

# 1 A COMPARATIVE STUDY OF MICROCRYSTALLINE CELLULOSE 2 ISOLATED FROM THE POD HUSK AND STALK OF FLUTED PUMPKIN

3 **ABSTRACT:** Microcrystalline celluloses (MCC) were prepared from  $\alpha$ -celluloses obtained from fluted pumpkin  
4 stalk and pod. The substrates were subjected to treatment with 2% (w/v) NaOH, 3.5% (w/v) NaOCl and 17.5% w/v  
5 NaOH solutions respectively to obtain alpha celluloses. Acid hydrolysis of the alpha-celluloses using 2.5 N  
6 hydrochloric acid were carried out. The study evaluates and compares the physicochemical properties of  
7 microcrystalline cellulose obtained from the pod and stalk of fluted pumpkin. Composition of cellulose,  
8 hemicellulose and lignin were also determined. Results showed cellulose; hemicellulose and lignin content of the  
9 pod husk and stalk were 49%, 26%, 9% and 41%, 24%, 26%, respectively. The morphology of the hydrolyzed  
10 MCCs' were investigated using scanning electron microscopy (SEM) and the results revealed the stalk (FS-MCC) to  
11 have an individual rod-like shaped fiber when compared with flat-shaped large aggregated form of the pod (FP-  
12 MCC). The particles sizes were also uneven with FP-MCC (6.689  $\mu\text{m}$ ) having larger particle sizes than FS-MCC  
13 (5.538  $\mu\text{m}$ ). The high cellulose content of the pod husk shows that the applications may be extended in the  
14 production of other cellulose derivatives while the high lignin content of the stalk shows that it may be used as  
15 alternative source of producing lignin for making textile dyes, coating and other agricultural chemical. Pod MCC  
16 (FP-MCC) had better physicochemical properties than the stalk MCC (FS-MCC).

17 **Key words:** Cellulose; MCC; Fluted pumpkin; SEM, Physicochemical

## 18 INTRODUCTION

19 Waste generation and accumulation have stimulated serious measure strategies not only in the Western  
20 World but also in the third World Countries. Reasonable efforts have been made to develop means for the recovery  
21 and utilization of biopolymers waste over the years. This became very important considering the fact that those  
22 wastes contain appreciable amount of dry matter, crude protein, fibre, ether extract, minerals, high molecular weight  
23 cellulose and hemicellulose which can be obtained at minimal cost [1, 2]. Cellulose, the most important chemical  
24 component in different lignocellulosic biomass (accounting for more than 50% by weight) has a linear homopolymer  
25 of glucopyranose residues, linked by  $\beta$ -1, 4 – glycosidic bond [3]. The chemical and mechanical degradations of  
26 cellulose result to the production of low molecular weight microcrystalline cellulose (MCC). MCC powder has a  
27 vast variety of applications in food, cosmetic, thin layer chromatography and pharmaceutical industries [4]. Wood  
28 and cotton linters have become the major precursors used in the production of MCC, but due to the negative impact  
29 of forest depletion on the environment worldwide, research is being focused on other alternative sources of MCC.  
30 Many studies have been reported using different non-woody sources to prepare MCC. These include groundnut  
31 shell, cereal straw [5,6], water hyacinth[7], bagasse and corn cob[8], Indian bamboo [9], sugar beet pulp [10], sugar  
32 cane bagasse [11-13], pineapple leaf [14], luffa cylindrical [15], banana plant waste [16] , peel of pear[17], orange  
33 mesocarps [18] , Pomelo peel [19], coffe husk [20] roselle fibers [21]etc.

34  
35 *Telfairia occidentalis Hook.f* generally known as fluted pumpkin is a perennial drought resistant crop which is  
36 grown as an annual crop under the West African traditional farming system. It is a dioecious herb which is coiled on  
37 creeping or usually branched tendrils with the root system ramifying the soil to top surface level. The stem is  
38 angular and glabrous which becomes fibrous when old. The fruit pod contains many ovoid seeds (about 192) of  
39 different genders of plant and the best method of harvesting this plant is by pruning [22, 23]. This creeping  
40 vegetative plant is cultivated and consumed in the South Eastern Nigeria extensively while the tender vine and  
41 foliage are consumed as potherb; the seed is consumed as nut. The leaf is rich in iron and finds herbal use in  
42 treatment of diabetes and anaemia [24]. The tender shoots, succulent leaves and immature seeds are cooked and  
43 consumed as vegetable. The plant also contains considerable amount of anti-nutrients such as tannin, phytic acid and  
44 saponin which may also be beneficial to the health [25, 26]. Processing of the wastes generated from this plant  
45 (fluted pumpkin stalk and pod husk) to  $\alpha$ -cellulose and subsequent conversion to microcrystalline cellulose for the

46 production of drugs, creams and lotion will add to its potentials and benefits to man. In this study, the  
47 physicochemical properties of the produced MCC will be evaluated and compared with commercial grade (C-MCC).

