



## Enhancing Cassava Peels Starch as Feedstock for Biodegradable Plastic

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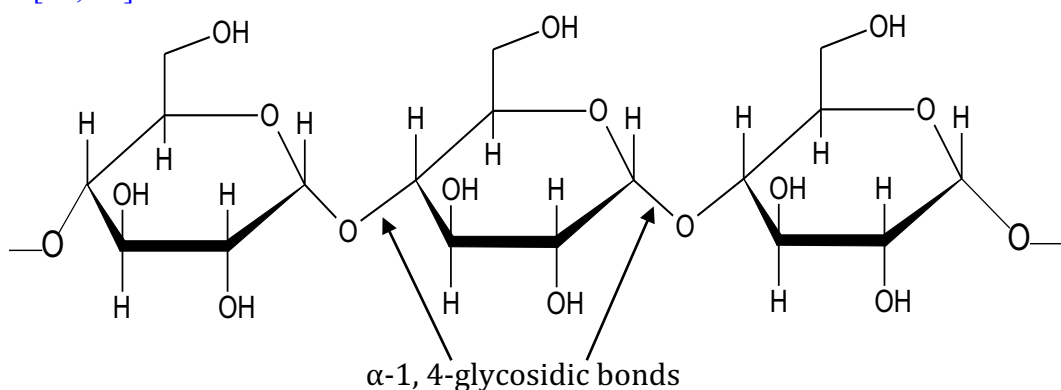
### Abstract

Cassava peels as an agro-industrial waste has potentials to serve as raw materials for industrial application. Growing interest geared towards harnessing the potential of these waste peels towards food sustainability has prospect for green synthesis. Cassava peels has high starch content that is biodegradable, inexpensive and abundantly available as polysaccharide molecule. Therefore modifying starch into biodegradable plastics that are less harmful to the environment than conventional plastics have attracted attention over the years because of its environmental sustainability. In the experimental procedure, 10 g of starch to sorbitol were prepared in the following proportions; 5 : 5, 6 : 4, 7 : 3, 8 : 2, 9 : 1 and 10 : 0. Further enhancement of starch to microcrystalline cellulose (MCC) with various concentration of sorbitol as plasticizer (10%, 20%, 30% and 40%) was investigated based on density, water absorption, solubility in water and biodegradability. Results from this study, reflect a clear indication that addition of microcrystalline cellulose (MCC) triggers lower density. The result also revealed that at starch to MCC ratio beyond 7:3, the impact of sorbitol on the bioplastic film begins to decrease. Use of sorbitol revealed an increase in water uptake after some hours with a 32 % lost in water-soluble organic matter. The biodegradability test, unveiled that the bioplastic without reinforcement with MCC, showed a higher percentage of degradation (55.46 %) after two weeks, indicating a higher weight loss. In conclusion, MCC addition triggered the enzymatic degradation more efficiently. These results also revealed the potential of MCC towards enhanced physical, and biodegradability properties.

