table 1 shows the liquefaction yield, the cellulose content, and the degree of polymerization of cellulose in the residue obtained at various reaction conditions. It is well known that the liquefaction yield depends on the liquefying reagents, the catalyst type and dosage, the liquid-to-substrate ratio, the reaction time, and the reaction temperature (El-barbary and Shukry 2008; Zhang *et al.* 2012). Like conventional liquefaction, increased temperature, prolonged reaction time, and/or an increased catalyst level enhanced the liquefaction yield in this work. The data indicated that the cellulose content in the residue was the major constituent (up to 93.9% (Experiment 12)).

The liquefaction conditions, *i.e.*, the temperature, time, and catalyst dosage, had a considerable impact on the DP, as shown in Table 1.

**Table 1.** The Influence of Reaction Conditions on Liquefaction Yield, Cellulose Content of the Residue, and Degree of Polymerization

Experiment No.	Catalyst Dosage (mmol/g sawdust)	Temperature (°C)	Time (min)	Yield of Liquefaction (%)	Cellulose Content in Residue (%)	DP of Cellulose
1	0.1	160	30	43.42	76.35	1139
2	0.15	160	30	55.14	89.05	816
3	0.2	160	30	62.13	92.83	441
4	0.25	160	30	63.26	93.39	280
5	0.3	160	30	64.62	93.56	223
6	0.2	160	15	57.70	90.13	675
7	0.2	160	60	63.85	92.89	281
8	0.2	160	90	66.51	92.82	183
9	0.2	140	30	53.72	83.39	1086
10	0.2	140	60	57.56	88.10	947
11	0.2	140	90	58.91	91.34	667
12	0.2	180	30	69.94	93.88	138
13	0.2	180	60	73.81	92.90	115
14	0.2	180	90	74.89	91.79	118

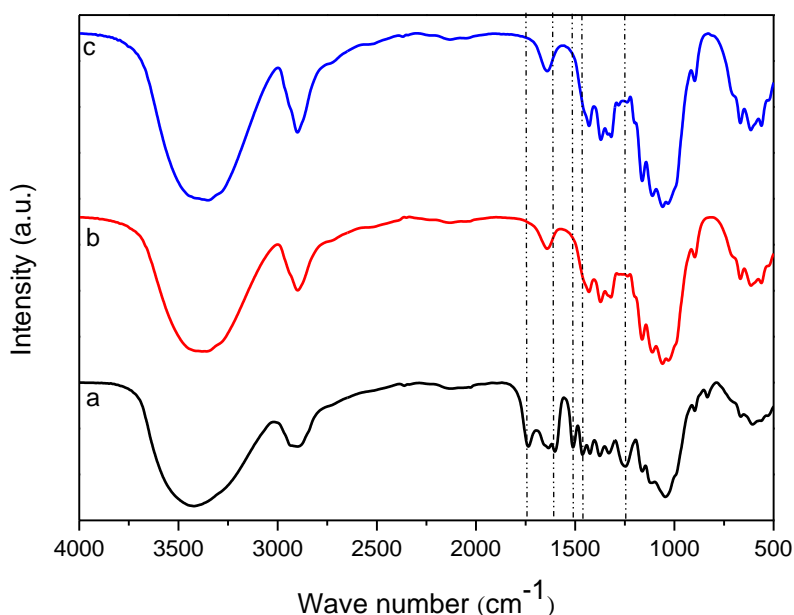
Note: DP of residues were measured after sodium chlorite purification

It is reported that raising the temperature, prolonging the time, or increasing the catalyst content facilitates the breakage of the glycosidic bonds of cellulose chains, which reduces the DP (Adel *et al.* 2011). Cellulose liquefaction in an acidic ethylene glycol was analogous to hydrolysis for the breakage of the glycosidic bonds of cellulose chains

(Jasiukaitytė *et al.* 2009); the DP is reduced to a floor-level value, which is denoted as the leveling-off degree of polymerization (LODP). The LODP refers to the weight-average degree of polymerization of the macromolecules comprising of the discrete, colloidal, and unbonded microcrystals of the polymers (Battista *et al.* 1956). The data in Table 1 shows that higher temperatures and catalyst dosages impacted the cellulose yield and DP. The DP of the cellulose isolated from the liquefaction residue did not change appreciably with time when the liquefaction was conducted with a catalyst dosage of 0.2 mmol/g and at a temperature of 180 °C (Experiments No. 12 through 14). Experiment 13 produced a cellulose with a LODP value. The cellulose product could have been reproducibly prepared under this condition. The cellulose obtained in Experiment 13 was used for the identification of microcrystalline cellulose. The test yielded a violet-blue color, which indicated the existence of MCC. The cellulose samples obtained in this work were also characterized by SEM, FT-IR spectra, and thermal analysis; the results from these analyses were compared to that of a commercial MCC (cotton linters).

