Effect of Drying Methods on the Powder and Compaction Properties of Microcrystalline Cellulose Derived from *Cocos nucifera*

**ABSTRACT**

**Aim:** This work aims at investigating the effect of drying methods on the powder and compaction properties of microcrystalline cellulose (MCC) obtained from the matured fruit husk of *Cocos nucifera* (CN).

**Study design:** Experimental design

**Place and Duration of study:** University of Nigeria, Nsukka from March 2014 to September, 2016

**Method:** Dried CN husk chips were treated with sodium hydroxide to obtain α-cellulose which on further treatment with dilute hydrochloric acid gave MCC. One portion was lyophilized at -45 ± 2 °C for 3 h (coded MCCL-Cocos) while a second portion was fluidized dried at 60 ± 1 °C for 2 h (coded MCCF-Cocos). The MCCs were characterized using standard methods. Avicel pH 102 was used as comparing standard.

**Results:** Physicochemical properties of the derived MCC such as degrees of polymerization DP and crystallinity, molecular weight, morphology and micromeritic properties were determined. The compaction properties were assessed using Kawakita and Heckel equation models. The MCCs had crystallinity index ranging from 81.25 to 82.12 %, DP of 222. MCCF-Cocos had better flow indices than MCCL-Cocos. The powders exhibited good densification behaviors based on the Kawakita model assessment. Compacts of
MCCL-Cocos were significantly ($P = 0.01$) harder than MCCF-Cocos compacts. Heckel analysis showed plastic behavior. The compaction properties - hardness and tensile strength of the CN MCCs were lower than that of Avicel PH 102.

**Conclusion**: Fluid bed dried MCC had better flowability while lyophilized MCC had superior densification and compaction properties. Thus drying methods had an effect on the powder and compaction behavior of *C. nucifera* MCC.

**Keywords**: [Cocos nucifera, microcrystalline cellulose, powder, compaction]

1. **INTRODUCTION**

The cells of all green plants are known to contain cellulose and lignin abundantly in their cell walls and this is responsible for the structure and mechanical strength exhibited by such plants [1]. Cellulose has over the years been found to be very useful in many activities of mankind because of its possession of such attributes as strong tensile strength, low density and eco-friendliness. Microcrystalline cellulose for industrial purposes is mostly derived from purified cotton linter and wood pulp. The desire for cheaper eco-friendly processes as well as the need to slow down the fast global deforestation has stimulated renewed interest in agro-fiber plants waste [2]. As a result of this, the husk from the fruit of the coconut plant (*Cocos nucifera*), which occurs as a huge agricultural waste globally and especially along the coastline areas in Nigeria, was investigated as a source for the production of MCC.

*Cocos nucifera* is a large palm plant of the family Arecaceae that has pinnate leaves and bears numerous fruits in bunches at a time. Each fruit contains a nut (shell) with a large volume of water or milk inside and three indentations on the head. The fruit consists of three layers: the exocarp, mesocarp and endocarp. The husk is made up of the exocarp and mesocarp while the mesocarp consists mostly of a fibrous material. The coconut plant is cultivated in many tropical and sub-tropical regions where the plant thrives as a result of climatic reasons – abundant sunshine, regular rainfall and high humidity [3]. It thrives well on sandy soils and is highly tolerant of salinity. Different brands of microcrystalline cellulose are
commercially available from different manufacturers in a quest to meet the various requirements of the
different consumers. Many of these brands exist as a result of the various physical powder characteristics
which each brand possesses. Such differences are imparted by variation of not only the pulp source but
also the processing steps during manufacture [4]. One of the processing steps is drying. Commercially,
most MCC is dried by the spray drying method which imparts a high inter particulate porous and
aggregate nature to the dried powder [5,6]. Fluid bed drying method aids powders to dry as discrete
particles with a low inter particulate void while lyophilization causes the iced portion of a frozen material to
be sublimed directly into vapour at a low partial pressure of water [7]. On drying, this leaves a material
with a high porous inter particle void [8]. All these different drying methods are expected to affect the
particle morphology, size, porosity and aggregation which in turn would affect the characteristic of the
material. Thus in this study, an investigation of the effect of drying procedure by lyophilization and
fluidization on the powder and compact characteristics of the MCC derived from the fruit husk of CN was
carried out.

2. MATERIAL AND METHODS

2.1 Materials

Sodium hydroxide (Merck, Germany), Hydrochloric acid, Magnesium nitrate (BDH, Poole England),
sodium hypochlorite 3.5 % w/v (JIK®, Reckitt and Colman Nig. Plc), Avicel® PH 102 (FMC Biopolymer,
USA), Potassium sulphate, Sodium chloride (J.T. Baker, New Jersey, USA), Talc, Magnesium stearate
(Sigma, USA) and distilled water (Pharm. Tech. Lab, Uniport). Dried chips of coconut fruit husk.