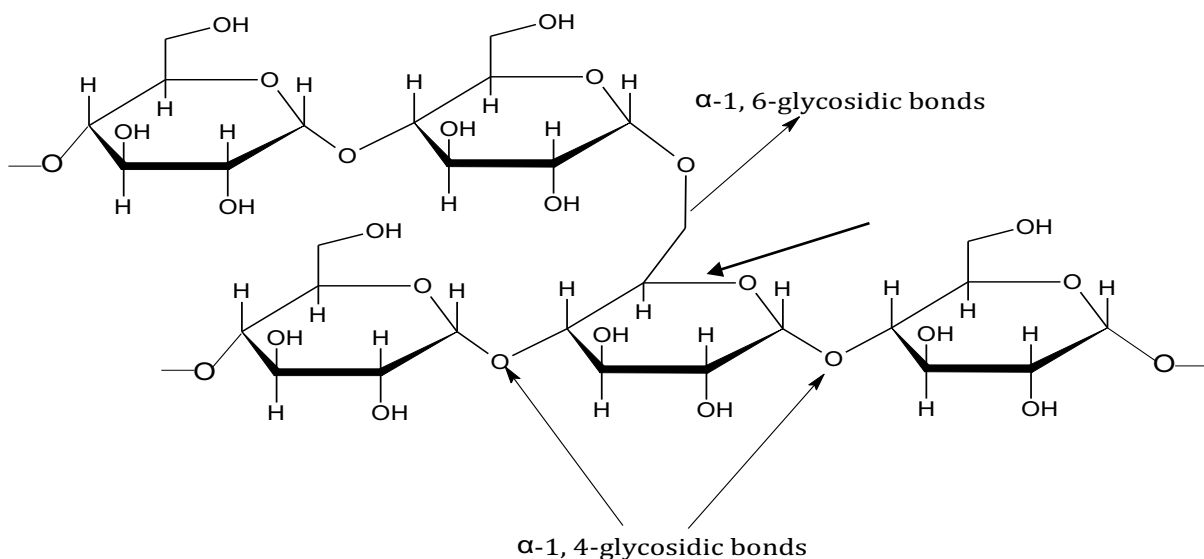
## 1. Introduction

Periodic accumulation of conventional plastic materials has raised concern over its waste management control protocol as it borders on their persistent nature in the environment [1-3]. Hence this factor has prompt current research focused in developing biodegradable materials targeted towards environmental sustainability [4-6]. The use of agro-industrial waste such as sugarcane bagasse, cassava peel, waste seed oil etc., has attracted growing interest due to the food sustainability mandate of World Health Organization (WHO) towards food security [7]. However, the source of the raw materials for bioplastics production is important [8, 9]. Therefore, natural raw materials for bioplastics production includes, polysaccharides (e.g. starch, cellulose, chitin and lignin), proteins (e.g. gelatine, casein and wheat gluten)

and lipids (e.g. plant oils and animal's fats) [10, 11]. Some agro-industrial waste has potentials to serve as raw materials for industrial application [12]. Biodegradability of bioplastics naturally in the environment is achieved when microorganisms in the environment metabolize and break down the structure of biodegradable plastic [13]. Biodegradable plastics are less harmful to the environment than conventional plastics [14]. Starch is one of the abundant natural raw material currently used for various industrial application, hence its application in bioplastic production has shown future prospect [15, 16]. Starch is a naturally occurring, biodegradable, inexpensive and abundantly available polysaccharide molecule [17, 18]. It is widely distributed in the form of tiny granules as the reserve carbohydrate in plants parts [19]. However, high paste viscosity, clarity and high freeze – thaw stability etc., are some important properties associated with starch, which upon various method of modification through chemical, physical and biological techniques can be harnessed into many useful products with industrial applications in food, paper, textiles, adhesives, beverages, confectionery, pharmaceuticals, building materials etc [20]. Starch can be extracted using various processes, depending on the plant source and end use [20, 21]. Starch a known polysaccharide carbohydrate consist of a large number of glucose units joined together by glycosidic bonds [22]. Its structural composition is made up of two major components: 22-26% amylose (a linear polysaccharide) and 74-78% amylopectin (a branched polysaccharide). The linear polymer units linked through an  $\alpha$ -D-(1,4)-glucosidic bonds is termed amylose, while the  $\alpha$ -D-(1,6)-glucosidic backbone linkage bonds, represents an amylopectin as shown in Figure 1 and 2 [17, 23]. Cassava (*Manihot esculenta Crantz*) is a perennial shrub grown throughout the lowland tropics for its starchy and thickened roots [24]. Its persistent nature makes it an all round year crop that can thrive in moderately fertile soil [25]. The quest for a waste crop with better starch yield, makes cassava peels a suitable alternative for this purpose. Therefore, the challenge of incessant disposal of waste peels of cassava after processing with subsequent danger in creating breeding grounds for microorganism will be tackled with a projection of converting waste to wealth. Starch modifications by chemical means have been reported to proceed via the following pathways; substitution, degradation or cross-linking [26]. The modification process has the capacity to disrupt the semi-crystalline starch granules thereby exposing the reactive hydroxyl groups of the amylopectin polymers, which becomes accessible to electrophilic reactants [26, 27].



**Fig 1.** Structure of Amylose [17]



**Fig 2.** Structure of Amylopectin [17]

Consequently, upon the poor mechanical properties resulting in its brittle nature, the need of an incorporated plasticizer for enhanced mobility of polymer by reducing their intermolecular forces with a corresponding increase in flexibility [28]. Glycerol and sorbitol are commonly used plasticizers in starch-based films as stated by Jantrawut *et al.* [29] and Maliuda *et al.* [10] who in their findings, noted that plasticizers only account for flexibility, but result in decrease tensile strength. In other study, Microcrystalline cellulose Avicel PH 101 was used as reinforcing filler because it offers higher density of hydroxyl groups on its surface that is available for hydrogen bonding [10, 30]. Therefore, the aim of this study is to evaluate the effect of reinforcement fillers and plasticizers on properties of bioplastics from cassava peels starch as agricultural waste source.