## 48 MATERIALS AND METHODS

### 49 Chemical reagents

50 Sodium hydroxide pellets, Sodium hypochlorite (Hypo® Multipro Enterprises Ltd, Nigeria), conc. hydrochloric  
51 acid, iodine crystals, zinc chloride (ZnCl<sub>2</sub>), potassium Iodide (KI), acetone (C<sub>3</sub>H<sub>6</sub>O), xylene,  
52 hexadecyltrimethylammonium bromide (C<sub>19</sub>H<sub>42</sub>Br) (Sigma-Aldrich, UK), conc. sulphuric acid, 2-octanol, hydrogen  
53 peroxide (H<sub>2</sub>O<sub>2</sub>), sodium dodecyl sulfate (Sigma-Aldrich, UK), EDTA disodium salt (dehydrate), Saturated KMnO<sub>4</sub>,  
54 glacial acetic acid, potassium acetate, tertiary butyl alcohol, silver nitrate (AgNO<sub>3</sub>), Fe(NO<sub>3</sub>)<sub>3</sub>.9H<sub>2</sub>O, oxalic acid  
55 dehydrate, ethanol, sodium borate, sodium phosphate dibasic (anhydrous), triethylene glycol, alpha amylase (Sigma-  
56 Aldrich, UK), sodium sulfite (Na<sub>2</sub>SO<sub>3</sub>) (Sigma-Aldrich, UK) and distilled water were utilized for the study.

### 57 Preparation of raw material

58 The fluted pumpkin pod husk and stalk were sourced from the sellers of the seeds and leaves at Main Market  
59 Onitsha in Anambra State and Mile 1 market in Rivers state, Nigeria respectively. The pod husk was soaked in water  
60 for 10 min, then washed properly to remove the pulp and other potential contaminants, while the stalk was cleaned  
61 with water. They were cut into irregular chips and then dried. The dried chips of the pod husk and stalk were  
62 pulverised using an electric grinder and then allowed to cool to room temperature. The resultant powdered pod husk  
63 and stalk of fluted pumpkin were sieved using 2.0 mm Laboratory test sieve (Endecotts Ltd London England).

## 65 COMPOSITIONAL ANALYSIS

66 In the determination of cellulose, hemicellulose and lignin content, a modification of the procedure by [27] was  
67 adopted.

### 68 Neutral detergent fibre (NDF)

69 5 g each of the samples were soaked in 50 mL of ethanol separately for 24 h, after which the samples were filtered  
70 and oven-dried at 105 °C for 6 h. 1 g of the dried biomass sample obtained after cold extraction with ethanol was  
71 weighed alongside with 0.5 g of sodium sulphite and placed in a Berzelium flask. 100 mL of the NDF solution was  
72 added at room temperature and refluxed for 1 h. Alpha amylase was added in dropwise to reduce splashing. At the  
73 end of the reflux period, each of the flask were removed separately and the sample solutions were slowly decanted  
74 into whatman 541 filter paper and vacuum filtered into a flask. Residues obtained were washed with several  
75 volumes of boiling water and oven-dried at 105 °C for 6 h, and then weighed to a constant weight.

$$76 \text{NDF} = \frac{[(\text{Dry filter paper} + \text{residue} - \text{Dry filter}) / \text{Initial sample weight}] \times 100}{77} \quad (1)$$

### 78 Acid-detergent fiber (ADF)

79 The dried residues obtained from NDF analysis were placed in Berzelium flask, added 3 drops of 2-octanol then  
80 refluxed for 1 h from onset of boiling. At the end of the reflux period, the sample solutions were filtered and washed  
81 thrice with boiling water and twice with acetone. The moist residues were oven-dried at 105 °C for 8 h, transferred  
82 to a desiccator and later weighed to a constant weight.

$$83 \text{ADF} = \frac{[(\text{Dry filter paper} + \text{residue} - \text{Dry filter paper}) / \text{initial sample weight}] \times 100}{84} \quad (2)$$

$$84 \text{Hemicellulose} = (\text{NDF}) - (\text{ADF}) \quad (3)$$

### 85 Determination of Permanent lignin (PML)

86 The dried residues obtained from ADF analysis were placed in a two different crucible immersed in a shallow  
87 enamel pan containing cold water. Saturated KMnO<sub>4</sub> was mixed with the buffer solution in the ratio of 3:1 and 25  
88 mL of that solution was added to the crucible. The crucible stand was allowed to stand for 90±10 min at room  
89 temperature while stirring continuously to break the lumps and draw up on sides of the crucible in order to wet all  
90 particles. At the end of delignification, the crucible was removed to filtering apparatus and suck dried, then placed  
91 back in the crucible and added demineralizing solution, after 5 min it was suck dried again then soaked back with  
92 demineralizing solution and left to stand till the fibre is white for 30 min. The resultant white fibre obtained was

