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Properties of microcrystalline cellulose obtained from coconut (*Cocos nucifera*) husk

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Abstract

This study was aimed at developing a pharmaceutical grade microcrystalline cellulose from coconut fruit husk fibre as a tablet excipient. The microcrystalline cellulose coded CH-MCC, was obtained from coconut *(Cocos nucifera)* fruit husk fibre by a two-stage sodium hydroxide delignification process followed by sodium hypochlorite bleaching and acid hydrolysis. CH-MCC was examined for its physicochemical and powder properties. The powder properties of CH-MCC were compared to those of best commercial microcrystalline cellulose grade, Avicel PH 101. The extraction yield of CH-MCC was approximately 19%. The cellulose material was composed of irregularly shaped fibrous cellulose particle with a moisture content of 6.2%. The true density was 1.53. The flow indices showed that CH-MCC flowed poorly. The hydration and swelling capacities were 3.15 and 48.6% respectively. The study revealed that the cellulose material compares favourably with Avicel PH 101 as well as official requirement specified in the British Pharmacopoeia 1993 for microcrystalline cellulose.

Keywords: Coconut fruit fibre, microcrystalline cellulose, powder properties.

Introduction

Microcrystalline cellulose, (MCC), is described as purified, partially depolymerised cellulose, prepared by treating α-cellulose, obtained as a pulp from fibrous plant with mineral acids. MCC is one of the most used filler-binders in direct compression. Its popularity in direct compression is because of its extremely good binding properties as a dry binder. It also works as a disintegrant and a lubricant and has a high dilution potential in direct compression formulations. In addition to its use in direct compression formulations, MCC is used as a diluent in tablets prepared by wet granulation, as a filler for capsules and for the production of spheres (Bolhuis and Chowhan, 1996). Commercially available MCC is derived from both gymnosperms (generally conifers) and other softwoods, and from hardwood dicotyledons. These woods differ considerably in chemical composition (proportions of cellulose, hemicelluloses and lignin) and structural organization which affect the composition of the α-cellulose extracted and the composition and crystallinity of MCC finally produced (Landin *et al.*, 1993). Besides the wood pulp as a source of cellulose and its derivatives, another commonest source is the purified cotton linters obtained from *Gossypium* species

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(Evans, 1989). Purified cotton linters and wood pulp are obtained from plantations specially grown in temperate climates, as such its production is expensive and the need for exploring other sources for MCC becomes imperative. Alternative sources for MCC recently investigated had examined agricultural wastes and other plant parts not traditionally used for MCC production (Alfa *et al.*, 2000; Audu-Peter *et al.*, 2004 and Ohwoavworhua *et al.*, 2004).

Coconut *(Cocos nucifera)* is widely distributed throughout West Africa and is found near villages on coast often inland. The fibres obtained from the fruit husk finds use as brushes, cordage, carpet-matting, belting, ship cables etc. being resistant to sea water as well as supple and elastic. It is also used for caulking in preference to hemp tow, being light and durable (Dalziel 1937). Coconut fruit husk has been reported in literature as a source of α-cellulose (Okhamafe *et al.*, 1987). The purpose of this investigation is to study the usefulness of microcrystalline cellulose obtained from it as a tablet excipient. This paper reports on the properties, physicochemical and powder, of the microcrystalline cellulose obtained from the fruit husk. The powder properties were compared with the commercial grade microcrystalline cellulose, Avicel PH 101.

Experimental

Materials. Sodium hydroxide (BDH, England), sodium hypochlorite as 'Jik' (Reckitt and Colman Ltd, Nigeria), hydrochloric acid (Fisons, UK), Avicel PH 101 (FMC corporation, USA), xylene, phloroglucinol and iodine crystals (Hopkin and Williams, London) were used as obtained. All other chemicals used were of analytical reagent grade and water was double distilled. Microcrystalline cellulose obtained from coconut fruit husk, coded CH-MCC, was prepared in our laboratory by the following method.