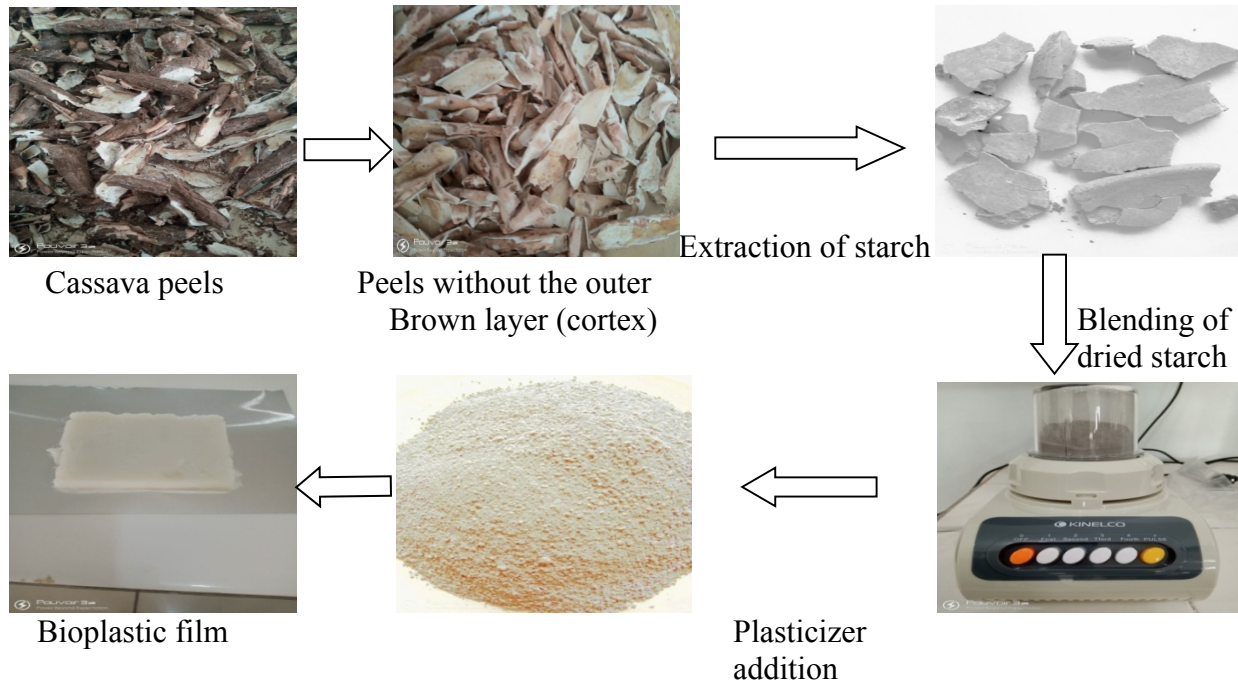
## 2. Experimental Details

### 2.1 Materials

Microcrystalline cellulose (MCC) Avicel PH101 was obtained from CDH Daryanj New Delhi-110002 India with particlesize 50  $\mu$ m, and sorbitol 99% was obtained from BDH chemical limited Poole England. All chemical reagents used in this study were of analytical grade.

### 2.2 Sample Collection and Extraction

The cassava peels used in this study were obtained from freshly harvested tubers at Agbahar-Otor market which is located in Ughelli North Local Government Area of Delta State, Nigeria on longitude 6° 2' 54" E / 5° 30' 40" N. The tubers were properly peeled and washed. The peels were further separated from the inner pulp (parenchyma) and the outer layer (cortex) with a clean knife. The separated cassava peels (100 g) was washed again with clean water before blending to small pieces with a blender. The blended cassava peels were then soaked in 100 ml of water for 45 minutes. Starch sediments were separated from the slurry. After separation, the residual white starch residue was sun-dried to remove all the moisture with further drying in an oven at 70 °C for removal of any residual water with subsequent blending into homogenized sizes [30].