2.1.1 Method

2.1.2 Isolation of Alpha Cellulose

Matured fruits of the coconut plant, Cocos nucifera, were harvested from different trees within Mgbeoba
town, Port Harcourt, Rivers State, Nigeria. The fruits were de-husked and husk fibers/strands cut into
small pieces of about 3 – 5 mm length and were air dried until completely dry and stored appropriately.
The method of Ohwoavworhua et al [9] with modification was adopted. An 800 g quantity of the dried
brittle fibers/strands of the coconut husk were placed in a 15 L stainless steel container to which 10 L of
3.5 % w/v solution of sodium hydroxide was added and digestion was effected for 4 h at a boiling
temperature of 100 °C on a sand bath to partially de-lignify the husk. The de-lignified mass was
thoroughly washed with water until neutral to litmus. Excess water was squeezed out through a muslin
cloth. First bleaching was done using 0.4 % w/v of sodium hypochlorite solution heated to 100 °C for 30
min. After washing thoroughly with water until neutral, the resultant pulp was further treated with 17.5 %
w/v solution of sodium hydroxide at 100 °C for 1 h. The resultant pulp was bleached two more times with
0.4 % w/v of sodium hypochlorite and followed by washing with distilled water until neutral to litmus.

2.1.3 Acid hydrolysis of alpha cellulose obtained from Cocos nucifera husk

A 60 g quantity of the alpha cellulose was put in a 2 L glass beaker (Pyrex®, England) and treated with
0.8 L of 2.5 N HCl at 105 ± 2°C in an oil bath. The hot acid mixture was poured into cold water and stirred
vigorously until cold, and allowed to settle. The supernatant was decanted and the material obtained was
washed until neutral to litmus and excess water squeezed out with a muslin cloth. The lumps of MCC
obtained were divided into two portions. One portion was fluid bed dried using a Tornado model
(Sherwood®, China) fluid bed dryer set at an inlet air temperature of 60 ± 1 °C for 2 h. The second portion
was lyophilized at - 45 °C for 3 h using an LGT 18 freeze dryer (Gallenkamp®, England). Each of the
material was milled using a domestic blender (Binatone®, Japan) and screened through a 250 µm
stainless sieve (Retsch®, Germany). The microcrystalline cellulose obtained by the different drying
processes was stored separately.

2.2 Physicochemical evaluation

2.2.1 Identification

The color, odor, taste and texture of the MCC were observed and recorded. A 2 g portion of the MCC
was soaked in iodine solution for 5 min and drained of excess reagent. Observations were made for
change in colour. Also, 2 drops of 60 % v/v of sulphuric acid solution were added to a fresh portion of the
MCC and observations were also recorded [10].

2.2.2 pH determination
This was done by shaking for 5 min a dispersion of 2 g of MCCF-Cocos, MCCL-Cocos and Avicel PH 102 in 100 mL of distilled water. This was allowed to settle and the pH of the supernatants determined using a pH meter (PHS® 25, India). Three replicate determinations were made.

### 2.2.3 Solubility

A few drops of excess of distilled water, acetone, 0.1 N hydrochloric acid (HCl) and ethanol were added to 1.0 g of MCCF-Cocos, MCCL-Cocos and Avicel PH 102. Observations were recorded.

### 2.2.4 Elemental / Heavy metal analysis

Analysis for the presence of heavy metals such as lead (Pb), iron (Fe), zinc (Zn) manganese (Mn) and Arsenium (As) was done using Atomic Absorption Spectroscopy, AAS (Model AA-7000, ROM version 1.01, S/N A30664700709 SHIMADZU, Japan).

### 2.2.5 Total ash content

The total ash content was determined by the measurement of the residue left after the independent combustion of 3 g of MCCL-Cocos in a furnace at 550°C for 5 h.

### 2.2.6 Scanning Electron Microscopy (SEM) test

The morphology and particle shape were investigated using a scanning electron microscope (Phenom Prox, Model no MVE016477830, Netherlands)

### 2.2.7 X-ray Diffraction (XRD)

An X-ray diffractometer (D/MAX-1200, Rigaku, Japan) powered by a 45 kv X-ray generator (RINT 2000, Rigaku, Japan) at an input of 35mA and set at 0.020° and step time of 29.10 sec (Cu Kα radiation) and a scan speed of 2’ per sec at a 2 theta (Θ) range of 3 - 80° was used. The degree of crystallinity of each sample was calculated from Equation 1

\[
\text{Crystallinity index (C.I)} = \left[\left(\frac{I_{002} - I_{am}}{I_{am}}\right)\right] \times 100
\]

\[\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldOTS
Where $I_{002}$ is the highest peak intensity of the crystalline fraction and $I_{am}$ is the low intensity peak of the amorphous region.

### 2.2.8 Degree of polymerization and Molecular weight

A U tube viscometer (Technicu size C100, 1983) and an ammoniacal solution of copper was used. Stock solutions of 2 % w/v of MCCL-Cocos and Avicel PH 102 were separately made. Serial dilutions of 1 %, 0.5 %, 0.25 %, 0.125 % were made from each MCC stock solution. Flow time measurements were made for each MCC solution at the different dilutions available and the flow rate measurements determined. The dilutions were considered adequate when the viscometric time measurements for the MCC solutions were identical to that of the ammoniacal solution of copper. The experiments were conducted at ambient temperature (29 ±1°C). The densities of the different MCC solutions were determined using pycnometric method. From the relative viscosity, the reduced viscosity was measured. The intercept of y-axis of the plot of the reduced viscosity versus concentration is the intrinsic viscosity. From the value of the intrinsic viscosity, the degree of polymerization was calculated from Equation 2 [12].

$$(DP)^{0.85} = 1.1 \times \eta \quad \text{................................................................. 2}$$

where $\eta$ is the intrinsic viscosity.

