Optimizing Yield of Microcrystalline Cellulose from Empty Fruit Bunch Via Hydrolysis Using Ionic Liquid

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Abstract. Microcrystalline cellulose (MCC) has been widely recognized as an important material with high potential in contributing to wide range of field including nanocomposite, biomedical applications, electroactive polymers, and many others. MCC has been isolated extensively using conventional method such as acid hydrolysis. However, utilization of hazardous and volatile solvent in acid hydrolysis has raised concern on the impact to the environment and the toxicity level of the as-produced MCC for potential application in biomedical field. In this study, new generation of environmental benign solvent named ionic liquid (IL) was utilized to extract MCC from empty fruit bunch (EFB). The yield of MCC has been optimized by varying three factors, namely temperature, reaction time and solvent concentration. It has been observed that the highest yield of MCC produced using IL is 31.44%, achieved at optimum conditions of 95º C, 129 minutes and 95% solvent concentration. The Fourier transform infrared (FTIR) spectra have confirmed the absence of absorption bands corresponding to lignin and hemicellulose, suggesting that almost pure MCC has been obtained. Scanning electron microscopy (SEM) micrographs have further revealed that the MCC had been agglomerated which resulted in micro-size cellulose ranging from 10 – 20 µm, highly probably due to inappropriate drying method.

1. Introduction

Cellulose, in conjunction with hemicellulose and lignin, is found abundantly in all plant materials. These biopolymers combine and are made up into the main constituent of the biomass termed as lignocellulosic. Cellulose, in particular acts as the utmost plenteous sustainable biopolymer material with high value and availability fits undeniably with the current growing demand in advantage of its biocompatible and environmental friendly properties [1]. Lignocellulosic materials can be isolated from fruit mesocarp, frond, trunk and empty fruit bunch (EFB). Among these varieties of fiber supply available in the tree’s part, EFB is more favourable in terms of its high availability, low price and its potential to yield high percentage of fibers making it possible to substitute the natural fiber resources [2–5]. Being the most abundant biomass materials which are constantly discharged as waste from the palm oil mill, efforts to extract the high value lignocellulosic materials as value-added products have gained increasing interest [5]. Being the most significant lignocellulosic material in EFB, cellulose is made up of adjoining glucose units bonded to each other by β-1,4-glycosidic bonds [6]. Microfibrils which are the linear cellulose chains bundled together are composed of amorphous and crystalline regions. Cellulose is recognized as an important renewable resource with wide applications. However,
since cellulose comprised of a straight glucose polymer alignment which forms a highly strong supramolecular arrangement due to hydrogen bonding, it is highly resistant to enzymatic degradation. From a greener perspective, designing an effective and environmental friendly cellulose hydrolysis method is highly desired [6–8].

Although cellulose is the most accessible polymer in nature, it is only rather recently that cellulose has attained notability as micro and nanomaterial, in form of microcrystalline cellulose (MCC) and nanocrystalline cellulose (NCC) [9]. MCC and NCC are commonly prepared via conventional acid hydrolysis. However, the use of conventional acids poses several problems such as producing potential hazardous organic compounds which led to environmental pollution. Hence, ionic liquid (IL) is proposed as an environmental benign and highly efficient solvent for the preparation of crystalline cellulose, particularly MCC [6–11]. In this study, one type of IL named 1-butyl-methylimidazolium chloride (BmimCl) is used to investigate the potential of IL in the preparation of MCC from EFB. In addition, the optimization study to determine the optimum conditions for the preparation of MCC is also conducted in this work. The effects of three process parameters namely temperature, reaction time and solvent concentration on the yield of MCC are investigated with the aid of Design of Experiment (DOE) software. Subsequently, the physical properties of raw EFB, treated EFB and the as-produced MCC are determined using Fourier transform infrared (FTIR) spectroscopy and scanning electron microscopy (SEM).

2. Materials and methods

EFB was collected from Felda Palm Industries Sdn. Bhd., Kulai, Malaysia. BmimCl used in this study was acquired from Shanghai Chong Jie Chemical Co. Ltd., China while sodium hydroxide (NaOH) was obtained from Sigma-Aldrich, Malaysia. All chemicals were used as received.