**Fig. 3** Chart of starch extraction for bioplastic film production

### 2.3 Bioplastic Preparation

Method according to Maulida *et al.* [30] was adopted for this study with some modifications. The ration of 10 g mixture of starch and sorbitol without MCC were initially prepared in the following proportion; 5 : 5, 6 : 4, 7 : 3, 8 : 2, 9 : 1 and 10 : 0. The bioplastic formed were analyzed based on these variations. Starch solution was heated and stirred on a hot plate at 70°C to ensure gelatinization. The second phase of analysis also used a 10 g mixture of starch and MCC also prepared in the following proportion; 5 : 5, 6 : 4, 7 : 3, 8 : 2, 9 : 1 and 10 : 0. The various starch was dissolved in distilled water with starch mixture : distilled water = 1 : 10 (w/v) ratio. Starch solution were heated and stirred on a hotplate for 10 minutes. Furthermore, periodic enhancement with sorbitol as plasticizer with variation at 10%. 20%, 30% and 40% were added to the mixture at 70°C to ensure gelatinization. The bioplastic produced from both phase was cooled, poured onto a flat mold and dried in an oven at 60° C for 24 hours. The bioplastic obtained from the process was removed from the mold, placed in a desiccator for further analysis [30].

### 2.4 Bioplastic Characterizations

#### 2.4.1 Density

Standard of ASTM D792-91 method of analysis was used to evaluate the film Density of bioplastic film with size approximately 1x1 cm<sup>2</sup> obtained by weighing the film using a standard chemical digital weighing scale accurate to 0.1 g. The film volume was calculated by a water displacement method with a specified weight of the film, by multiplying the film area by the thickness [31-34]:

$$Density = \frac{mass (g)}{Volume (cm^3)}$$

### 3.4.2 Water Absorption

Water uptake of film samples was investigated by weighing film samples with an area of approximately 1 x 1 cm<sup>2</sup>. The samples were then dried at 70 °C for 3 hours, cooled, and then immediately weighed. The film samples were then submerged in distilled water for 3 hours without agitation. After the immersion period, the samples were then removed from the water and weighed. The percentage water absorption was calculated thus [30-32].

$$\text{Water absorption (\%)} = \frac{[M_2 - M_1]}{M_1} \times 100$$

Where:  $M_2$ =Final Weight,  $M_1$ =Initial Weight

### 3.4.3 Solubility in Water

Dried samples with an area of approximately 1 x 1 cm<sup>2</sup> were weighed. Each sample was subsequently immersed in 50 mL of distilled water under constant agitation for 3 hours at room temperature. Insoluble portion of the film was air dried for 24 hours and reweighed. The water solubility (%) of the films was calculated thus [30-32].

$$\text{Solubility(\%)} = \frac{W_0 - W_1}{W_0} \times 100$$

Where:  $W_0$ =Weight before submersion,  $W_1$ =Weight after submersion

### 3.4.4 Biodegradability Test

Biodegradation test method by Edhirej *et al.* [28] was adopted thus; Bioplastic film (2 x 2 cm<sup>2</sup>) were analyzed based on changes in weight and other physical morphology before and after the films were buried in soil under natural environmental conditions. Half of the film samples were buried in 1.5 cm of soil resulting in half a sample being buried while half is exposed to the open air. After a week, the samples were removed from the soil and gently cleaned by wiping gently with a soft brush. Samples were further air-dried for another week, inspected again and weighed. The percentage weight loss was calculated using the expression.

$$\text{Weightloss (\%)} = \frac{W_0 - W_1}{W_0} \times 100$$

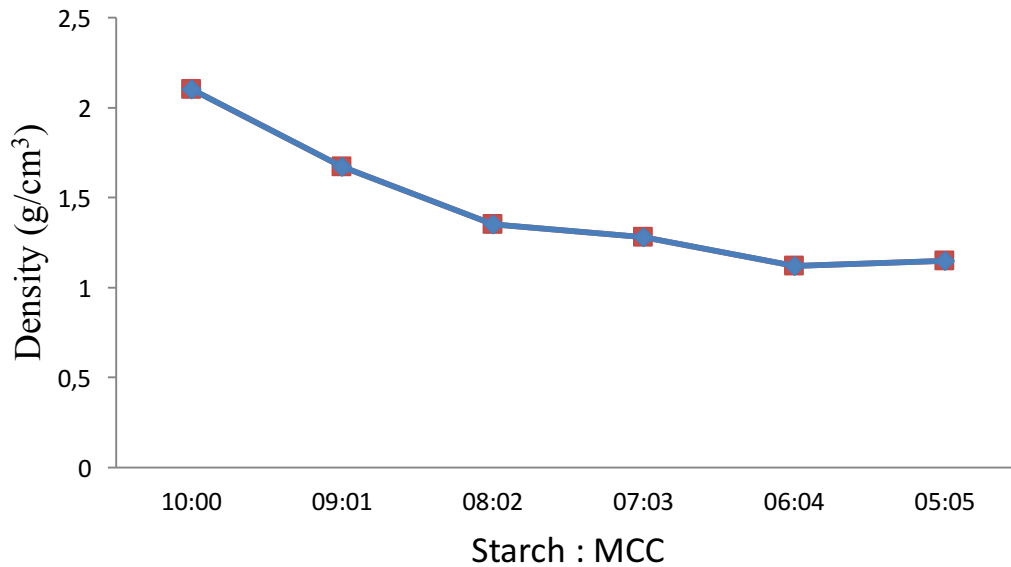
Where:  $W_0$ = InitialWeight,  $W_1$ = Final Weight

