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Isolation and characterization of microcrystalline cellulose from pomelo peel

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ABSTRACT

In this study, cellulose from pomelo peel (PP-C) was isolated using alkaline hydrogen peroxide liquor extraction, and PP-C was further hydrolyzed by hydrochloric acid to yield microcrystalline cellulose (PP-MCC). Several analytical methods were used to determine the structural characteristics, crystalline behavior, morphological properties, thermal properties, and water/oil binding capacity of PP-C and PP-MCC. Fourier-transform infrared spectra and morphological analysis showed that the alkaline hydrogen peroxide treatment and acid hydrolysis successfully removed hemicellulose and lignin from pomelo peel fibers. Both PP-C and PP-MCC had only a cellulose I polymorph structure, and the crystallinity index of PP-MCC increased compared with that of PP-C. The degradation temperatures of PP-C and PP-MCC were approximately 257 °C and 280 °C, respectively. The water/oil binding capacity of PP-MCC was significantly higher than that of the commercial MCC. Our results thus suggested that PP-MCC might be suitable for use as a food stabilizer and pharmaceutical additive.

1. Introduction

Pomelo (*Citrus grandis* (L.) Osbeck), also known as pummelo, shaddock or Chinese grapefruit, is widely cultivated and consumed worldwide due to its flavor and unique taste [1–3]. It has been reported that pomelo contains large amounts of fiber, pectin, carotenoids, flavonoids, limonoids, vitamin E, and essential oils [4]. These bioactive components are related to pomelo's pharmacological activity that has been shown to convey anti-oxidation, anti-inflammation, and anti-diabetes health benefits [5, 6].

During the processing of pomelo fruit for juice, jam or canning products, the pomelo peel (PP) represents about 50% of the total weight of the fruit and is the primary byproduct [7, 8]. However, most of the PP is disposed of and has little economic value [9]. PP is inedible to humans and is thus discarded in landfills where it releases carbon dioxide along with noxious gases resulting in environmental pollution. Consequently, there has been considerable interest in the search for alternative disposal methods for PP. PP contains many natural chemical components, such as cellulose, flavonoids, essential oils, pectin etc. that make it a good source for valuable extracts [10]. As a result, high value-added utilization of PP is a subject of significant research interest.

For example, pectin isolated from PP has been used in the manufacture of low-sugar products such as low-sugar jam and jelly [9, 11–13]. Essential oils from PP have widespread applications as aromatic flavor in many food products and in the pharmaceutical industry [4]. Li et al. prepared a type of porous carbon using carbonizing PP as an adsorbent material for the removal of methyl orange [14]. A novel family of biomass-based carbon aerogels derived from PP for the removal of organic pollutants/oils was reported by Zhu et al. [7].

Several methods for fabricating simple and effective oil adsorption materials using botanic cellulose have been reported [15, 16]. PP is comprised primarily of cellulose and hemicellulose, which are responsible for the moisture uptake and high oil sorption capacity of PP [17]. The isolation of microcrystalline cellulose from plant cellulose and applications of this result are also areas of research interest. Microcrystalline cellulose is conventionally prepared by treating alpha cellulose with an excessive amount of mineral acids [18]. However, the isolation and characterization of microcrystalline cellulose from pomelo peel has never been described. The objective of this study was to extract cellulose from pomelo peel (PP-C) and to further use PP-C to prepare microcrystalline cellulose (PP-MCC). Multiple analysis techniques such as SEM, FTIR, XRD and TG were used to investigate the characteristics of PP-MCC, and the water/oil holding ability of PP-MCC was also determined.

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2. Materials and methods

2.1. Materials

Pomelo peel was obtained from a local supermarket in Wuhan, China, and then dried and ground into powder. Analytical grade chemicals and solvents were used in this study.

2.2. Extraction of cellulose from pomelo peel

Cellulose extraction procedures were based on reported methods [19, 20] with modification. Pomelo peel powder was treated with a 4% (w/w) sodium hydroxide solution containing 0.9% (v/v) of hydrogen peroxide at 80 °C for 4 h. The solution was then filtered and washed with distilled water until neutral pH to obtain crude cellulose from the pomelo peel (PP-CC). The PP-CC was then subjected to a rapid purification treatment with a mixture of 80% acetic acid-68% nitric acid (v/v = 10:1) at 100 °C for 15 min to remove resistant hemicellulose and lignin associated with the cellulose [21]. Finally, the cellulose was freeze-dried using a freeze drier (Betr 2-8 LD plus, Christ, Germany) for 48 h to obtain purified cellulose from the pomelo peel (PP-C).

2.3. Preparation of PP-MCC

PP-C was used to prepare PP-MCC by the hydrochloric acid hydrolysis method. In a reactor, 1 g of PP-C was mixed with 20 ml of an aqueous solution of 6% (w/w) hydrochloric acid in a 90 °C water bath for 100 min [22]. Next, the solution was filtered and washed with distilled water until it reached a neutral pH. Finally, the retentate was freeze-dried to obtain PP-MCC.