2.1. Pre-treatment of EFB

EFB was first sun-dried for 5 days and then was cut into smaller pieces. The EFB was further dried in an oven at 80°C overnight before it was ground into smaller size using a grinder. Next, 50 g of the ground EFB was pretreated using 500 mL of 2% (w/v) NaOH solution. The pretreatment process was carried out at 80°C for 2 hours while continuously stirred. Then, the mixture was left to cool down before it was washed with distilled water to remove the NaOH solution [12]. The pretreated EFB was subsequently dried at 60°C for 8 hours and kept for further used.

2.2. Hydrolysis of EFB using IL

Hydrolysis of EFB was carried out by treated the EFB at different operating conditions using BmimCl. Three process parameters (i.e. temperature, reaction time and solvent concentration) were investigated to optimize the yield of MCC from EFB. For each designated run, 60 mL of desired concentration of BmimCl was first heated to its desired temperature using a heating plate (Model 11-102-50SH, Fisher Scientific, USA). 3 g of pretreated EFB were then added to the BmimCl. The mixture was stirred continuously at a designated temperature for a specific reaction time. Afterwards, the cold water was added to the mixture to stop the reaction. Next, the mixture was diluted with distilled water and then was centrifuged at 8000 rpm for 10 minutes to extract the MCC. MCC suspension was collected during the centrifugation step until no more cloudy suspension was obtained. The MCC suspension was dried in an oven at 80°C overnight to obtain MCC powder. The as-produced MCC powder was then weighed for yield measurement and kept in a desiccator for further characterization. The yield of MCC was calculated using equation (1) as shown below.

\[
\text{Yield} (\%) = \frac{\text{Weight of dried MCC}}{\text{Initial weight of treated EFB}} \times 100 \tag{1}
\]
2.3. Optimization of Yield of MCC

2.3.1. Preliminary Tests
Preliminary test was conducted to determine the range of each process parameter to be used for optimization study. Ranges selected for preliminary test for the effect of temperature, reaction time and solvent concentration were 60°C – 130°C, 40 min – 130 min and 40% - 100%, respectively. In this test, one-factor-at-a-time (OFAT) method was used.

2.3.2. Design of Experiment (DOE)
The smaller ranges of the process parameters were determined from the preliminary tests and subsequently selected to be used in the optimization study using Design of Experiment (DOE). In this study, Central Composite Design (CCD) under Response Surface Methodology (RSM) was used to investigate the optimum process parameters for the preparation of higher yield of MCC.

2.4. Fourier Transform Infrared (FTIR) Spectroscopy
The change of functional groups of raw EFB, treated EFB and the as-produced MCC was recorded using a FTIR spectrometer (Model Spectrum 65, Perkin Elmer). All samples were mixed with potassium bromide (KBr) powder with weight ratio of 1:100 and subsequently compressed into a disc. The spectra for each sample were recorded as an average of 64 scans at a resolution of 4 cm\(^{-1}\) ranging from 500 cm\(^{-1}\) to 4000 cm\(^{-1}\) [13,14].

2.5. Scanning Electron Microscopy (SEM)
The surface morphology of raw EFB, treated EFB and MCC was analysed using a scanning electron microscope (Model JSM-6100, Jeol). All samples were gold coated and then mounted in the microscope using conductive carbon tape. All samples were ensured to be fully dried before scanned at accelerating voltage of 10kV.

3. Results and discussion

3.1. Preliminary Tests
The yield of MCC from preliminary tests using OFAT for different process parameters (i.e. temperature, reaction time and solvent concentration) were shown in Figure 1. Preliminary tests were carried out to determine the smaller range of process parameter prior to the optimization study. From Figure 1(a) – (c), the selected range for optimization study for temperature, reaction time and solvent concentration are 80°C – 120°C, 90 min - 130 min and 55% - 95%, respectively. For all three process parameters, it can be observed that the increase of temperature, reaction time and solvent concentration led to the increasing yield of MCC until it reached a maximum value. This is highly probably due to more amorphous region have been removed by the reaction between IL and cellulose fibres. However, as all three parameters were further increased, the MCC yield started to decline as the fibres have further broken down into soluble sugar.