## 4. Results and Discussion

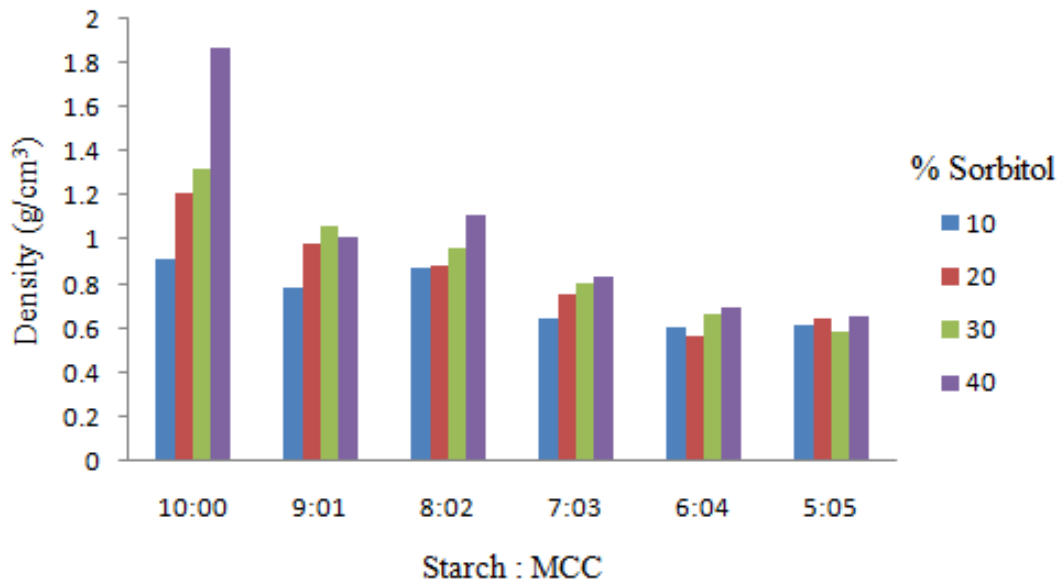
### 4.1 Density

The ratio of starch to sorbitol as it affects the density is shown in Figure 4. From the result, periodic increase in sorbitol results to subsequent decrease in the density. The result in this study recorded an average value of 2.10, 1.67, 1.35, 1.28, 1.12 and 1.15 for starch to sorbitol ratio of 10:0, 9:1, 8:2, 7:3, 6:4 and 5:5 respectively. The results reflect clear indication that increase sorbitol triggers lower density.

Result from this finding is higher than  $1.04 \text{ g/cm}^3$  as reported by Reis *et al* [35] and also higher than  $1.11 \text{ g/cm}^3$  as reported in other findings [33]. Likewise report from other study recorded higher values without MCC [30]. Figure 5 reflects a clear indication that further reinforcement with MCC, also affect the density of bioplastics. The study revealed that an increase in the MCC concentration with a corresponding increase in sorbitol result in the decrease density of the bioplastic.



