Carbohydrate Polymers 148 (2016) 11–20

Contents lists available at ScienceDirect

Carbohydrate Polymers

journal homepage: www.elsevier.com/locate/carbpol

Characterisation of microcrystalline cellulose from oil palm fibres for food applications

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ARTICLE INFO

Article history:
Received 14 January 2016
Received in revised form 11 April 2016
Accepted 12 April 2016
Available online 14 April 2016

Keywords:
Microcrystalline cellulose
Oil palm fibres
Physicochemical properties
Rheology

ABSTRACT

Microcrystalline cellulose (MCC) extracted from empty fruit bunches (EFB), stalk and spikelet were characterised through physicochemical and microstructure analyses. Raw stalk fibres yielded the highest cellulose content (42.43%), followed by EFB (32.33%) and spikelet (18.83%). Likewise, lowest lignin and residual oil content was reported in raw stalk fibres compared to EFB and spikelet. SEM revealed significant changes on fibres' surface morphology throughout the extraction process. FTIR analysis showed that main characteristic peaks of hemicellulose and lignin was absent on the extracted MCC. The crystallinity index for MCC extracted from EFB (82.5%), stalk (82.2%) and spikelet (86.5%) was comparable to commercial MCC (81.9%). Results suggested stalk fibres is more preferable for the production of MCC compared to EFB and spikelet. Further rheological studies showed viscoelastic behaviour with no significant differences between commercial and stalk-based MCC, while modelling work showed ability to simulate complex deformation of the MCC-hydrogel/food mixture during processing/handling stage.

1. Introduction

Microcrystalline cellulose (MCC) is one of the cellulose derivatives that is described as purified, partially depolymerised cellulose, prepared by treating α-cellulose from fibrous plant material with mineral acids (Thoorens, Krier, Leclercq, Carlin, & Evrard, 2014). It is characterised with high degree of crystallinity, which values are typically in the range from 55% to 80% (Chuayjuljit, Su-uthai, & Charuchinda, 2010). MCC gained major interest in various applications, such as stabilizer, fat replacer and texturing agent in food industry (Singh, Kanawjia, Giri, & Khetra, 2015), binder and water retainer in pharmaceutical industry (Johansson & Alderborn, 2001), and reinforcing agent in plastic industry (Wittaya, 2009).

Considering Asian countries like Malaysia produced large amounts of oil palm biomass, extraction of MCC from oil palm fibres has gained popular. Empty fruit bunches (EFB) is the largest solid waste produced from the palm oil mills after the fresh palm fruits are processed to obtain oil. EFB is made up of two major parts, namely stalk and spikelet. Even though a number of studies were conducted to obtain MCC from oil palm fibres (Mohamad Haafiz, Eichhorn, Hassan, & Jawaid, 2013; Mat Soom, Abd Aziz, Wan Hassan, & Md Top, 2009; Ramli, Junadi, Beg, & Yunus, 2015), these works mainly focused on EFB as a whole, without specific investigation on different parts of oil palm EFB (*i.e.* stalk and spikelet). In addition, the works by Mohamad Haafiz et al. (2013), Nazir, Wahjoedi, Yussof, and Abdullah (2013) and Ramli et al. (2015) concentrated on the extraction methods of MCC production from EFB (though acid hydrolysis, formic acid-hydrogen peroxide, and ultrasonic-alkali treatment respectively), without much emphasis on the industrial application of the MCC produced (*e.g.* food and pharmaceutical applications). It is likely MCC extracted from different parts of a lignocellulosic source can influences physicochemical properties such as crystallinity, thermal stability, porous structure, surface area and moisture content. This can likely affect the MCC rheological behaviour when added as bio-filler in food (*e.g.* ice cream and mayonnaise) or pharmaceutical products (*e.g.* skin cream and tablets).

Therefore the aim of this work is to characterise MCC obtained from the different components of oil palm fibres (*i.e.* EFB, stalk and spikelet) through physicochemical and microstructure analyses. From the analyses results, we then proposed a type of oil palm fibres and conduct rheological testing and modelling. The production of MCC from oil palm fibres are conducted using chemical extraction method (details provided in Section 2.2), whereas the rheological testing is performed by adding different mass percentages of the MCC into hydrogel. Note that the analyses of the MCC are compared to investigate whether the MCC produced here are comparable to the commercial ones.

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http://dx.doi.org/10.1016/j.carbpol.2016.04.055
0144-8617/

2. Materials and methods

2.1. Materials

Empty fruit bunches (EFB) were collected after threshing process from Besout Palm Oil Mill in Perak, Malaysia, and were kept at −20 °C to prevent fungal contamination. The EFB were then separated manually into stalk and spikelet, before being washed with tap water and a 2% of detergent solution to remove residual oil and dust, before being dried in an oven at 60 °C for 24 h. The dried EFB, stalk and spikelet were cut into 0.2 mm by using a power cutting mill (Retsch, SM 200, Germany). A commercial available MCC (Catalogue no. 435236, Sigma Aldrich, USA) was used as reference, which is denoted as commercial microcrystalline cellulose (CMCC).