![Figure 1. Yield of MCC from preliminary tests using OFAT for different process parameter, (a) temperature, (b) reaction time, and (c) solvent concentration.](image-url)
3.2. *Optimization of Yield of MCC*

The optimization study was conducted using DOE by applying the results from the preliminary tests. For simplicity reason, the three process parameters (i.e. temperature, reaction time and solvent concentration) investigated in the optimization study were denoted as A, B and C, respectively. The yield of MCC prepared at different designated conditions is shown in Table 1. The highest MCC yield (31.33%) was obtained at conditions of 100°C, 110 min and solvent concentration of 95% (run 2). Meanwhile, the lowest MCC yield (3.33%) was recorded from run 3, where the MCC was prepared at conditions of 120°C, 90 min and solvent concentration of 55%.

### Table 1. Yield of MCC prepared at different designated conditions.

| Run | Process Parameter | Response |
|-----|-------------------|----------|
|     | Temperature, A (°C) | Reaction time, B (min) | Solvent concentration, C (%) | Yield (%) |
| 1   | 80                | 90        | 95        | 18.33     |
| 2   | 100               | 110       | 95        | **31.33** |
| 3   | 120               | 90        | 55        | 3.33      |
| 4   | 100               | 90        | 75        | 9.33      |
| 5   | 100               | 110       | 75        | 14.00     |
| 6   | 100               | 110       | 75        | 13.00     |
| 7   | 120               | 90        | 95        | 14.33     |
| 8   | 120               | 110       | 75        | 7.67      |
| 9   | 80                | 110       | 75        | 7.00      |
| 10  | 80                | 130       | 95        | 30.67     |
| 11  | 80                | 90        | 55        | 3.67      |
| 12  | 100               | 130       | 75        | 10.00     |
| 13  | 100               | 110       | 75        | 11.00     |
| 14  | 80                | 130       | 55        | 4.00      |
| 15  | 100               | 110       | 75        | 9.67      |
| 16  | 120               | 130       | 55        | 5.33      |
| 17  | 120               | 130       | 95        | 22.33     |
| 18  | 100               | 110       | 75        | 11.33     |
| 19  | 100               | 110       | 55        | 6.33      |
| 20  | 100               | 110       | 75        | 9.00      |

3.2.1. *Analysis of Variance (ANOVA)*

The analysis of variance (ANOVA) was applied in DOE to measure the precision of the model obtained from the results of the experiments. The statistical data of the variation of mean square ratio and mean square residual error was shown in Table 2. R-squared represents the determination coefficient which denotes the square of correlation coefficient for the response. In this study, the R-squared obtained is 0.9504. The closer the R squared value to 1, the better the correlation between the experimental and the predicted values. Hence, the model will predict the response and fits the data better. The adjusted R-squared value is 0.9058 while the predicted R-squared value is 0.6538. Although the predicted R-squared value is not as close to the adjusted R-squared value, the predicted R-squared value is considered to be in reasonable agreement with the aforementioned adjusted R-squared value.
### Table 2. Statistical data for ANOVA.

| Parameter   | Value  | Parameter          | Value  |
|-------------|--------|---------------------|--------|
| Std. Dev.   | 2.47   | R-Squared           | 0.9504 |
| Mean        | 12.08  | Adj R-Squared       | 0.9058 |
| C.V. %      | 20.48  | Pred R-Squared      | 0.6538 |
| PRESS       | 427.58 | Adeq Precision      | 15.7360|

Meanwhile, the adequate precision measures the signal to noise ratio. An adequate precision value of more than 4 is desirable so that the effective determination of the model could be obtained [15]. The adequate precision was determined to be 15.7360, which implies an adequate signal and validating the model precision. The standard deviation value of 2.47 which is relatively low value shows that the model has a good precision and lower dispersion. Whilst the coefficient of variation (C.V.) value obtained is 20.48%. This value represents the ratio of standard deviation to mean and expresses precision and repeatability. The low value of C.V. shows that the factors are highly reliable during the optimization.

The result of ANOVA for response surface quadratic mode is shown in Table 3. The ANOVA result indicates that the investigated process parameters are significantly affecting the yield of MCC, as implied by the model F-value of 21.30. There is only 0.01% (p-value) possibility that the F-value of a model this large could occur due to noise. The result also revealed that the model terms were significant. The individual effect of solvent concentration (C) is influencing the yield of MCC the most, as shown by high F-value of 145.33. In addition, B, C, BC, A2 and C2 were also concluded as significant model terms, in which all their p-values are less than 0.1000. The F-value of lack of fit of 2.36 implies that the lack of fit is not significant relative to the pure error value of 18.21. There is an 18.36% possibility that the F-value of lack of fit this large could occur due to noise. Non significant lack of fit is desirable as it shows that the model fits well.