Extraction of α- cellulose. The alpha cellulose was prepared according to the method of Ohwoavworhua *et al.* (2004) with minor modification. Coconut fruit husks were shredded into its fibres and dried in air oven at 60 ^{0}C for 16 h. A 125 g quantity of the material was placed in a stainless steel container to which was added 5 L of 2% w/v sodium hydroxide and digestion was effected for 3 h at 80 ^{0}C in a water bath (FGL 1083 Karl Kolb scientific). This step, also removes lignin in the form of soluble complexes. Following thorough washing and filtration, it was bleached once with 2.4 L of a 1:1 aqueous dilution of sodium hypochlorite for 30 minutes at 100 ^{0}C. The washed and filtered material was then treated with 2.0 L of 17.5 % w/v sodium hydroxide at 80 ^{o}C for 1.25 h. The resulting alpha-cellulose was washed thoroughly. The extraction process was completed by whitening with a 1:1 aqueous dilution of sodium hypochlorite for 5 min. at 80 ^{0}C and was washed until it was neutral. The cellulose material was filtered, pressed and manually reduced to small lumps, which were dried in a fluidized bed dryer (laboratory model, Copley) at an inlet air temperature of 57 – 60 ^{o}C for 1.5 h.

Production of microcrystalline cellulose (MCC). The procedure reported earlier (Ohwoavworhua *et al.*, 2004) with slight modification was used. A 50 g quantity of the alpha cellulose obtained was place in a glass container and hydrolyzed with 1.0 L of 2.5 N hydrochloric acid at boiling temperature for 15 min. The hot acid mixture was poured into cold tap water which was followed by vigorous stirring with a glass rod for 5 min and the mixture was allowed to stand overnight at room temperature. The product was washed with water until neutral, pressed and dried in a fluid bed dryer at an inlet air temperature of 57 – 60 ^{o}C for 60 min. Following further milling and sieving, the fraction passing through 1.0 mm sieve was obtained and stored at room temperature in a

desiccator.

Physicochemical properties of CH-MCC. The organoleptic characteristic, identification, organic impurities, starch and dextrin, solubility, total ash and water-soluble substances were carried out in accordance with BP 1993 specifications. An optical microscope, Nikon model Larphot 2 (Nikon Inc. Japan) was used for preliminary assessment of the nature of particles in CH-MCC. The combination of low and high power objective lenses of 100 and 400 times magnification was used.

pH determination. This was done by shaking 2 g of the powder material with 100 ml of distilled water for 5 min and the pH of the supernatant liquid was determined using a pH meter (Corning, model 10 England) (Ohwoavworhua *et al.*, 2004).

Powder properties

Particle size analysis. An Endicott's sieves shaker, (Endicott's Ltd UK) was used for this. Test sieves ranging from 1.18 mm to 75 μm were arranged in a descending order. A 20 g quantity of CH-MCC powder was placed on the top sieve and was shaken for 5 min. and the weight of material retained on each sieve determined.

True density. The true densities, (D_t), of cellulose powders were determined by the liquid displacement method using xylene as the immerse fluid (Okhamafe *et al*, 1991).

Flow properties

Angle of repose. The static angle of repose, *a,* was measured according to the fixed funnel and free standing cone method (Train, 1958). A funnel was clamped with its tip 2 cm above a graph paper placed on a flat horizontal surface. The powders were carefully poured through the funnel until the apex of the cone thus formed just reached the tip of the funnel. The mean diameters of the base of the powder cones were determined and the tangent of the angle of repose calculated using the equation:

$$\text{Tan } a = 2h/D \quad \text{.....1}$$

Where h is height of heap of powder and D is the diameter of the base of heap of powder.

Bulk and tap densities. A 10 g quantity of powder samples were each, placed into 50 ml clean, dry measuring cylinder and the volume, V_o, occupied by each of the samples without tapping was determined. After 500 taps using Stampfvolumeter (Model STAV 2003 JEF, Germany), occupied volumes, V_{500} were determined. The bulk and tap densities were calculated as the ratio of weight to volumes (V_0 and V_{500} respectively) (Ohwoavworhua *et al.*, 2004).

Hausner and Carr's compressibility indices. These were calculated as reported earlier (Ohwoavworhua *et al.*, 2004).

Powder Porosity. This was derived from the values of true and bulk densities when fitted into the equation:

$$e = 1\text{-}B_b/D_t \times 100 \quad \text{.....2}$$

Where B_b is the bulk density , D_t is the true density and e is the porosity

Hydration capacity. The method of Kornblum and Stoopak (1973) was used. A 1.0 g each of the samples was placed in each of four 15 ml plastic centrifuge tubes and 10 ml distilled water was added from a 10 ml measuring cylinder and then stoppered. The contents were mixed on a vortex mixer (Vortex-Gennie Scientific Industry, USA) for 2 min. The mixture was allowed to stand for 10 min. and immediately centrifuged at 1000 rpm for 10 min. on a Gallenkamp bench centrifuge (Gallenkamp, England). The supernatant was carefully decanted and the sediment weighed. The hydration capacity was taken as the ratio of the weight of the sediment to the dry sample weight

Swellability. This was measured at the same time as the hydration capacity determination using the method of Okhamafe *et al.* (1991).