### FT-IR Analysis

The FT-IR spectra of the eucalyptus sawdust, MCC from liquefaction residual, and commercial MCC are shown in Fig. 1.



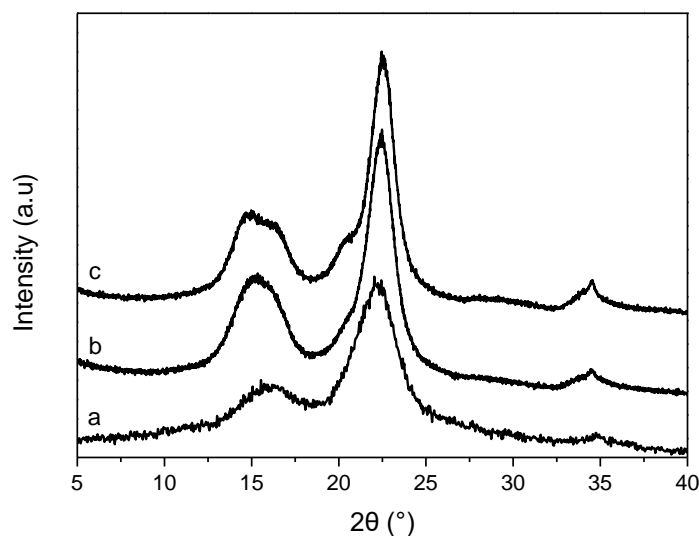
**Fig. 1.** FT-IR spectra of (a) the eucalyptus sawdust, (b) the MCC produced from sawdust, and (c) the commercial MCC (cotton linters)

The broad band at 3400  $\text{cm}^{-1}$  to 3500  $\text{cm}^{-1}$  was assigned to the stretching of the hydroxyl groups, and the absorption at 2900  $\text{cm}^{-1}$  was related to  $\text{CH}_2$  groups. The band at 1731  $\text{cm}^{-1}$  of the eucalyptus sawdust can be attributed either to the acetyl and uronic ester groups of the hemicelluloses or to the carbonyl ester of the *p*-coumeric acid units of lignin (Sun *et al.* 2005; Sain and Panthapulakkal 2006). The bands located at 1510  $\text{cm}^{-1}$ , 1610  $\text{cm}^{-1}$ , and 1436  $\text{cm}^{-1}$  in the sawdust represented the  $\text{C}=\text{C}$  stretch of the aromatic rings of lignin (Sain and Panthapulakkal 2006). The band at 1368  $\text{cm}^{-1}$  and 1256  $\text{cm}^{-1}$  was derived from the methoxyl groups of lignin. In the MCC spectra, these bands did not appear, which

suggested that the MCC did not contain lignin and hemicelluloses. The absorption at  $1640\text{ cm}^{-1}$  in the purified cellulose was indicative of the absorption of water. This band is related to the -OH groups from the water molecules strongly interacting with cellulose (Liu *et al.* 2006). The absorption band at  $1163\text{ cm}^{-1}$  and  $1061\text{ cm}^{-1}$  corresponded to C-O-C stretching, and the peak at  $896\text{ cm}^{-1}$  was associated with the C-H rocking vibration from cellulose; these absorptions appeared in all of the spectra. No differences were found in the spectra of the MCC from sawdust liquefaction and from commercial MCC (*i.e.*, cotton linters). This observation suggested that the reaction to obtain MCC from sawdust removed lignin and hemicelluloses completely, and that the molecular structure of cellulose was not changed.

### XRD Characterization

The X-ray diffraction (XRD) patterns of the eucalyptus sawdust, the MCC from the sawdust, and the MCC from cotton linters (commercial) are presented in Figs. 2(a) through (c). The crystallinity data from the samples are listed in Table 2. From Figs. 2(a) and (b), it is clear that the diffraction peak located at  $22.5^\circ$  became sharper than that of the sawdust, which indicated a higher amount of crystallinity. The crystallinity values were estimated as 58.0% and 79.0% for the eucalyptus sawdust and the MCC produced, respectively. The increase of crystallinity was attributed to the removal of hemicelluloses, lignin, and amorphous cellulose, leading to an increase in the proportion of crystalline regions of cellulose molecules (Haafiz *et al.* 2013). From Figs. 2(b) and (c), the XRD patterns were the same for the sawdust MCC and the commercial MCC; this pattern was typical of cellulose I polymorph with no cellulose II present. The diffractograms showed a peak at  $2\theta = 22.5^\circ$  and a shoulder in the region  $2\theta = 14^\circ$  to  $17^\circ$ . The crystallinity of the commercial MCC was 78.66%. From the XRD results, it was concluded that the purified cellulose that reached the LODP value displayed high crystallinity, which was similar to that of the commercial product (cotton linters).