### Table 3. ANOVA for response surface quadratic mode.

| Source       | Sum of Square | df  | Mean Square | F-Value | p-value (Prob>F) |
|--------------|---------------|-----|-------------|---------|-----------------|
| Model        | 1174.01       | 9   | 130.45      | 21.30   | < 0.0001        |
| A-Temperature| 11.41         | 1   | 11.41       | 1.86    | 0.2022          |
| B-Reaction time | 54.48   | 1   | 54.48       | 8.90    | 0.0137          |
| C-Solvent concentration | 889.81 | 1   | 889.81      | 145.33  | < 0.0001        |
| AB           | 0.89          | 1   | 0.89        | 0.15    | 0.7108          |
| AC           | 22.21         | 1   | 22.21       | 3.63    | 0.0860          |
| BC           | 40.55         | 1   | 40.55       | 6.62    | 0.0277          |
| A^2          | 47.34         | 1   | 47.34       | 7.73    | 0.0194          |
| B^2          | 9.10          | 1   | 9.10        | 1.49    | 0.2508          |
| C^2          | 148.40        | 1   | 148.40      | 24.24   | 0.0006          |
| Residual     | 61.23         | 10  | 6.12        | 2.36    | 0.1836          |
| Lack of Fit  | 43.02         | 5   | 8.60        | 2.36    | Not significant |
| Pure Error   | 18.21         | 5   | 3.64        |         |                 |
| Cor Total    | 1235.24       | 19  |             |         |                 |

#### 3.2.2. Response Surface Analysis

The interaction and relationship between process parameters and the response (MCC yield) can be indicated by three dimensional (3D) contour plots. Figure 2 portrays 3D contour view for the effects of different combination of process parameters on MCC yield. From Figure 2(a), the combined effect of temperature and process time is less significant on the MCC yield, as represented by blue and green region showing less significant response. Meanwhile, a saddle shape was observed in Figure 2(b) which represented the combined effect of temperature and solvent concentration on MCC yield. The
The aforementioned shape was observed as the solvent concentration is more significantly affected MCC yield as compared to the temperature. Similar saddle shape also has been observed in Figure 2(c), in which solvent concentration also shown superior effect on MCC yield as compared to the reaction time. The result indicates that reasonably high MCC yield could be prepared by employing high solvent concentration coupled with moderate values of temperature and reaction time.  

**Figure 2.** 3D contour view for the combined effects of (a) temperature and reaction time, (b) temperature and solvent concentration, and (c) reaction time and solvent concentration on MCC yield.

### 3.2.3. Optimization Validation

In order to validate the model, the DOE proposed a solution run with the predicted maximum yield of MCC (Table 4). The suggested optimum run was carried out for three times and the average percentage error between actual and predicted yield was calculated and recorded. The average percentage error (3.15%) was revealed to be less than 5%, thus the predicted model was realistically accepted.

**Table 4.** Experimental validation for the optimization of the yield of MCC.

| Solution no. | Temperature, A (°C) | Reaction time, B (min) | Solvent concentration, C (%) | Predicted yield (%) | Actual yield (%) | Percentage error (%) |
|-------------|---------------------|------------------------|-----------------------------|--------------------|------------------|----------------------|
| 1           | 95                  | 129                    | 95                          | 31.37              | 32.00            | 2.02                 |
| 2           | 95                  | 129                    | 95                          | 31.37              | 32.33            | 3.07                 |
| 3           | 95                  | 129                    | 95                          | 31.37              | 30.00            | 4.36                 |
| **Average** |                     |                        |                             | **31.44**          |                  | **3.15**             |