93 washed twice with 80% ethanol and subsequently with acetone and then suck dried. The residue obtained was oven-  
94 dried at 105 °C for 6 h and then weighed.

$$95 \text{ PML} = [(\text{Dry filter paper} + \text{residue} - \text{Dry filter})/\text{initial sample weight}] \times 100 \quad (4)$$

$$96 \text{ Crude lignin containing ash} = (\text{ADF}) - (\text{PML}) \quad (5)$$

97 The resultant dried residue obtained after delignification was ashed. The sample was weighed and placed in a  
98 crucible and the ash content was estimated by weighing the residue that remained after combustion at 550 °C for 4 h  
99 until all the carbon is eliminated.

$$100 \text{ Pure lignin content} = \text{PML weight before ashing} - \text{ash content} \quad (6)$$

### 101 ISOLATION OF ALPHA CELLULOSE (POD HUSK)

102 A slight modification of the method by [28] was adopted in production of alpha cellulose from the pod husk. 1.2 kg  
103 of the sieved fraction of the pulverized dried pod husk was de-lignified using 18 L of 2% w/v sodium hydroxide  
104 solution in a stainless steel container immersed in a water bath (Precisdig (6001197) JP Selecta water bath) which  
105 was maintained at 80 °C for 3 h. The wet mass obtained after delignification was treated with 8 L of 3.5% w/v  
106 sodium hypochlorite solution, at 80 °C for 30 min. The resultant bleached mass was further digested with 4L of  
107 17.5% w/v aqueous sodium hydroxide solution at 80 °C for 1 h. Finally, the alpha cellulose obtained after washing  
108 was further bleached with 4 L of (1:1) aqueous solution of 3.5% w/v sodium hypochlorite at 50 °C for 20 min,  
109 washed severally with distilled water until the washings were neutral to litmus paper. It was filtered and squeezed  
110 through the muslin cloth to obtain a small mass and then oven-dried (JP Selecta Digiheat Oven) at 65±1.5 °C for 12  
111 h.

### 112 STALK

113 1000 g of the sieved fraction of milled, dried stalk was de-lignified using 5 L of 2% w/v aqueous sodium hydroxide  
114 solution in a stainless steel container immersed in a water bath (Precisdig (6001197) JP Selecta water bath) and  
115 maintained at 80 °C for 3 h. The moist material obtained after delignification was treated with 3.5 L of 3.5% w/v  
116 sodium hypochlorite solution at 80 °C for 30 min. The resultant bleached mass was treated with 3.750 L of 17.5%  
117 w/v aqueous sodium hydroxide solution at 80 °C for 1 h. The resultant moist residue (the crude alpha cellulose)  
118 obtained after several times of washing with distilled water was bleached with 1.4 L of 1:1 dilution of 3.5% w/v  
119 aqueous sodium hypochlorite solution repeatedly at 80 °C for 1 h until the material became milky white. Finally, the  
120 alpha cellulose was further treated with 2.5 L of 13.5% v/v hydrogen peroxide at 80 °C for 1 h to obtain a snow  
121 white colour, which was washed severally with distilled water until the washings were neutral to litmus paper. It was  
122 then filtered through the muslin cloth to obtain a small mass and then oven-dried (JP Selecta Digiheat Oven) at  
123 65±1.5 °C for 12 h.

### 124 PRODUCTION OF MICROCRYSTALLINE CELLULOSE

125 A slight modification of the method by [29] was adopted in preparing microcrystalline cellulose. The α-cellulose  
126 (109 g and 161 g) of pod and stalk respectively were each placed in a beaker and hydrolyzed with 2 L of 2.5 N  
127 hydrochloric acid at a boiling temperature for 15 min. The resulting mixture was poured into 3 L of distilled water  
128 followed by vigorous stirring, then allowed to stand overnight. The crystals obtained were washed with distilled  
129 water until neutral to litmus paper, filtered through a muslin cloth and then oven-dried at 65±1.5 °C for 12 h. Further  
130 milling and sieving were carried out to produce smaller crystals of the aperture sieve size of less than 250 μm.

### 131 PHYSICOCHEMICAL PROPERTIES

132 Identification, Organoleptic characteristics, starch and dextrin, solubility tests were carried out according to British  
133 Pharmacopoeia (BP) specifications [30]

134 pH: This was determined by shaking 1 g of each MCC with 50 mL of distilled water for 5 min and the pH of the  
135 supernatant liquid was determined using pH meter (pHep® pocket-sized pH meter).