**Fig. 4** Effect of the ration of starch to Sorbitol on the density of bioplastic film

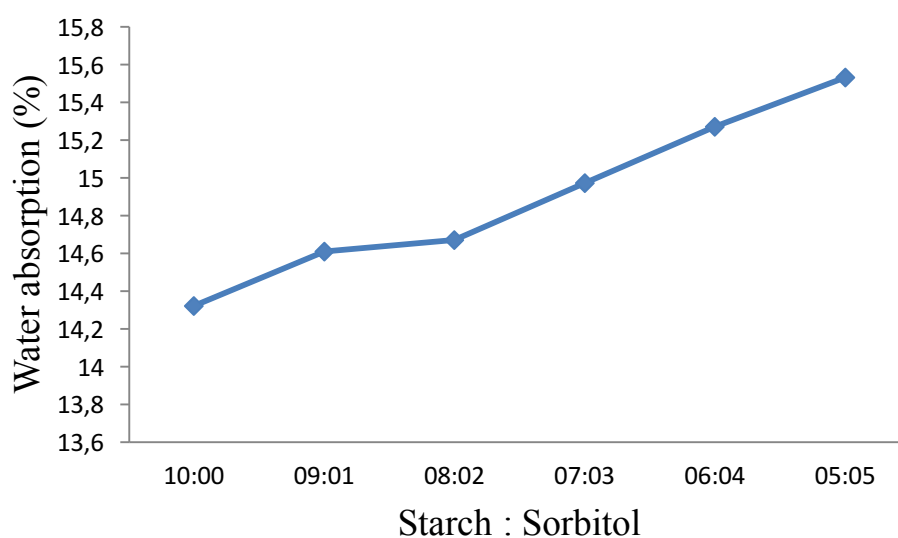


**Fig. 5** Effect of plasticizer concentration on bioplastic film

The result also revealed that at starch to MCC ratio beyond 7:3, the impact of sorbitol on the bioplastic film begins to neutralize. The result is in agreement with report by Sanyang *et al.* [36] on account that an increase in plasticizers content causes starch network to swell, thereby resulting in the decrease of network density. Results from this study are also in agreement with reports according to Maulida *et al.* [30]. This result is in line with report by Zhang *et al.* [37], who noted that the decrease density could be attributed to decrease in crystalline index of MCC after ultrasonic treatment. Also, Bierley *et al.* [38] and Hahladakis *et al.* [39], attributed lower density plastic to possess an open structure which can be penetrated by fluids, such as H<sub>2</sub>O, O<sub>2</sub> or CO<sub>2</sub>. Crystallinity leads to an increase in amorphous fraction that has attributes to lower polymer mass because of the un-uniformity and less dense molecules that result in lower density [37].

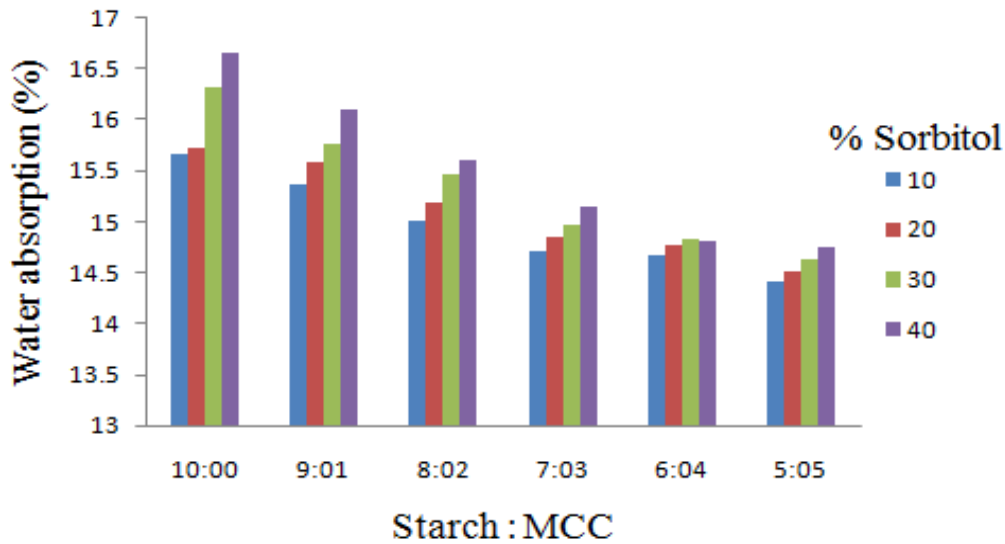
#### 4.2 Water Absorption

Increase in the water absorption was observed with subsequent increase in the Sorbitol content as reflected in Figure 6. This trend is in line with the assertion that describes the hydrophilic property of cellulose, thereby influencing their water sensitivity [40]. Report in other finding according to Maulida *et al.* [30], further supported the claim by noting that cellulose has strong hydrogen bond and characteristic that has the difficulty to bond compared to starch. Report for the water absorption from this study was slightly higher than reported value within the range of 12.9 – 13.9 for various reaction ratio of corn and rice starch [41], but is lower compare to reported values as reported in other findings [42]. Further enhancement with sorbitol as plasticizer as shown in Figure 7, revealed an increase in water uptake after some hours. This trend was accounted for based on the high hydrophilic property of the solid sugar alcohol [43]. Consequently, other findings reported that the gelatin content in the waxy starch films would increase the water solubility of the thermoplastic starch [44].