The molecular weight can be calculated from Equation 3

$$DP = \frac{M}{M_o} \quad \text{................................................................. 3}$$

Where $M$ is the molecular weight of the material or polymer, and $M_o$ is the molecular weight of glucose.

Calculations and plots to determine the intrinsic viscosity, relative viscosity, the molecular weight and degree of polymerization were done for each MCC sample.

### 2.2.9 Swelling capacity determination

The swelling capacity of the microcrystalline cellulose samples MCCF-Cocos, MCCL-Cocos and AV-102 were determined by using the method of Bowen and Vadino [13] with slight modification. A 3 g quantity
of MCCF-Cocos, MCCL-Cocos and AV-102 was individually placed in a graduated glass measuring cylinder and tapped to obtain the tapped volume, \( V_t \). A dispersion of each powdered sample was made in 85 mL of water with thorough shaking before making up the volume to 100 mL with more water. The mixture was allowed to stand undisturbed for 24 h on a flat surface and the volume of the sediment formed, \( V_v \) noted. Triplicate determinations were done and the swelling capacity calculated as a percentage using Equation 4 [14].

\[
\text{Swelling capacity (S.C.)} = \left( \frac{V_v - V_t}{V_t} \right) \times 100
\]

2.2.10 Hydration capacity

The hydration capacity was determined using the Kornblum and Stoopaks method [15] with slight modification. A 1 g quantity of each of the sample was put in a 15 mL plastic centrifuge tube and 10 mL of distilled water added to each. Each tube was shaken intermittently over a 2 h period and left to stand for 30 min. Centrifugation at 1000 revolutions per minute (rpm) was done for 10 min. The supernatant was carefully decanted and the wet sediment weighed. The hydration capacity was calculated as:

\[
\text{Hydration capacity (H.C.)} = \frac{x}{y}
\]

Where \( x \) is the weight of the wet sample/powder sediment and \( y \) is the weight of the dry sample/powder.

2.2.11 Moisture content

A 5 g quantity each of MCCF-Cocos, MCCL-Cocos and AV-102 were placed individually in tarred white porcelain crucibles and dried in a hot air oven (Mermmet, England) at 105°C until constant weights were obtained. The moisture content was determined as the percentage of the ratio of the weight of the sample after drying to the weight of the sample before drying [16]:

\[
\text{Moisture content (M.C.)} = \left( \frac{W_f}{W_i} \right) \times 100
\]

Where \( W_f \) is the final weight of powder after drying, and \( W_i \) is the initial weight of powder before drying.
2.2.12 Moisture sorption test

A 2 g quantity each of the MCCF-Cocos, MCCL-Cocos and AV-102 were placed on the surface of three different 7 mm tarred Petri dishes and kept in desiccators of relative humidities of 96, 84, 75 and 52 % respectively at room temperature of 29 ± 1°C. The weight gained over a 5 day period was calculated for each sample from Equation 7. Determinations were done in triplicates.

\[
\text{Moisture sorbed (M.S.)} = \left(\frac{W_2 - W_1}{W_1}\right) \times 100
\]

Where \( W_1 \) is the weight before exposure and \( W_2 \) is the weight after exposure

2.2.13 Particle size analysis

The method of nest of sieves was used. Stainless steel sieves (Retsch®, Germany) ranging from 1000 µm to 45 µm arranged in descending order with a collection pan underneath the sieves was placed on a sieves shaker (Retsch® Ltd, Germany). The sieves were weighed empty. A 15 g sample of each of MCCF-Cocos, MCCL-Cocos and AV-102 were placed on the topmost sieve and was agitated for 5 min at an amplitude of 1.5 mm/g. The weight of material retained on each of the sieves was obtained by deducting the weight of the empty sieves from its weight with the powder sample retained. The percentage of samples retained were calculated. Triplicate determinations were done and the mean weight at each determination calculated as [17]:

\[
\text{Average diameter} = \left(\frac{\Sigma (\% \text{ retained} \times \text{mean aperture})}{100}\right)
\]

2.2.14 Bulk and Tapped density

A 10 g quantity each of MCCF-Cocos, MCCL-Cocos and AV-102 was poured freely under gravity into a 50 mL clean, dry, graduated measuring cylinder and the volume occupied by the sample noted as the bulk volume (Vb). The bulk density, \( Db \) was calculated from Equation 9 [17]:

\[
Db = \frac{M}{Vb}
\]

Where \( M \) is the mass of the MCC powder.
The cylinder was tapped on a flat wooden platform by dropping the cylinder from a height of about 2 – 3 cm at 2 – 3 sec intervals until there was no further reduction in the volume of the MCC powder. The tapped volume \( V_t \) was noted and the tapped density, \( D_t \) was calculated from Equation 10:

\[
D_t = \frac{M}{V_t}
\]  

**2.2.15 Particle density**

The particle density of each of the MCC samples was determined by the liquid displacement method using xylene as the immersion fluid. A pycnometer of 25 mL volume was weighed empty, filled with xylene, stoppered and excess fluid wiped off the body of the pycnometer. This was weighed and denoted as, a. A 1 g quantity of each of the powdered samples was weighed, and noted as \( W_p \). It was placed in the stoppered pycnometer, wiped clean of excess fluid and reweighed. The particle density was calculated from Equation 11 [18].

\[
P_d = \frac{W_p}{(a + W_p) - b} \times \text{S.G.}
\]

Where \( P_d \) is the particle density, S.G. is specific gravity of the xylene, a is the weight of pycnometer and xylene, \( W_p \) is the weight of powder and b is the weight of pycnometer + xylene + powder. Triplicate determinations were made for each powder sample.