2.4. Fourier-transform infrared spectra (FT-IR) analysis

The dried samples of PP-C, PP-MCC and commercial microcrystalline cellulose (C-MCC) were mixed with spectroscopic-grade potassium bromide powder and then ground and pressed into pellets for the FT-IR measurement. FT-IR spectra were recorded using a Nexus 470 FT-IR spectrometer (Thermo Nicolet, USA) with a frequency range of 4000–400 cm^{-1}.

2.5. Morphological analysis

The morphologies of the PP-C, PP-MCC, and C-MCC were observed by a Scanning Electron Microscope (SEM) using a JSM-6390LV. Samples were coated with gold before observation.

2.6. X-ray diffraction (XRD) analysis

The crystalline structures of PP-C, PP-MCC, and C-MCC were analyzed using an X-ray diffractometer (D8 Advance, Bruker) equipped with Cu-Kα radiation (λ = 0.154 nm) in the range of 2θ from 5 to 80°. The operating voltage was 40 kV, and the current was 40 mA. The crystallinity index (CI) was calculated using the following empirical equation:

$$CI(\%) = \frac{I_{002} - I_{am}}{I_{002}} \times 100\%$$

Here, I_{002} is the maximum diffraction intensity of crystalline from plane (002) at 2θ = 22.6° and I_{am} is the background scatter intensity measured at 2θ = 18° [23].

2.7. Thermogravimetric analysis

The thermal stability of PP-C, PP-MCC, and C-MCC was conducted in a nitrogen gas atmosphere of 10 ml/min using a thermal-gravimetric analyzer (TG 209C, NETZSCH Inc.). The samples weight was approximately 10 mg, and the temperature was raised from 25 °C to 500 °C at a heating rate of 10 °C/min.

2.8. Water/oil holding capacity analysis

The water holding capacity (WHC) of the PP-MCC and C-MCC was measured by centrifugation using the method described by Fuentes-Alventosa et al. [24] with some modifications. Each 5.000 g of cellulose samples was homogenized in 50 ml of water for 1 h at room temperature. Next, the mixture liquid was centrifuged at 3000*g* using an Avanti J-E centrifuge (BECKMAN COULTER, USA) for 15 min. Supernatants were carefully discarded and the resultant hydrated cellulose was collected and weighed. The oil holding capacity (OHC) was determined under the same condition as the WHC using soybean oil (0.917 g/ml density). The WHC and OHC were expressed as grams of water/oil retained per gram of sample and calculated as:

$$WHC/OHC\ (g/g) = (W_1 - W_0)/W_0$$

Here, W_0 and W_1 are the weights of the dry and swollen samples, respectively.

2.9. Statistical analysis

Results were expressed as mean values and standard deviation (SD). The analysis of variance was performed using the one-way analysis of variance (ANOVA) and differences between the means of samples were analyzed by Duncan's test at a significance level of 0.05.

3. Results and discussion

3.1. Infrared spectra assay of PP-MCC

The infrared spectra of PP-C, PP-MCC, and C-MCC ranged from 4000 cm^{-1} to 400 cm^{-1} are shown in Fig. 1. The absorptions of approximately 3400 cm^{-1}, 2900 cm^{-1}, 1430 cm^{-1}, 1370 cm^{-1}, 890 cm^{-1} exhibited in all spectra were associated with the characteristics of native cellulose [25]. All infrared spectra of the samples displayed a broad and intense peak at around 3400 cm^{-1} per the characteristic absorption of the OH stretching vibration of cellulose [18, 26]. The peaks at 2917 cm^{-1} were attributed to the aliphatic saturated C—H bonds in the cellulose. There was a strong absorption band at 1745 cm^{-1} indicating

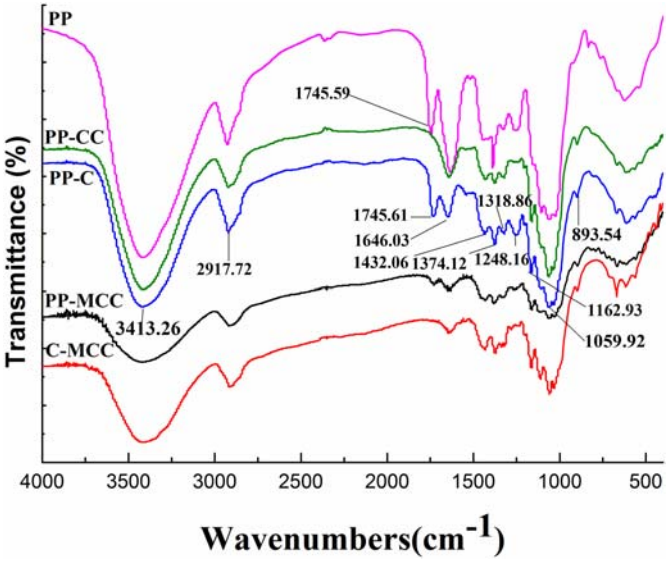

Fig. 1. FT-IR spectrum of PP, PP-CC, PP-C, PP-MCC, and C-MCC.