2.2. Production of microcrystalline cellulose

The microcrystalline cellulose (MCC) was obtained from the EFB following the method by Mat Soom et al. (2009). Ten different EFB samples were mixed and treated with 0.7% (w/v) sodium chlorite ($NaClO_2$) solution (collected after the threshing process), where the acetic acid was added to acidify the $NaClO_2$ solution until the pH reached the scale of 4. The fibres were then heated in acidified $NaClO_2$ for 2 h at 70–80 °C, with a fibre to $NaClO_2$ ratio of 1:50 (g/ml^{-1}). The process was repeated five times, before the residues were washed several times with distilled water until the yellowish colour and odour from the chlorine oxide were removed. The samples were then dried at 60 °C overnight. After that, the bleached fibres were treated with 17.5% (w/v) sodium hydroxide (NaOH) solution (with the fibre to NaOH solution ratio of 1:25 (g/ml^{-1})) for 2 h. The mixture was then filtered, washed and dried in an oven at 60 °C for 24 h, yielding α-cellulose fibres. The MCC was produced by hydrolysing the α-cellulose fibres in 55% (w/w) sulphuric acid solution for 45 min at 45 °C under constant stirring, after which large volume of cooled distilled water was added to stop the reaction. The diluted suspension was then washed and filtered until the pH reached the scale of 5 before dialysis was conducted until the pH became constant. Similar procedures were applied to produce the MCC from oil palm stalk and spikelet.

2.3. Lignocellulosic compositions

The cellulose, hemicellulose and lignin contents were determined via neutral detergent fibre (NDF), acid detergent fibre (ADF), and acid detergent lignin (ADL) method analyses as described by Mohammed, Salmiaton, Wan Azlina, & Mohamad Amran (2012). The NDF content was measured by refluxing the sample in a boiling neutral reagent for one hour. Then, the solution was cooled and filtered. The residues were washed with distilled water and acetone, before being dried. The ADF analysis is similar to the NDF, but different detergent solution was used (acid detergent solution). The ADL content was then determined by hydrolysing the residual fibres from the ADF analysis with 72% (w/v) sulphuric acid. The fibres were washed, dried, before being ignited in a furnace. The percentage of NDF, ADF, and ADL were calculated from the initial and final weight difference. The percentage of cellulose, hemicellulose and lignin were then computed using the following equations:

$$\text{Cellulose}(\%) = \text{ADF} - \text{ADL} \quad (1)$$

$$\text{Hemicellulose}(\%) = \text{NDF} - \text{ADF} \quad (2)$$

$$\text{Lignin}(\%) = \text{ADL} \quad (3)$$

2.4. Determination of residual oil content

The oil content in the EFB, stalk and spikelet were determined using the Soxtec extraction method (Soxtec™ 2050, Foss Analytical, Denmark). A sample size of 1 g was weighed into a labelled extraction thimble for each extraction process. A defatted cotton plug was placed on top of each sample to keep the sample immersed during the boiling step and to prevent any sample loss from the top of the thimble. The clean extraction cups were dried in an oven at 105 °C for 30 min, before being cooled in a desiccator. Then, approximately 80 mL of the hexane solvent that was used to extract the oil residues in the samples was added to each weighed extraction cup. The Soxtec extraction unit was assembled and was allowed to reflux for 75 min. Once the extraction process was completed, the extraction cup was removed and the sample were dried at 105 °C for one hour to remove the residual hexane solvent and was cooled in a desiccator, before being weighed. The oil content present in the sample was calculated using:

$$\text{Oil content}(\%) = \frac{\text{Thimble with oil(g)- Thimble(g)}}{\text{Sample(g)}} \times 100\% \quad (4)$$

2.5. Scanning electron microscope (SEM)

The surface morphology changes of EFB, stalk and spikelet throughout MCC extraction processes were observed through a scanning electron microscopy (E-1010, Hitachi, Japan). The sample in the range of 2–5 mm was mounted on an aluminium stub and sputter-coated with platinum prior to the morphological assessment. An accelerating voltage of 15–25 kV was used throughout the scanning process.

2.6. Fourier transmission infrared (FTIR)

FTIR spectra of EFB, stalk and spikelet throughout the MCC extraction processes were obtained using Fourier transform infrared spectroscopy (Nicolet 6700, USA) to determine the chemical changes caused by the extraction processes. Scanning was conducted in a range of 500–4000 cm^{-1} wavenumber, at a resolution of 4 cm^{-1} with a total of 32 scans for each sample.

2.7. X-ray diffraction (XRD)

The crystallinity of the samples was determined with an X-ray diffractometer (Italstructures APD 2000, Italy) using a CuK_α radiation. The sample was scanned and the intensity was recorded in a 2θ range from 5° to 30°. The crystallinity index (CrI) of the sample was calculated as follows (Segal, Creely, Martin, & Conrad, 1959):

$$CrI = \frac{(I_{002} - I_{am})}{I_{002}} \times 100 \quad (5)$$

where, I_{002} is the maximum intensity of the (002) lattice diffraction at 2θ between 22° and 23° for cellulose I, and between 18° and 22° for cellulose II, while I_{am} is the minimum intensity of lattice diffraction attributed to amorphous cellulose at 2θ between 18° and 19° for cellulose I, and between 13° and 15° for cellulose II (Roncero, Torres, Colom, & Vidal, 2005).

2.8. Preparation of MCC water gel-like suspension

MCC hydrogels with different concentrations (10%, 12%, 14%, 16%, and 18% (w/v)) were prepared in distilled water. The mixture was then sonicated for three minutes using a high intensity ultrasonic disruptor (FB-705, Fisher Scientific, UK) (Mihranyan, Edsman, & Strømme, 2007), to produce a gel-like sample. Upon production