**2.2.16 Flow rate and angle of repose**

A 10 g quantity of MCC powder was poured into a clamped stoppered clean glass funnel whose orifice was 5 cm above a flat surface. The powder was allowed to flow freely from the funnel unto the platform. The time of flow, the diameter and height of the powder heap formed were measured. The flow rate and tangent of the powder heap were calculated as:

\[
F.R. = \frac{M}{F.T.}
\]

Where F.R. is flow rate, F.T. is flow time and \( M \) is mass of powder used.

\[
\text{Angle of repose } (\Theta) = \tan^{-1}\left(\frac{h}{r}\right)
\]
2.2.17 Hausner’s quotient (ratio) and Carr’s index

The Hausner’s quotient and Carr’s index for each powder were calculated from Equations 14 and 15 [12].

Hausner’s quotient (H.Q.) = $\frac{D_t}{D_b}$ .............................. 14

Carr’s Index (C.I.) = $(1 - \frac{D_b}{D_t}) \times 100$ ............................ 15

2.2.18 Powder porosity

Powder porosity, $\varepsilon$ is given as: [13].

$\varepsilon = (1 - \frac{D_b}{P_d}) \times 100$ ................................. 16

where $P_d$ is the particle density, and $D_b$ is the bulk density

2.2.19 Compactibility and powder cohesion

The Kawakita Equation describes the relationship between the volume reduction of a powder column and the applied pressure on tapping. A 10 g quantity of the MCC was poured freely into a 50 mL graduated, glass measuring cylinder. The bulk volume, $V_0$ was noted and the cylinder was mechanically tapped at pre-determined incremental number of taps and the volumes occupied by the powder noted each time until there was no further decrease in volume, $V$. The degree of volume reduction, $C$ was calculated from the values of $V$ and $V_0$. The degree of cohesion and compactibility of the powder was calculated using the Kawakita Equation 17 [20].

$\frac{N}{C} = \frac{N}{a} + \frac{1}{ab}$ .......................................................... 17

$C$ is derived from $(V_0 - V)/V_0$. The cohesiveness of the powder sample is described by $1/b$ while ‘$a$’ is considered as the compactibility of the powder. When $N/C$ is plotted against $N$ in a graph, $1/a$ is the slope while $1/ab$ is the intercept. The procedure was carried out on MCCF-Cocos, MCCL-Cocos and AV-102.
2.3. Compaction of microcrystalline powders

The MCCF-Cocos, MCCL-Cocos and AV-102 were compressed at a target compact weight of 300 mg at different compression loads ranging from 4.90 to 14.71 MPa (Megapascal) using a 10 mm diameter flat faced set of punches for 30 sec. The powder was manually fed into a single punch hydraulic tablet press (Model C, Carver Inc., Winscosin, USA). The punches and dies were lubricated with stearic acid powder before each compaction cycle.

2.3.1 Evaluation of microcrystalline cellulose compacts

The batches of the compacts were each allowed a 24 h post compression relaxation time before evaluation for weight uniformity, thickness, hardness, friability, disintegration time and tensile strengths.

2.3.2 Weight uniformity test

Twenty compacts randomly selected from each batch of MCCF-Cocos, MCCL-Cocos and AV-102 were individually weighed. The mean, standard deviation and coefficient of variation were determined. Acceptance or rejection was based on British Pharmacopoeia tolerance limits for uncoated tablets of > 250 mg which is ± 5 % [6].

2.3.3 Thickness/height and diameter test

With the aid of a micrometer screw gauge ten compacts randomly selected from each of the batches were measured for thickness and diameter. The mean and standard deviation was calculated for each batch.

2.3.4 Hardness test

The hardness of ten compacts randomly selected from each batch of MCC was determined using an Erweka® TBH 200 hardness tester (Erweka®, Germany). The mean, standard deviation, and coefficient of variations were determined.

2.3.5 Disintegration time test
Six tablets randomly selected from each batch were used for the test. The test was carried out using an Erweka® ZT-3 double basket disintegration tester (Erweka®, Germany) and each compact held in place with a glass disc inside each cylindrical hole. Each beaker was filled with 500 mL of 0.1 N HCl heated to a temperature of 37 ± 1°C. The time taken for each compact to completely break up and pass through the mesh was noted. Triplicate determinations were made.