136 **Moisture content:** 2 g of each of the powdered samples was weighed, transferred into a petri dish and then dried in  
137 an oven for 3 h at 105 °C to a constant weight. The moisture content (%) was then computed based on the initial air-  
138 dried weight.

139 **Bulk ( $B_d$ ) and Tapped ( $T_d$ ) density:** 5 g of each MCC samples was weighed and transferred into a 50 mL dry  
140 measuring cylinder. The volume occupied by the sample was noted as the bulk volume and the bulk density was  
141 determined by dividing the mass of the material by the bulk volume as expressed [31]:

$$142 \quad \text{Bulk density } (B_d) = [M / V_B] \quad (7)$$

143 Where M is the mass of the sample,  $V_B$  is the bulk volume of sample  
144 The measuring cylinder was then tapped on a wooden platform by dropping the cylinder from a height of one inch at  
145 2 seconds intervals until there was no observable change in volume reduction. The volume occupied by the material  
146 was recorded as the tapped volume. The tapped density was determined using the expression:

$$147 \quad \text{Tapped density } (T_d) = [M / V_T] \quad (8)$$

148 Where M is the mass of the sample,  $V_T$  is the tapped volume of sample

149 **True density ( $D_t$ ):** The true density was determined by the liquid displacement method by completely immersing  
150 the sample in a pycnometer bottle (26 mL) capacity using xylene as the immersion fluid. The volume of the liquid  
151 that was displaced was measured and the density was computed according to the following equation:

$$152 \quad \text{True density } (D_t) = [w / \{(a + w) - b\} \times SG] \quad (9)$$

153 where w is the weight of powder, SG is specific gravity of xylene, a represents sum of weights of the bottle and  
154 solvent and b represents the sum of weights of bottle, solvent and the MCC powder.

155 **Packing fraction ( $P_f$ ):** The packing fraction ( $P_f$ ) is expressed as the ratio of the bulk density ( $B_d$ ) to that of the true  
156 density ( $D_t$ ) as: [32]

$$157 \quad P_f = B_d / D_t \quad (10)$$

158 **Hausner's ratio ( $H_r$ ):** This is calculated as the ratio of tapped density to bulk density of the sample [15]

$$159 \quad H_r = T_d / B_d \quad (11)$$

160 **Powder porosity ( $e$ ):** This was determined from the values of true and bulk densities when fitted into the equation;

$$161 \quad e = 1 - B_d / D_t \times 100 \quad (12)$$

162 Where  $B_d$  is the bulk density,  $D_t$  is the true density and  $e$  is the porosity

163  
164 **Compressibility index  $C_1$ :** This was calculated by fitting bulk and tapped densities data into the equation as  
165 expressed by [15]

$$166 \quad \text{Compressibility index } (C\%) = [(T_d - B_d) / T_d] \times 100 \quad (13)$$

167  
168 **Hydration capacity ( $H_c$ ):** The method of [33] was used to determine the hydration capacity of MCC. 1 g each of  
169 the samples was placed in each of the four 15 mL plastic centrifuge tubes and 10 mL distilled water was added from  
170 a 10 mL measuring cylinder and then stoppard. The content was mixed for 2 min; then the mixture was allowed to  
171 stand for 10 min and was then immediately centrifuged at 1000 rpm for 10 min on a bench centrifuge (Sorvall-GLC-  
172 4). The supernatant was carefully decanted and the sediment weighed. The hydration capacity is determined as the  
173 ratio of the weight of the sediment to the dry sample weight.

$$174 \quad H_c = [(\text{weight of sediment} - \text{weight of tube}) / \text{oven-dried}] \quad (14)$$

175  
176 **Swelling capacity (Swellability) ( $S_C$ ):** This was determined at the same time as the hydration capacity  
177 determination using the method reported by [28] and was calculated as follows:

$$178 \quad \text{Swelling capacity } (S_C) = (V_2 - V_1) / V_1 \quad (15)$$

179  $V_1$  = tapped volume occupied by the sample prior to hydration  
 180  $V_2$  = volume occupied by sample after hydration  
 181 **Angle of repose (a):** The static angle of repose ‘a’ was measured according to the fixed funnel and free standing  
 182 cone method. A funnel was clamped with its tip 2 cm above a graph paper placed on a flat horizontal surface. The  
 183 MCC powders were carefully poured through the funnel until the apex of the cone thus formed just reached the tip  
 184 of the funnel. The mean diameters of the base of the powder cones were determined and the tangent of the angle of  
 185 repose was calculated using the equation:

$$186 \quad \text{Tan } a = 2h/D \quad (16)$$

187 where **h** is the height of the heap of powder and **D** is the diameter of the base of the heap of powder [34]

188 **Particle size analysis:** This was determined by microscopic method. The particles were viewed through the  
 189 trinocular microscope (SXY-m50) and the s-viewer application was used in taking accurate readings. The average  
 190 diameters of the magnified particles were determined for the sample size of 100 particles.