Moisture sorption capacity. Two grams of the cellulose materials were accurately weighed and evenly distributed over the surface of a

70 mm tarred *Petri* dish. The samples were then placed in a large desiccator containing distilled water in its reservoir (RH = 100%) at room temperature and the weight gained by the exposed samples at the end of a five-day period was recorded. The amount of water sorbed was calculated from the weight difference (Ohwoavworhua *et al.*, 2004).

Moisture content. Five grams of powdered samples were transferred, each, into a *Petri* dish and then dried in an oven at 60 °C until a constant weight was obtained. The % moisture content was then determined as the ratio of weight of moisture to weight of sample expressed as percentage (Ohwoavworhua *et al.*, 2004).

Results and Discussion

The yield of alpha cellulose was about 25 percent w/w of the original material. The yield of the microcrystalline CH-MCC, obtained from alpha-cellulose was approximately 74 percent w/w. Thus the yield of CH-MCC was approximately 19 percent w/w of the starting dry plant material. The results of some of the physicochemical properties investigated are shown in Table 1. The results indicate a high level of purity of the cellulose material. The organoleptic qualities of the CH-MCC produced were good as the material was odourless, tasteless, white and granular in texture.

Table 1: Some physicochemical properties of CH-MCC

Tests	CH-MCC
Organoleptic	Odourless, white, tasteless, coarse powder
Organic impurities	Nil
Identification	Turns violet-blue with iodinated $ZnCl_2$
Starch and dextrins	Nil
pH	6.5
Solubility*	Complete and no residue
Water-soluble substance	< 0.2%
Microscopy	Irregularly shaped fibrous particles which are a mixture of primary particles and spherical aggregates.

*(In ammoniacal solution of copper tetrammine)

Table 2: Powder properties of CH-MCC and Avicel PH 101

Parameters	CH-MCC	Avicel PH 101
True density (g/ml)	1.53 (0.16)	1.40 (0.06)
Bulk density (g/ml)	0.36 (0.0)	0.31 (0.04)
Tapped density (g/ml)	0.5 (0.0)	0.42 (0.12)
Porosity (%)	76.5	78
Flow properties:		
(a) Angle of repose	37.11 (0.36)	41.20 (0.46)
(b) Hausner index	1.39	1.35
(c) Compressibility index (%)	28	26
Hydration capacity	3.15 (0.19)	2.17 (0.01)
Swelling capacity (%)	48.6 (0.15)	21.4 (0.03)
Moisture sorption capacity (%)	15.0 (0.16)	16.6 (0.24)
Moisture content (%)	6.2 (0.32)	7.4 (0.4)

Values are mean and standard deviation is in parenthesis; Number of replicates, N=3