2.3.6 Friability test.

Ten tablets randomly selected from each batch of the compacts were de-dusted and collectively weighed into one of the drums of an Erweka model TAR 200 (Erweka®, Germany) twin drum electronic friabilator programmed to revolve at 25 revolutions per minute (rpm) for 4 min. At the end of the exercise the tablets were collected and de-dusted and any broken tablets were rejected. The tablets were reweighed and the abrasion resistance (B) calculated from Equation 18 [21].

\[ B = 100 \left( 1 - \frac{W}{W_0} \right) \text{ or } 100 \left( \frac{W_0 - W}{W_0} \right) \]  

Where \( W_0 \) is the initial weight and \( W \) is the final weight.

2.3.7 Tensile strength

The tensile strengths of the compacts were determined based on the hardness, thickness and tablet diameter using Equation 19 [22].

\[ \text{Tensile strength (TS)} = \frac{2P}{\pi dt} \]

Where \( P \) is the breaking force, \( d \) is the tablet diameter, \( t \) is the tablet thickness.

2.3.8 Heckel analysis

The Heckel Equation analyzes the relative density of a powder bed to the applied compression pressure during tableting. It describes the densification behavior of the powder bed from the point the die is filled to the time a quantified pressure is applied. It also describes the deformation mechanism of the powder in forming the compact. The Equation is stated as [23].
ln (1 / 1-D) = KP + A .................................................

where D is the relative density of a powder compact at pressure P, K is a constant to measure the plasticity of the compressed material, A is the Y axis intercept, related to die filling and particle rearrangement before deformation and bonding of the separate particles.

2.3.9 Reworking potential of microcrystalline cellulose

Compacts of the microcrystalline cellulose compressed at the different compression loads were randomly selected, crushed to fine particles, screened through a 250 μm stainless sieve and recompressed at the same compression loads and target compact weights of 300 mg using 10 mm flat faced punches. Reworked compacts were evaluated 24 h post compression for uniformity of weight, thickness, hardness, and friability. Plots of hardness against compression pressure were made for the initial compacts and the reworked compacts. The area under the curve (AUC) of each plot was determined using Wolfram/Alpha Widget software. The re-workability was calculated as the percentage of AUC of reworked MCC (AUCr) against AUC of initial MCC (AUCi) compact.

Reworking Potential (R.P.) = (AUCr/AUCi) x 100 ........................................... 20

2.4. Statistical evaluation

The data obtained were statistically analyzed using ANOVA and students t test (SPSS version 21). Values were considered significant at P = 0.00 or below or equal to 0.05.

3. RESULTS AND DISCUSSION

3.1 Physicochemical properties

The yield of α-cellulose and microcrystalline cellulose obtained from CN were 29.22 and 13.88 % respectively with reference to the dry starting material. Results of organoleptic and some physicochemical evaluation tests show the MCCs to be fine, odorless, tasteless and brownish in color. Except for the color, all the other characteristics were similar to Avicel PH 102.
The absence of a dark blue color on the treatment with iodine showed the absence of starch while the presence of a blue color when treated with sulphuric acid confirmed that the sample is MCC (Table 1). The insolvibility of the MCCs in water and some organic solvents is a feature common with MCC and is attributable to their high crystallinity and structure. The samples were near neutral as shown by the pH values obtained (6.58 ± 0.19 to 6.49 ± 0.21). This quality makes them suitable for the formulation of both acidic and basic drugs. The low ash content (1.94) shows that the material was well processed and contains minimal organic matter. The moisture content ranged from 6.58 to 8.88 and falls within compendial set limits [10]. The DP of 222 obtained is typical of MCC and conforms to values of < 350 that is characteristic of MCC [10] (Table 1). The molecular weight of MCC derived from CN (39,994.61) is dependent on the number of polymer chains that are attached to it after polymerization and the value obtained is characteristic of processed cellulose such as Avicel PH 102 [24, 25, 26]. Diffractograms of the samples (Figures 1 & 2) showed diffraction patterns that are similar to Avicel PH 102 (Fig.3). Peaks were observed at 22.74° and 17.00° of 2 theta (θ) for the crystalline and amorphous regions respectively for MCCF-Cocos and at 22.86° and 17.50° of 2θ for MCCL-Cocos. These gave percentage crystallinity values of 82.25 and 80.15 % for MCCF-Cocos and MCCL-Cocos respectively. These values were also comparable with that obtained for Avicel PH 102 which was 80.15 % at 23.32° and 16.24° of 2θ. Crystallinity values of 60 – 80 % have been reported for MCC [27, 28]. The micrographs from SEM test (Fig.4) for MCCL-Cocos showed strands whose morphology, texture and size resembled AV-102 (Fig.5). Elemental analysis results show that the samples were safe as the value of heavy metals were within compendial and World Health Organization (WHO) safe limits [10, 23].

Fig.1: X-ray diffractogram of MCCF-Cocos. Fig.2: X-ray diffractogram of MCCL-Cocos.
Operations: Smooth 0.150 | Background 1.000, 1.000 | Import

File: SAIFXR151014A-07(AV-102).raw - Step: 0.020 °  - Step time: 29.1 s - WL1: 1.5406 - kA2 Ratio: 0.5 - Generator kV: 40 kV - Generator mA: 35 mA - Type: 2Th/Th locked

L in (Counts)

Fig.3: X-ray diffractogram of AV-102

Fig.4: SEM Micrograph of MCCL-Cocos.