191 **Sieve analysis:** This was determined using a sieve shaker, containing standard test sieves ranging from 250 $\mu\text{m}$ -  
 192 63 $\mu\text{m}$  arranged in a descending order according to their aperture sizes. 20 g of each MCC powder was placed on the  
 193 topmost sieve (U.S.A standard test sieve, ASTM E-11 specification) and after 5 min of shaking; the weight of MCC  
 194 retained on each sieve was determined by subtracting the weight of the empty sieves from the weight of sieves  
 195 containing the powder. The percentage retained was then determined. The average diameter was calculated using the  
 196 following relationship as reported by [35]:

$$197 \quad \text{Average diameter of the MCC particles} = [\Sigma(\% \text{ retained}) \times (\text{mean aperture})] / 100 \quad (17)$$

198 **Degree of polymerization and Molecular weight determination:** This was determined at 25 °C using a Cannon-  
 199 fenske viscometer of the type described in the A.S.T.M standard method. 0.25 g of the sample (dry weight) was  
 200 dissolved in 50 mL of Cuprammonium hydroxide solution and 0.1 g of cuprous chloride was added and shaken until  
 201 dissolved and tested in a capillary viscosimeter. Five readings were made and the average of 3 measurements was  
 202 taken after discarding both the lowest and highest readings. The viscosity was determined from efflux time of the  
 203 sample solution ( $t$ ) and the blank solution ( $t_0$ ) using the equation of Solomon and Cuitta [36]

$$204 \quad [\eta] = \frac{[2(\eta_{sp} - \ln \eta_r)]^{1/2}}{C} \quad (18)$$

205  
 206 Where  $[\eta]$  is the intrinsic viscosity (cP)  
 207  $\eta_{sp}$  is the specific viscosity ( $\eta_r - 1$ )  
 208  $\eta_r$  is the relative viscosity ( $t_{\text{solution}}/t_{\text{solvent}}$ )  
 209  $C$  is the concentration of the sample ( $\text{g}/\text{cm}^3$ )  
 210  $t_{\text{solution}}$  is solution flow time (s)  
 211  $t_{\text{solvent}}$  is the solvent flow time (s)

212 **Degree of polymerization (D.P):** The degree of polymerization was determined from the viscosity using the  
 213 equation [37]

$$214 \quad \text{D.P} = 598.4 \ln [\eta] + 118.02 (\ln [\eta])^2 - 449 \quad (19)$$

215 **Molecular Weight:** The molecular weight was determined using the equation

$$216 \quad \text{DP} = \frac{M}{M_0} \quad (20)$$

217  
 218 Where  $M_0$  is the molecular weight of an anhydroglucose unit (AGU)

219  $M$  is the molecular weight of the material

220 **Scanning Electron Microscopy (SEM):** SEM was carried out on the sample to study the surface morphology. Each  
 221 of the samples was sputter-coated with gold for 3–4 min to dissipate the static charges occurring due to electron  
 222 bombardment and then observed in SEM, JSM 5400 (JEOL Ltd., Japan) at an accelerated voltage of 10 kV.



223  
224

## RESULTS AND DISCUSSION

**Table 1:** Some properties of MCC (FS-MCC and FP-MCC)

Test	FS-MCC	FP-MCC
Organoleptic characteristics	White, odourless, tasteless, granular	White, odourless, tasteless, granular
Identification (Iodinated zinc chloride)	Turns violet blue	Turns violet blue
Starch and Dextrin	Negative	Negative
pH	4.9	5.4
Water soluble substance (%)	≤ 0.99	≤ 0.64
Solubility (n-hexane, water, acetone, ethanol)	Insoluble	Insoluble

225

226 From Table 1, identification test gave a violet-blue colour in the samples indicating the presence of cellulose, while  
227 the solubility test showed that the samples were insoluble in the test solvents. No colour changes were observed for  
228 the starch and dextrin test and were thus considered absent. The organoleptic properties are acceptable as the  
229 materials were odourless, tasteless, white granular powders. The percentage yield of  $\alpha$ -cellulose obtained from pod  
230 husk and stalk were 18.2% w/w and 16.1% w/w respectively while the MCC yield obtained from their  $\alpha$ -celluloses  
231 were 88.16% w/w and 88.19% w/w respectively, thus the yield of MCC from their starting dry plant materials were  
232 approximately 16% w/w and 14% w/w respectively. As expected, the amount of MCC obtained was reduced due to  
233 large amount of amorphous regions that get solubilized and eliminated during acid hydrolysis.