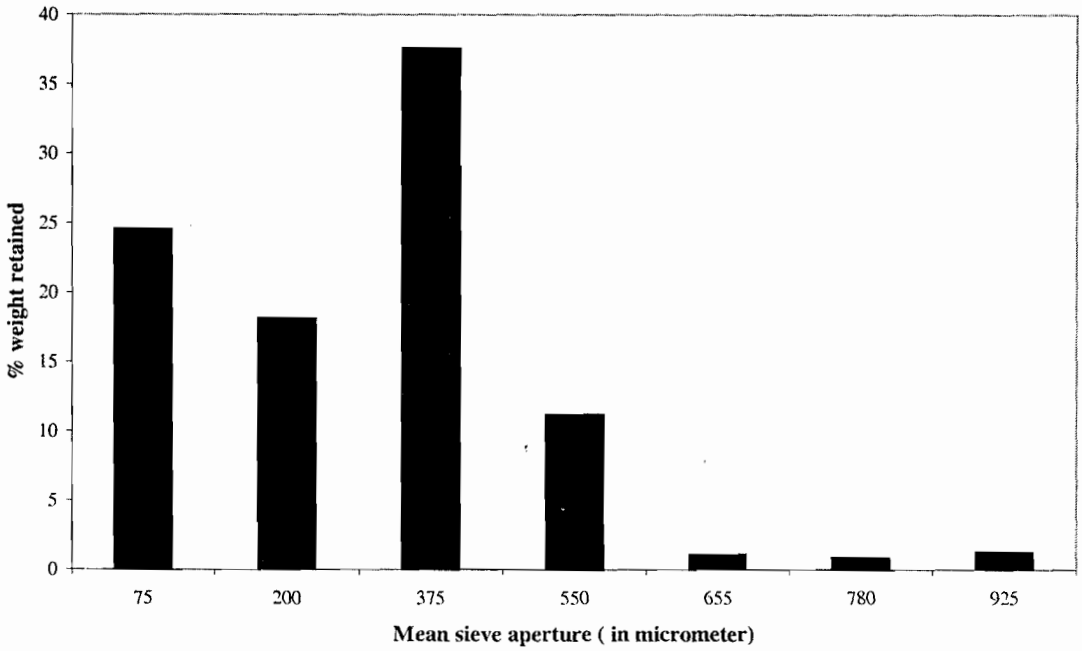

Figure 1: Particle size distribution of CH-MCC powder

The powder properties of CH-MCC and Avicel PH 101 are presented in Table 2 while the result of particle size analysis of CH-MCC powder is as shown in Figure 1. The true density of 1.53 for LC- MCC (Table 2) is high when compared to 1.40 for Avicel PH 101. Stamm (1964) had pointed out that a direct correlation exists between the degree of crystallinity of cellulose and its true density when determined in a non-polar liquid. Consequently, the slightly high true density value for CH-MCC is suggestive that CH-MCC has higher degree of crystallinity than Avicel PH 101. The moisture content of CH-MCC produced was about 6.2% which is well below the official limit of 8 % in British Pharmacopoeia, 1993. This low value is indicative of the suitability of CH-MCC as a diluent in the formulation of hydrolysable drugs such as aspirin.

The flow properties of the powder are essential in determining the suitability of a material as a direct compression excipent. The angle of repose, Hausner index and Carr's percent compressibility are considered as indirect measurements of powder flowability (Staniforth, 1996). The angle of repose of CH-MCC is high (Table 2), which is indicative of very poor flow (Well and Aulton, 1996). While the Hausner index is indicative of interparticle friction, the Carr's index (%) shows the aptitude of a material to diminish in volume (Staniforth, 1996). As the values of these indices increase, the flow of the powder decreases. In general however, Hausner ratio greater than 1.25 indicates poor flow and Carr's compressibility index below 16 % indicate good flowability while values above 35 % indicate cohesiveness (Staniforth, 1996). Thus, the flow indices (Table 2) showed that both CH-MCC and Avicel flowed poorly. As such a glidant will be needed when they are to be used in solid dosage formulation.

Swelling which is generally accepted as an indication of tablet disintegration ability (Caramella, 1991) can be assessed by the determination of hydration capacity, swelling

capacity and moisture sorption profile. The hydration capacity value (Table 2) indicates that CH-MCC is capable of absorbing more than thrice its own weight of water. The swelling capacity, which reflects the increase in volume of cellulose following water absorption, was 48.16 % (Table 2). This value indicates that only a small portion of absorbed water actually penetrated the individual cellulose particles causing them to swell. The bulk of the absorbed water would exist in a 'free' state between the particles. Thus, if the cellulose was incorporated in tablet formulation as a disintegrant it would probably produce tablet disintegration by two mechanisms: capillary or wicking due to interparticulate water and swelling. Additionally, the slightly higher hydration and swelling capacities values observed for CH-MCC could possibly be due to its low moisture content (Table 2).

The moisture sorption capacity values for CH-MCC and Avicel PH 101 are comparable. This property is a measure of moisture sensitivity of material. Stamm (1964) reported that the crystallite portion of cellulose does not adsorb water and that the extent of water adsorption by cellulose should thus be proportional to the amount of amorphous cellulose present. Thus, the result is indicative of the high degree of crystallinity as well as suggestive of comparable amount of amorphous cellulose expected of these materials. Additionally, larger surface area due to small particle size (approximately 50 μm) of Avicel powder could have, in part, accounted for the slightly high amount of moisture taken up by it. Also, study of water sorption is of importance since it reflects the relative physical stability of tablets made from CH-MCC when stored under humid condition. In all, this property showed that the cellulose powders are sensitive to atmospheric moisture and should therefore be stored in air tight container.

Conclusion

The cellulose product, CH-MCC, obtained from coconut fruit husk conformed to the official specifications in the British Pharmacopoeia (1993). The powder properties indicate that CH-MCC and Avicel PH101 are comparable, hence microcrystalline cellulose obtained from coco-nut fruit husk, CH-MCC, is a potential tablet excipient.

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