**Fig. 6** Effect of Sorbitol on the water absorption of bioplastic film

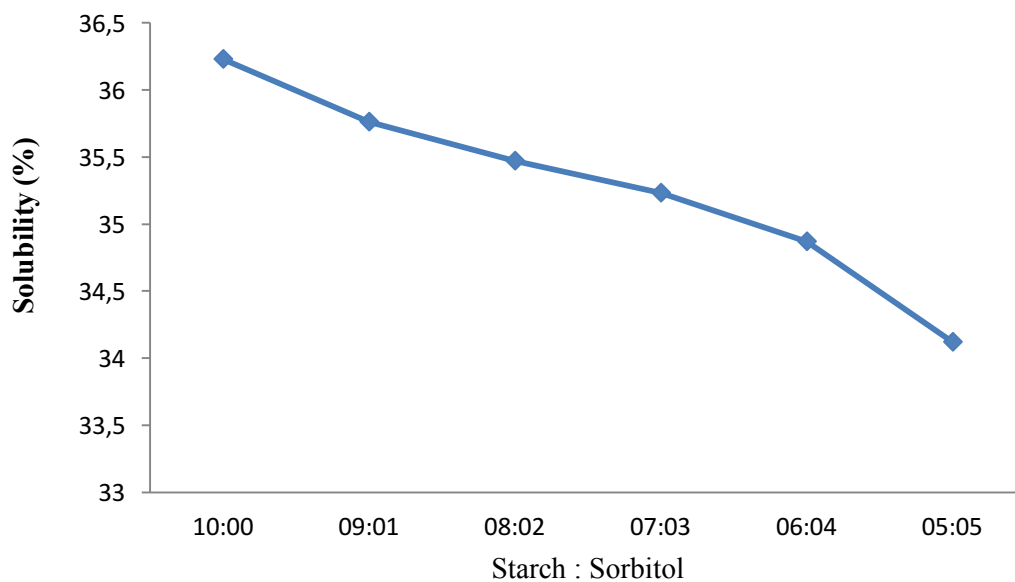




**Fig. 7** Effect of plasticizer on water absorption

### 4.3 Solubility in Water

From the earlier statement on the strong hydrogen bond and characteristic that limits effective bonding of cellulose with water. [Figure 8](#) reveals a decreasing solubility with increase MCC, thereby confirming their weaker polar property. Decrease solubility trend reported in this study is in line with other findings [\[42\]](#). The study unveils average of 32 % lost in water-soluble organic matter which is below reported value of 40 % [\[42\]](#).



**Fig. 8** Effect of MCC on the solubility of bioplastic film



These results suggest that the loss of soluble matter corresponds basically to the hydrophilic character of plasticizer used, which are easily release into the medium. These results are similar to report in other findings, thereby attributing this outcome to the loss of plasticizer [45, 46]. Lower solubility in water is a vital property towards an effective bioplastic property, hence this study highlights 30 % sorbitol concentration with a lower solubility as shown in Figure 9.