Fig.5: Scanning Electron Micrograph of AV-102.

The moisture hysteresis results show an increase in the amount of moisture adsorbed as the relative humidity increased. MCCL-Cocos significantly (P = 0.00) adsorbed more water than MCCF-Cocos at the different relative humidities tested except at 96%. Avicel PH 102 adsorbed more water than the MCCs from CN at the different relative humidities tested.

Hydration capacity has been described as the amount of water a material is able to absorb on hydration while swellability indicates increase in volume of water taken up after absorption [30]. The hydration capacity of the MCCF-Cocos was $2.33 \pm 0.11$ (Table 1) and is lower than that obtained with the lyophilized powder ($2.84 \pm 0.09$). Both values were lower than that of AV-102 ($3.55 \pm 0.07$). Swelling capacity values of the MCCF-Cocos was $33.63 \pm 2.31$ % and this represents about one-third of that for
AV-102 (111.57 ± 3.53). These moisture values impart positively in the disintegration characteristic of MCC in tablet formulations as water uptake could occur by two mechanisms: wicking and swelling.

3.2 Micromeritic properties

Some of the powders properties are shown in Table 3. The bulk and tapped densities, flow rate, and packing fraction of MCCF-Cocos was higher than those of the lyophilized powder, MCCL-Cocos. Also angle of repose (26.63 ± 2.86˚), Hausner’s quotient (1.16 ± 0.01) possessed by the fluidized dried powder was lower than angle of repose (35.91 ± 1.29), Hausner’s quotient (1.28 ± 0.01) and Carr’s index (27.36 ± 5.25 %) for MCCL-Cocos. This implies that the MCCF-Cocos has a greater tendency to flow than MCCL-Cocos. However, the porosity and particle density values obtained for the lyophilized powder were higher than the fluid bed dried powder. This would lead to better densification. Porosity of MCCL-Cocos was significantly (P = 0.01) higher than MCCF-Cocos but comparable with AV-102 implying higher voids/air spaces in the powder bed for MCCL-Cocos. The packing fraction of MCCF-Cocos (0.87 ± 0.01) was higher than that of MCCL-Cocos (0.79 ± 0.01) and shows a more densely packed and consolidated powder bed (Table 2) which can translate to better flowability for MCCF-Cocos than MCCL-Cocos.

The Kawakita plot of the powders is shown in Figure 6. Compactibility and cohesiveness values derived from the graph (Table 3) show higher values for MCCL-Cocos than MCCF-Cocos. The compactibility values obtained suggests that the lyophilized powder bed would undergo greater densification on application of stress or agitation by tapping. Cohesiveness describes the ability of the powders to adhere to each other leading to aggregation and when values are high, flow would be impaired. The lyophilized MCC had a higher value than the fluid bed dried MCC and thus would have a poorer flow. Comparatively, MCCL-Cocos has a significantly (P = 0.02) higher compactibility than MCCF-Cocos but lesser than AV-102.
Table 1: Some physicochemical properties of MCCF-Cocos, MCCL-Cocos and AV-102.

<table>
<thead>
<tr>
<th>Sample/Parameter</th>
<th>MCCF-Cocos</th>
<th>MCCL-Cocos</th>
<th>AV-102</th>
</tr>
</thead>
<tbody>
<tr>
<td>Iodine solution</td>
<td>Reddish brown</td>
<td>Reddish brown</td>
<td>Reddish brown</td>
</tr>
<tr>
<td>Sulphuric acid(60% v/v)</td>
<td>Blue color</td>
<td>Blue color</td>
<td>Blue color</td>
</tr>
<tr>
<td>Solubility in water, alcohol, acetone, dilute mineral acid.</td>
<td>Insoluble</td>
<td>Insoluble</td>
<td>Insoluble</td>
</tr>
<tr>
<td>Solubility in ammoniacal solution of copper tetramine.</td>
<td>Completely soluble</td>
<td>Completely soluble</td>
<td>Completely soluble</td>
</tr>
<tr>
<td>pH ± SD</td>
<td>6.58 ± 0.19</td>
<td>6.49 ± 0.21</td>
<td>6.74 ± 0.31</td>
</tr>
<tr>
<td>Ash content</td>
<td>1.94</td>
<td>1.94</td>
<td>1.95</td>
</tr>
<tr>
<td>% crystallinity</td>
<td>82.14</td>
<td>81.25</td>
<td>80.15</td>
</tr>
<tr>
<td>Molecular weight</td>
<td>39,994.61</td>
<td>39,994.61</td>
<td>42,156.48</td>
</tr>
<tr>
<td>Degree of polymerization</td>
<td>222</td>
<td>222</td>
<td>234.40</td>
</tr>
<tr>
<td>Hydration capacity</td>
<td>2.33 ± 0.11</td>
<td>2.84 ± 0.09</td>
<td>3.55 ± 0.07</td>
</tr>
<tr>
<td>Moisture/RH 96 %</td>
<td>4.80 ± 0.01</td>
<td>4.80 ± 0.22</td>
<td>7.80 ± 0.05</td>
</tr>
<tr>
<td>Moisture/RH 84 %</td>
<td>1.10 ± 0.10</td>
<td>1.40 ± 0.15</td>
<td>1.60 ± 0.20</td>
</tr>
<tr>
<td>Moisture/RH 75 %</td>
<td>0.50 ± 0.02</td>
<td>0.70 ± 0.10</td>
<td>1.10 ± 0.01</td>
</tr>
<tr>
<td>Moisture/RH 52 %</td>
<td>0.20 ± 0.05</td>
<td>0.40 ± 0.01</td>
<td>0.60 ± 0.03</td>
</tr>
<tr>
<td>Pb</td>
<td>0.00</td>
<td>0.00</td>
<td>0.00</td>
</tr>
<tr>
<td>Mn</td>
<td>0.00</td>
<td>0.00</td>
<td>≤ 10</td>
</tr>
<tr>
<td>Zn</td>
<td>0.12</td>
<td>0.12</td>
<td>≤ 10</td>
</tr>
<tr>
<td>Fe</td>
<td>0.00</td>
<td>0.00</td>
<td>≤ 10</td>
</tr>
<tr>
<td>Na</td>
<td>1.09</td>
<td>1.09</td>
<td>≤ 10</td>
</tr>
<tr>
<td>As</td>
<td>0.01</td>
<td>0.01</td>
<td>≤ 10</td>
</tr>
<tr>
<td>Swelling capacity (%)</td>
<td>33.63 ± 2.31</td>
<td>23.38 ± 4.13</td>
<td>111.57 ± 3.53</td>
</tr>
<tr>
<td>Loss on drying (%)</td>
<td>8.88</td>
<td>7.96</td>
<td>6.58</td>
</tr>
</tbody>
</table>