234 The pH value of the pod husk was within the acceptable limit of 5-7.5 as specified in the British pharmacopeia [30]  
235 while that of the stalk was slightly lower than the acceptable limit. This can be enhanced by washing with excess  
236 water in order to ensure neutralization. From the pH values conclusion may also be drawn that the presence of  
237 sodium hydroxide is highly improbable. The composition of cellulose, hemicelluloses and lignin content of the pod  
238 and stalk were 49%, 26%, 9% and 41%, 24%, 26% respectively.

239 The water soluble substance values for the pod husk and stalk were not in compliance with the standard ( $\leq 0.26\%$  or  
240 12.5 mg) as stipulated by the British Pharmacopoeia[30]. The low value of water soluble substances can be  
241 attributed to the hydrolysis process, where glucose content of  $\alpha$ -cellulose is diluted; as a result the microcrystal  
242 cellulose became low indicating that the crystal phase had appeared in MCC [37, 38]. Thus, the high value obtained  
243 for the two samples (FP-MCC and FS-MCC) may be due to lower crystal phase appearance of the MCC.

244 **Table 2:** Comparative studies of powder properties of microcrystalline cellulose FS-MCC, FP-MCC and  
245 Commercial MCC (C-MCC))

Parameters	FS-MCC	FP-MCC
True density (g/ml)	1.340 (0.27)	1.310 (0.02)
Bulk density (g/ml)	0.197 (0.005)	0.240 (0.009)
Tapped density (g/ml)	0.340 (0.01)	0.390 (0.02)
Angle of repose ( $\Phi^0$ )	36° (1.04)	31° (0.6)
Hausner's ratio	1.600	1.720
Compressibility index (%)	42.23	37.66
Hydration capacity	3.070 (0.20)	3.071 (0.20)
Swelling capacity (%)	17.650 (0)	24.730 (1.86)
Moisture content (%)	6.920 (0.13)	6.250 (0.34)
Porosity (%)	85.300 (1.04)	81.730 (0.35)
Packing fraction	0.147(0.01)	0.183 (0.004)
Degree of polymerisation	248	220
Molecular weight (g/mol)	40176	35692

246 \*value is mean and standard deviation is in parenthesis, number of replicate =3

247 From Table 2, the moisture content determined for the samples were within the maximum acceptable limit of 7%  
248 [30]. The low % moisture content may be due to the crystalline structure. The crystalline phase finds it difficult

249 absorbing and storing water when compared to the amorphous phase. The FS-MCC has higher moisture content than  
 250 the FP-MCC. This may be due to having higher amorphous content portion than the crystalline portion. Regulation  
 251 of moisture in tablet or drug formulation is very essential as high moisture content may interfere with active  
 252 ingredient [32, 39]

253 Bulk and tapped densities of the powders provide information on their packing, compactness and densification  
 254 behaviour. The higher the bulk and tapped densities value, the better the potential for a material to flow as well as to  
 255 re-arrange under compression. From the results, FP-MCC had higher bulk and tapped densities when compared to  
 256 FS-MCC. This suggests that FP-MCC may have a better flow property than FS-MCC. The porosity value of FP-  
 257 MCC is lower than that of FS-MCC. This may be due to increased bulk density which is associated with decrease in  
 258 total pore space. From the results of packing fraction, FP-MCC exhibited the largest maximum volume reduction  
 259 due to packing when compared to FS-MCC.

260 The flow characteristics and frictional forces in powder samples were measured using angle of repose. Results  
 261 showed the angle of repose of FS-MCC is higher than that of FP-MCC. Decrease in particle size is suggested to lead  
 262 to increase in angle of repose of FS-MCC. From the limits of angles established as reported by [40], the samples  
 263 have reasonable flow potential.

264 The Carr's index and Hausner ratio predict the flow and compressibility of powders. The Hausers ratio and Carr's  
 265 index of FS-MCC was higher than that of FP-MCC. This may be due to the high moisture content of FS-MCC  
 266 which is said to directly decrease the flow property of the powder by increasing its cohesiveness of powder. From  
 267 the values established by the Carr's index and Hausners ratio, values obtained for the two samples show that they  
 268 have poor floor properties. Hence, addition of a glidant would be needed when using these materials in solid dosage  
 269 production process.