### 3.3. FTIR Analysis

FTIR spectra of raw EFB, treated EFB and the as-produced MCC are shown in Figure 3. All three spectra show two main absorbance regions which are the low wavelength region at 500 cm\(^{-1}\) to 1750 cm\(^{-1}\) and the high wavelength region ranging from 2800 cm\(^{-1}\) to 3500 cm\(^{-1}\). The absorption peaks found in all spectra in the range of 3400 cm\(^{-1}\) to 3450 cm\(^{-1}\) are attributed to the hydroxyl groups which are one of the functional group found in lignocellulosic materials [16]. On the other hand, the peaks found in the range of 2850 cm\(^{-1}\) to 2930 cm\(^{-1}\) are attributed to the aliphatic saturated C–H stretching vibrations in cellulose, hemicellulose and lignin [17]. In comparing the raw EFB spectrum with both treated EFB and MCC spectra, the most noticeable difference is the absence of the vibration peak at 1737.39 cm\(^{-1}\). This peak is associated with the C=O stretching of the methyl ester and carboxylate groups in pectin and the acetyl and uronic ester groups in hemicelluloses. It may also be caused by the ester linkage of carboxylic group of the ferulic and p-coumaric acids of lignin and/or hemicelluloses [18]. Hence, the disappearance of this peak indicates that the hemicellulose and lignin components have been effectively removed post alkaline treatment.
On the other hand, the peaks present at 1630 cm\(^{-1}\) to 1645 cm\(^{-1}\) correspond to the O-H bending due to absorption of water by cellulose [19]. This peak is found in both treated fibres probably due to reaction of sodium hydroxide with the hydroxyl groups of celluloses and hence subsequently lead to formation of water molecules. The same vibration peak is also found at 1642.25 cm\(^{-1}\) of untreated fibres. This shows that even though the fibres were subjected to drying process, the water adsorbed in the cellulose molecules was very difficult to be extracted due to the cellulose-water interaction [20]. The peaks at 1500 cm\(^{-1}\) to 1600 cm\(^{-1}\) are due to C=C aromatic skeletal vibration of lignin [21]. This can be seen in the spectra of raw EFB at 1550 cm\(^{-1}\) indicating the presence of lignin. Peaks in this range however do not appear in both treated EFB and MCC spectra. This indicates that lignin has been removed post alkaline and IL treatment. The vibration band in the range of 1450 cm\(^{-1}\) to 1475 cm\(^{-1}\) could be identified as the bending vibration of CH\(_2\) group in aliphatic compounds and is found in all spectra. The peak observed in all of the spectra at 1030 cm\(^{-1}\) to 1060 cm\(^{-1}\) is due to the C-O stretching assignment of primary alcohol [21]. The increase in the intensity of this band indicates an increase in crystallinity of the samples. Another significant absorption band, which increases continuously with alkaline and IL treatment is the peak at 897 cm\(^{-1}\) which corresponds to glycosidic –C-H- deformation, with a ring vibration contribution and –O-H bending. This peak features the characteristic of the β-glycosidic linkage between the anhydroglucose units in cellulose [22]. The deformation, wagging and twisting modes of anhydro-glucopyranose vibration is shown from 600 cm\(^{-1}\) to 750 cm\(^{-1}\) [23].

3.4. SEM Analysis

SEM micrographs of raw EFB, treated EFB and MCC at different magnification are shown in Figure 4. Raw and rough surface morphology has been observed for the raw EFB sample while the treated EFB sample shows smooth and holes-like surface. The holes-like surface of the treated EFB was observed highly probably due to successful removal of lignin and hemicellulose post alkaline treatment. Whereas the SEM micrographs of the as-produced MCC at 1000 and 5000 magnifications are portrayed in Figure 4(c) and (d), respectively. The MCC produced are made up of the crystalline regions from the cellulosic fibres after the removal of amorphous regions at the cellulose polymer. The MCC sample displays a rigid and conglomerate texture with size ranging from 10 – 20 µm, in which the fibers are fused together giving a relatively homogeneous macrostructure. It is anticipated that smaller particles in nanosize could be prepared by applying more appropriate drying approach such as freeze drying method.
4. Conclusion
Successful production of MCC from EFB could be achieved via hydrolysis using IL. The highest yield of MCC (31.44%) can be prepared at optimized conditions of 95°C, 129 minutes and at solvent concentration of 95%. Almost pure MCC is prepared as lignin, hemicellulose and other non-cellulosic components have been successfully removed from the raw EFB, as shown by FTIR spectra. SEM analysis also have revealed that the as-produced MCC shown rigid and conglomerate structure with size ranging from 10 – 20 µm. As more environmental-friendly approach attracts ever-increasing attention in the field of nanomaterial production, IL has shown remarkable potential to be used as environmental benign solvent for the preparation of micro- and nano-size cellulose.

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