Table 2: Some micromeric properties of MCCF-Cocos, MCCL-Cocos and AV-102.

<table>
<thead>
<tr>
<th>Sample / Parameter</th>
<th>MCCF-Cocos</th>
<th>MCCL-Cocos</th>
<th>AV-102</th>
</tr>
</thead>
<tbody>
<tr>
<td>Parameter</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Bulk density (g/mL)</td>
<td>0.40 ± 0.04</td>
<td>0.32 ± 0.01</td>
<td>0.31 ± 0.04</td>
</tr>
<tr>
<td>Tap density (g/mL)</td>
<td>0.47 ± 0.03</td>
<td>0.41 ± 0.03</td>
<td>0.38 ± 0.02</td>
</tr>
<tr>
<td>Sample</td>
<td>Angle of repose (°)</td>
<td>Flow rate (g/s)</td>
<td>Carr’s index (%)</td>
</tr>
<tr>
<td>--------</td>
<td>---------------------</td>
<td>-----------------</td>
<td>------------------</td>
</tr>
<tr>
<td></td>
<td>26.63 ± 2.86</td>
<td>5.46 ± 0.44</td>
<td>12.77 ± 1.36</td>
</tr>
<tr>
<td></td>
<td>35.91 ± 1.29</td>
<td>Poor flow (5.23 ± 0.22)</td>
<td>5.25 ± 3.85</td>
</tr>
<tr>
<td></td>
<td>30.52 ± 2.35</td>
<td>Poor flow (5.23 ± 0.22)</td>
<td>18.96 ± 0.67</td>
</tr>
</tbody>
</table>

**Table 3: Compactibility and cohesiveness of MCCF-Cocos, MCCL-Cocos and AV-102**

<table>
<thead>
<tr>
<th>Sample</th>
<th>1/a</th>
<th>1/b(Pk)</th>
<th>1/ab (%)</th>
<th>R²</th>
</tr>
</thead>
<tbody>
<tr>
<td>MCCF-Cocos</td>
<td>0.22</td>
<td>4.60</td>
<td>9.09</td>
<td>22.40</td>
</tr>
<tr>
<td>MCCL-Cocos</td>
<td>0.27</td>
<td>4.56</td>
<td>7.14</td>
<td>19.37</td>
</tr>
<tr>
<td>AV-102</td>
<td>0.22</td>
<td>3.39</td>
<td>9.09</td>
<td>12.42</td>
</tr>
</tbody>
</table>