270 One common feature of all theories of disintegration is that penetration of water or any liquid medium must precede  
 271 disintegration and this can be assessed by the determination of the hydration capacity, swelling capacity and porosity  
 272 [41]. The swelling capacity showed a higher increase in volume of FP-MCC when compared with FS-MCC. This  
 273 demonstrates that FP-MCC may be a better disintegrant than the FS-MCC and if incorporated in tablet formulation  
 274 as a disintegrant, would probably produce tablet disintegration by two mechanisms: capillary or wicking and  
 275 swelling [32]. Furthermore, high swelling capacity of FP-MCC could also be that it has higher amorphous cellulose  
 276 content than [42]

277 Hydration capacity of polymers is directly related to its porosity, since the rate and extent of water uptake by a  
 278 material depends on the porosity. The hydration capacity of a material is a measure of the amount of water that can  
 279 be taken-up by the material [43]. The hydration capacity values of the two samples were almost close in terms of  
 280 numeric value. This shows that the two samples have high porosity. The degree of polymerization the samples were  
 281 within the standard limits (DP < 350) as stipulated by British Pharmacopoeia.

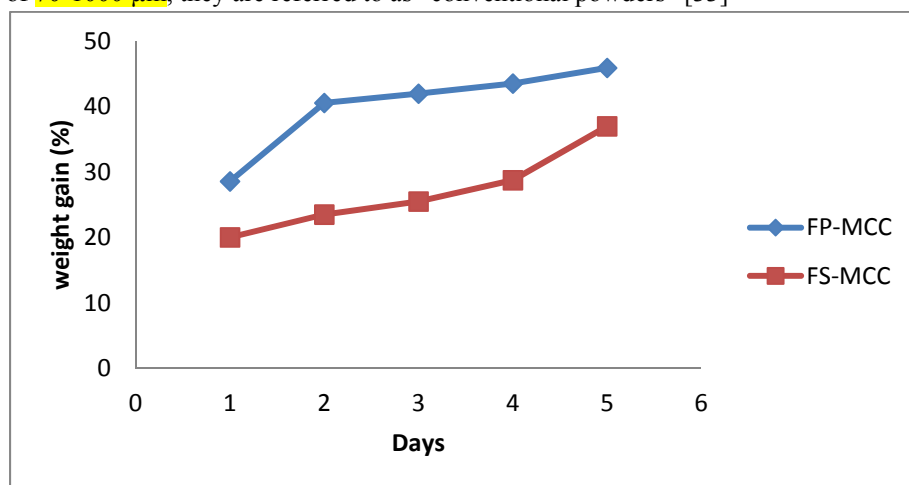
282 True density measures the density of a solid material excluding the volume of any open and closed pores. High true  
 283 density suggests high crystallinity which is also in accordance with the report by [42]. Thus, FS-MCC had a higher  
 284 true density value than that of FP-MCC

285 **Table 3:** Particle sizes of the cellulose

Samples	Weight (%) retained at the mean size aperture (µm)					Average diameter	Average particle size(µm)
	250	180	125	90	63		
FS-MCC	2.300	27.167	29.667	25.500	11.500	122.860	5.538
FP-MCC	1.833	34.717	25.550	16.000	15.000	121.930	6.689

286

287 The particle size analysis results shown in Table 3 above were in the range of 63–250  $\mu\text{m}$ . It was observed that over  
288 95% of the particle population was less than 250  $\mu\text{m}$  for both samples. The highest number of MCC particles were  
289 retained in sieve aperture of 180  $\mu\text{m}$  (35% out of the 20 g sample) for FP-MCC, while the highest number of MCC  
290 particles were retained in sieve aperture of 125  $\mu\text{m}$  (30% out of the 20 g sample) for FS-MCC. This revealed that  
291 FP-MCC had more of the particle sizes greater than 180  $\mu\text{m}$  while FS-MCC had more of the particle sizes less than  
292 180  $\mu\text{m}$  and the coarsest portion had the least quantity. The calculated average diameters were 123 and 122 for FP-  
293 MCC and FS-MCC respectively. The average diameters of the two prepared MCC samples were comparable may be  
294 due to the same acid (i.e HCl) used during hydrolysis unlike the results reported for bean and rice hull by [43],  
295 where two different acids were used (i.e. HCl and  $\text{H}_2\text{SO}_4$ ) which had effect on the average diameter of the resulting  
296 MCC samples. Furthermore, since the calculated average particle diameters of the two samples are within the range  
297 of 70–1000  $\mu\text{m}$ , they are referred to as “conventional powders” [33]