**Fig. 2.** X-ray diffraction patterns of (a) the eucalyptus sawdust, (b) the MCC produced, and (c) the commercial MCC

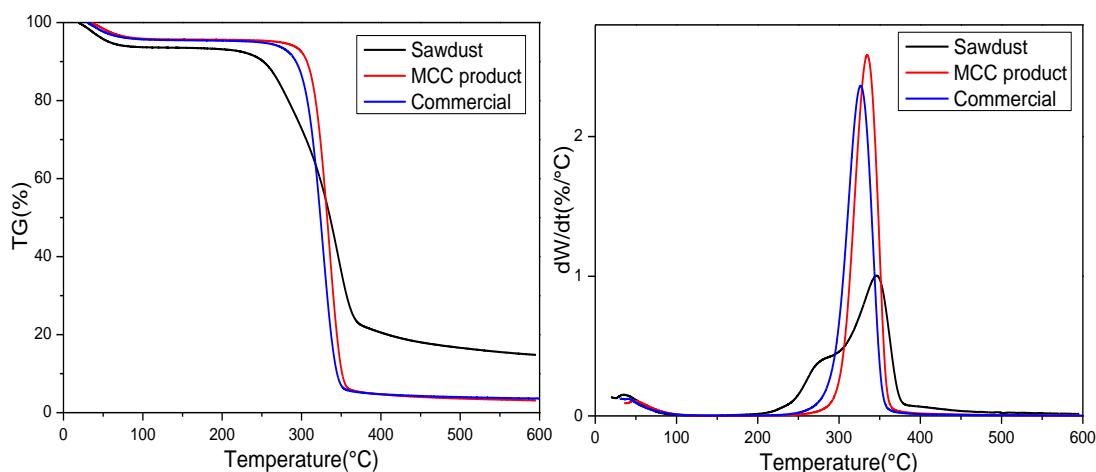
**Table 2.** Crystallinity of the Eucalyptus Sawdust, the MCC Produced, and Commercial MCC

Samples	Crl (%)
Eucalyptus Sawdust	58.95
MCC Produced from Sawdust	78.98
Commercial MCC (Cotton Linters)	78.66

### Thermal Decomposition

Figure 3 shows the TG and DTG curves of the eucalyptus sawdust, the MCC produced from the liquefaction residual, and the commercial MCC (cotton linters). The initial weight loss of the samples (below 100 °C) was due to the vaporization and removal of bound water. The hemicelluloses, lignin, and pectin were easily decomposed by heating. The DTG curve of the eucalyptus sawdust showed an early weight loss that started at approximately 210 °C, and a much wider temperature range of weight loss that occurred from 210 °C to 375 °C. A higher onset of degradation temperature indicated an increase in the thermal stability of the material. The MCC produced from the liquefaction residual and the commercial MCC started to decompose at 300 °C and 285 °C, respectively; these temperatures were higher than the thermal degradation of the eucalyptus sawdust. This demonstrated that the MCC produced from the liquefaction residual had better thermal stability than the MCC from the commercial source.