Fig.6: Kawakita plots of MCCF-Cocos, MCCL- and AV-102
3.3 Compact properties

The compacts formed weighed between 290.10 ± 2.75 to 301.60 mg ± 2.48 %. These values fall within the BP set limits of ± 5 % for tablets weighing above 250 mg. Increased compression load resulted in a decreased thickness of the compacts. The friability values were all below 1% and decreased as the compression pressure increased. MCCL-Cocos had values that were consistently lower than their corresponding MCCF-Cocos compacts at the same compression pressures. There was also a general increase in hardness as the compression load increased. The lyophilized compacts were significantly (P = 0.01) harder than the fluidized dried compacts. All values were above 40 N (Fig.8) and could be considered strong enough to withstand rigors of handling and transportation. Tensile strength values (Fig.9) were in the range of 1.73 – 4.73 MN/m² for the MCCF-Cocos compacts and 1.98 – 5.30 MN/m² for the MCCL-Cocos. The lyophilized compacts were consistently and significantly (P = 0.01) stronger at all compression loads. Values for AV-102 ranged from 0.73 – 7.31 MN/m². The disintegration time of all the compacts was within 15 min except for MCCL-Cocos compacts compressed at 12.26 and 14.71 MPa. The MCCL-Cocos compacts had disintegration time values that were significantly (P = 0.02) higher than the MCCF-Cocos compacts (Fig. 7). The hardness, disintegration time and friability were within both BP
2012 and USP 2009 specifications and thus were adjudged good [10, 31]. The Heckel parameters (Table 4) were derived from the straight line portion of the Heckel plot (Fig. 10). The yield pressure, Py describes the tendency of the material to plasticize or fragment under an applied pressure which can be related to its compressibility. MCCF-Cocos and MCCL-Cocos values were 0.14 and 0.18 respectively while AV-102 was 0.22. This implies that MCCF-Cocos exhibited a faster plasticity or onset of deformation than MCCL-Cocos and AV-102. The lyophilized powder compacts had a plastic behavior equivalent to the commercial sample, AV-102. The $D_o$ (initial packing in the die as a result of filling by powder) value of MCCF-Cocos was higher (0.26), than MCCL-Cocos and AV-102. This implies that MCCF-Cocos exhibited a higher degree of packing and re-arrangement on die filling than both the lyophilized and AV-102. In terms of degree of particle packing at zero and low pressure ($D_A$), particle re-arrangement and fragmentation ($D_B$), in the early phase of compression, AV-102 had the highest value followed by MCCF-Cocos and MCCL-Cocos. Generally, all the MCC powders had good compactibility and plasticity, MCCL-Cocos was more compactible than MCCF-Cocos. The reworking potentials of the powders are shown in Fig. 11. All were re-workable implying that after compaction or exposure to similar stresses, the powders do not lose much of its compressibility and densification behavior. It could still be useful in the formulation of tablets that would have good mechanical properties.

### Table 4: Parameters of Heckel plot

<table>
<thead>
<tr>
<th>Sample</th>
<th>K</th>
<th>Py (MNm$^{-2}$)</th>
<th>$D_o$</th>
<th>$D_A$</th>
<th>$D_B$</th>
<th>A</th>
<th>R$^2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>MCCF-Cocos</td>
<td>11.14</td>
<td>0.14</td>
<td>0.26</td>
<td>0.57</td>
<td>0.31</td>
<td>0.18</td>
<td>1.00</td>
</tr>
<tr>
<td>MCCL-Cocos</td>
<td>4.09</td>
<td>0.18</td>
<td>0.20</td>
<td>0.31</td>
<td>0.11</td>
<td>0.16</td>
<td>0.99</td>
</tr>
<tr>
<td>AV-102</td>
<td>4.46</td>
<td>0.22</td>
<td>0.19</td>
<td>0.91</td>
<td>0.00</td>
<td>0.21</td>
<td>0.98</td>
</tr>
</tbody>
</table>
Fig. 8: Disintegration efficiency of *MCCF-Cocos*, *MCCL-Cocos* and *AV-102*.

Fig. 9: Tensile strength of *MCCF-Cocos*, *MCCL-Cocos* and *AV-102*. 
4. CONCLUSION

The powder characterization test results show that variation in processing method such as drying procedure significantly affected the characteristics of the MCC derived from *Cocos nucifera* in terms of flowability and the indices associated with it. The bulk and tapped densities, flow rate, Carr’s Index, and packing fraction of the *MCCF-Cocos* were higher than *MCCL-Cocos* while angle of repose, Hausner’s quotient, porosity and particle density of *MCCL-Cocos* were higher than *MCCF-Cocos*. This obviously
means that the MCCF-Cocos flowed better than the MCCL-Cocos. The flowability of MCCF-Cocos was similar to that of AV-102. The densification, volume reduction and compactibility as assessed by the Kawakita model show that MCCF-Cocos differed from the MCCL-Cocos but was similar to AV-102. In terms of mechanical strength and tabletability, compacts formed from MCCL-Cocos were significantly (P = 0.01) stronger than the compacts formed from the MCCF-Cocos. Compacts of AV-102 were stronger than compacts of both MCCF-Cocos and MCCL-Cocos. Thus hardness, tensile strength, and disintegration of compacts formed from MCCL-Cocos and AV-102 were of higher value than those MCCF-Cocos while the compact friability was in the order AV-102 < MCCL-Cocos < MCCF-Cocos respectively. The hardness, disintegration time and friability were adjudged good because they conformed to both BP 2012 and USP 2009 specifications for uncoated tablets. Heckel evaluation showed that the MCCs undergo plasticity and slippage even on the application of low pressure which explains their good mechanical properties. All the MCCs: AV-102, MCCL-Cocos and MCCF-Cocos were quite reworkable but AV-102 had the highest reworking potential.

CONSENT (WHERE EVER APPLICABLE)

Not applicable.

ETHICAL APPROVAL (WHERE EVER APPLICABLE)

Not applicable.

REFERENCES

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[22]. Odeku OO, Awe OO, Poopola B, Odeniyi MA, Itiola AO, Compression and Mechanical Properties of Tablet Formulations containing Corn, Sweet Potato, and Cocoyam starches as binders, Pharmaceutical Technology, 2005: 82-90.