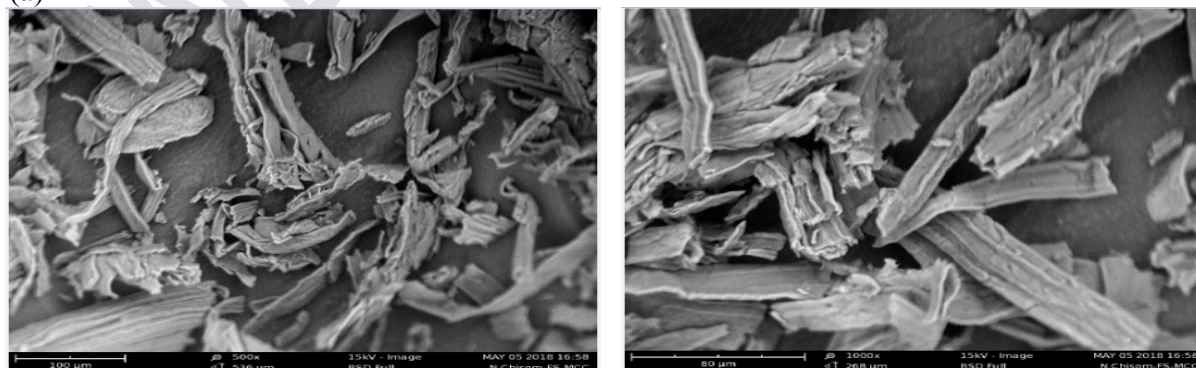


298

299 **Fig 1:** Moisture sorption profile of FS-MCC and FP-MCC

300 The moisture sorption capacity is a measure of moisture sensitivity of a material and it reflects relatively the  
301 physical stability of the tablets when stored under humid conditions [28]. From fig.1, results showed that FS-MCC  
302 adsorbed less moisture than FP-MCC. This could indicate that when FS-MCC is used in tablet formulation, it would  
303 adsorb the least moisture and thus eventually give tablets with better physical stability than FP-MCC. According to  
304 the report by [42], FS-MCC may probably have high crystalline portion of cellulose than FP-MCC which makes it  
305 adsorb less water.

306 (a)



307

308

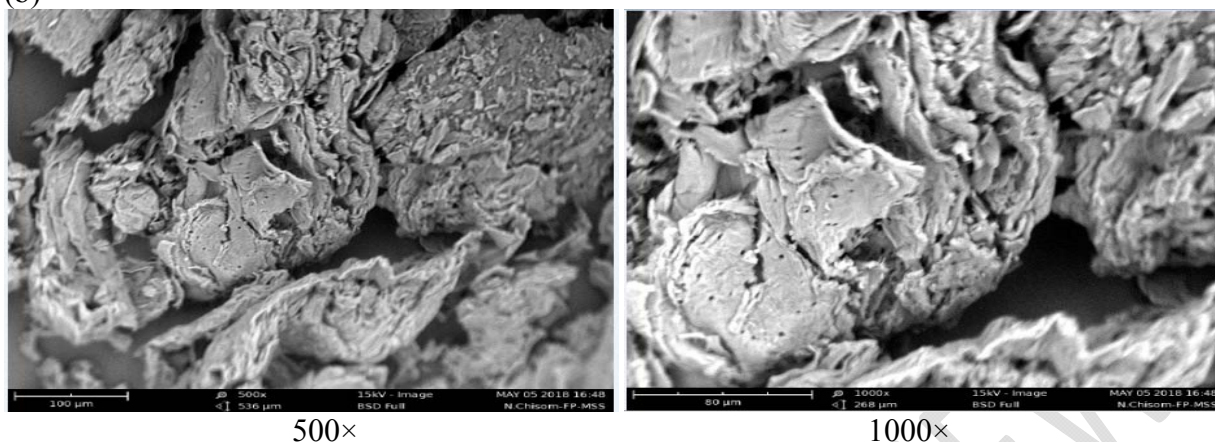
309

500×

1000×



310 (b)



311  
312 500×  
313 1000×  
**Fig 2:** Scanning Electron micrograph of (a) FS-MCC (b) FP-MCC

314 Fig 2 (a and b) are the results of the scanning electron micrographs of FP-MCC and FS-MCC at different  
315 magnifications. Results revealed that stalk-MCC (FS-MCC) exist as irregular, individual rod-like fibres with few  
316 bundle forms as well as an uneven surface while the pod-MCC (FP-MCC) exists as irregular, flat-shaped aggregated  
317 fibres. Both MCCs had a rough surface which favours the production of nanocrystals through hydrolysis [44].  
318 Particle shapes were also reflected in the porosity, as particles with larger sizes have lower porosity [45].

#### 319 CONCLUSION

320 Microcrystalline cellulose has been successfully prepared from the waste biomasses. Both MCC powders had poor  
321 flow properties which is not different from some other established reports. This may be resolved by the addition of  
322 glidant during solid dosage production. The Pod husk has high cellulose content when compared to the stalk. Thus,  
323 may be used as an alternative source of producing other cellulose derivatives, while the high lignin content of the  
324 stalk may be an alternative source or used for making textile dyes, coating and other agricultural chemical.

325  
326 **CONFLICT OF INTEREST:** The authors declare that they have no conflict of interest.

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