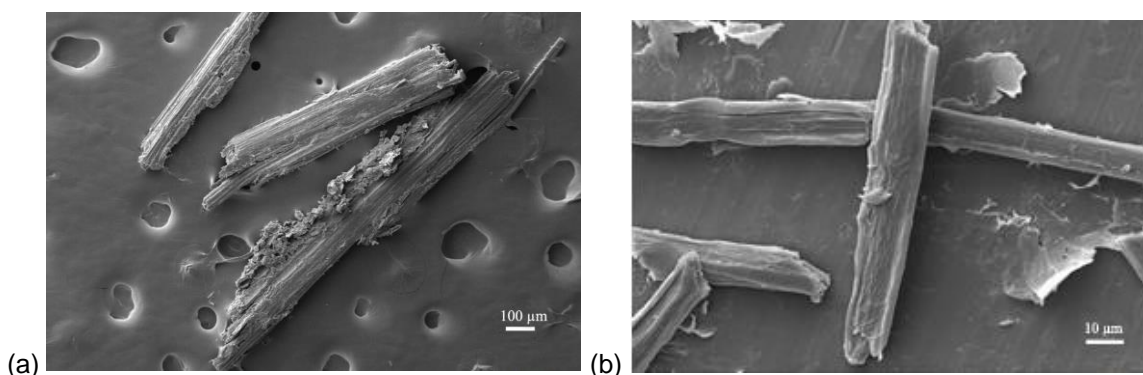
On the other hand, it was observed in Fig. 3 that both cellulose samples had only a relatively narrow weight-loss peak and a lower weight of the thermal decomposition residue than that of sawdust. The narrow weight-loss peak indicated that the composition and chemical structure of the material was relatively simple. According to the references (Hu *et al.* 2007; Yang *et al.* 2006), the pyrolysis of cellulose and hemicellulose usually occurs in the ranges 300 to 400 °C and 200 to 300 °C, respectively. Lignin has a wide pyrolysis temperature range (200 to 700°C) and flat DTG peaks. In addition, the lower weight of the thermal decomposition residue illustrated that the carbon content of the MCC was low. These observations supported the conclusion that nearly all of the hemicelluloses and lignin were removed from the eucalyptus sawdust during liquification, and that the MCC from the residuals were quite pure. Based on the above results, the MCC prepared in this work was thermally stable and could be used in preparing biomass composites.

**Fig. 3.** TGA and DTG thermograms of the eucalyptus sawdust, the MCC produced from liquefaction residual, and the commercial MCC (cotton linters)

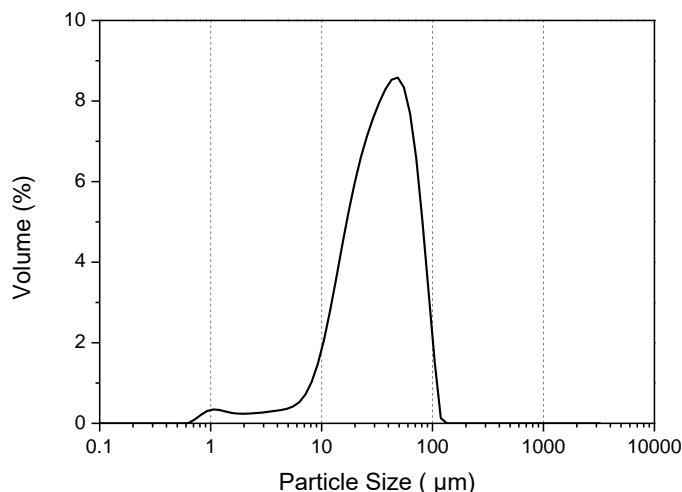


### Morphological Analysis and Particle Size Distribution

The microstructure of the eucalyptus sawdust and the MCC produced from the liquefaction residual was measured by SEM. The SEM images of the eucalyptus sawdust and the MCC prepared from the liquefaction residue were at 100 and 1000 magnifications, respectively. As shown in Fig. 4(a), very well-organized, long, and compact fiber bundles were observed with the eucalyptus sawdust. The size of the MCC was considerably smaller when compared to the sawdust. The MCC from the liquefaction residuals had flat and rigid rod-like morphology with a diameter approximately 10  $\mu\text{m}$ . This observation indicated that the rigid fiber bundles cracked into small irregular fragments during liquefaction. In contrast, the liquefaction reaction could cleave the amorphous regions of the cellulose transversely, which resulted in a diameter reduction of the fibers. The results from the particle size analysis of the MCC produced from liquefaction residuals are shown in Fig. 5. The particle sizes of the purified cellulose ranged from 1  $\mu\text{m}$  to 100  $\mu\text{m}$  with a mean of 38.6  $\mu\text{m}$ .



**Fig. 4.** SEM images of (a) the eucalyptus sawdust, and (b) the MCC produced from the liquefaction residual



**Fig. 5.** Particle size analysis of the MCC produced from the liquefaction residual

## CONCLUSIONS

1. Microcrystalline cellulose (MCC) could be produced from eucalyptus sawdust by controlling liquefaction conditions to afford degraded cellulose with a LODP value.
2. The MCC prepared from eucalyptus sawdust had similar characteristics as commercial microcrystalline cellulose from cotton linters. The chemical structure of the cellulosic fragments was not influenced by liquefaction. The MCC prepared in this study had a high crystallinity index of 79.0% and had good thermal stability.
3. The MCC particles were flat, small, and had a rigid rod-like shape. The particle sizes of the MCC prepared from the sawdust ranged from 1  $\mu\text{m}$  to 100  $\mu\text{m}$  with a mean of 38.6  $\mu\text{m}$ .

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