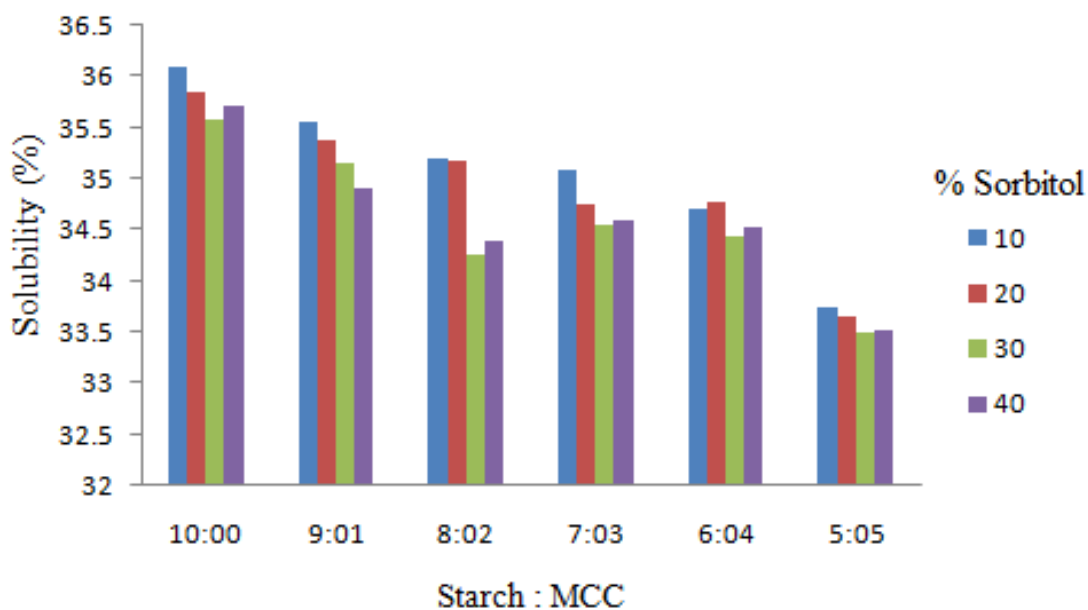


Fig. 9 Effect of plasticizer on bioplastic solubility

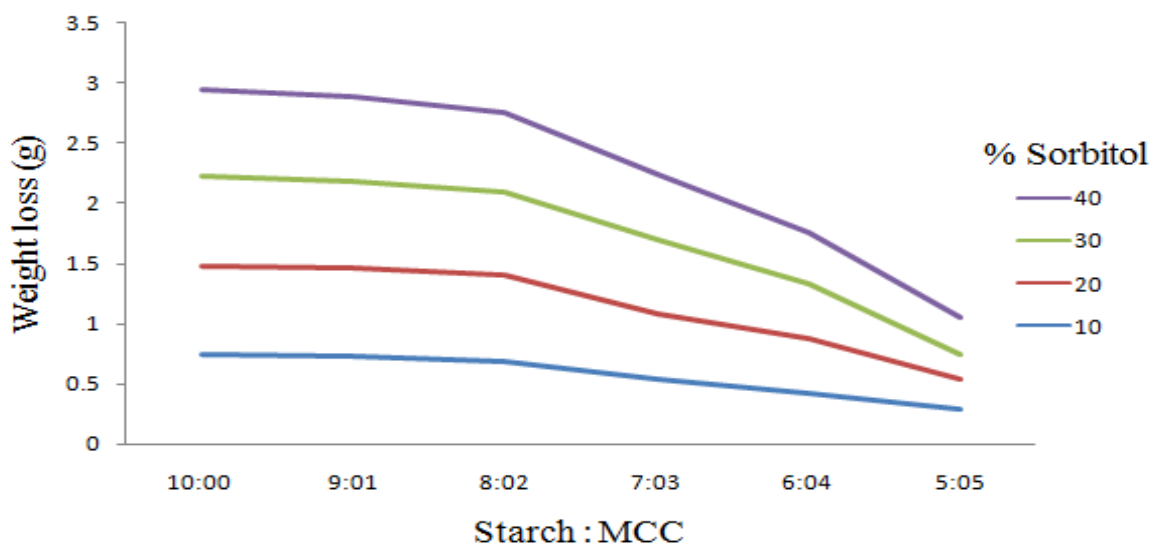
#### 4.4 Biodegradability Test

As shown in Table 1, the biodegradability test, unveiled that the bioplastic without reinforcement with MCC, showed a higher percentage of degradation (55.46 %) after two weeks, indicating a higher weight loss. Consequently, upon addition of filler (MCC), there was a decline in the biodegradability. The increase weight loss of the bioplastic could be as a result of the presence of insoluble, crystalline microfibrils, which are highly resistant to enzymatic hydrolysis [47]. The result from this study is in agreement with research by Mukuze *et al.* [33], who reported similar trend. Furthermore, Laxmeshwar *et al.* [47], noted that crystalline regions are more difficult to degrade, hence the ability of films to degrade depends greatly on physicochemical characteristics of the substrate, such as the degree of crystallinity and polymerization of cellulose, of which the crystallinity degree of cellulose is the most important structural parameter.

Subsequent reinforcement with sorbitol, showed only a slight resistance in the biodegradability at 10:0 (Starch : MCC) as shown in Figure 10. This finding is in line with report by Rinaldi *et al.* [48] who noted that longest decomposition time in all concentration of sorbitol for plastic without the addition of carboxymethyl cellulose (CMC).

**Table 1** Weekly biodegradability test

Starch : MCC	Initial weight (g)	Weight (g) after Week 1	% Weight Loss	Weight (g) after Week 2	% Weight Loss
10:0	1.1	0.74	32.73	0.49	55.46
9:1	0.87	0.76	12.64	0.68	21.84
8:2	0.78	0.71	8.97	0.59	24.36
7:3	0.62	0.57	8.07	0.47	24.19
6:4	0.49	0.45	8.16	0.39	20.41
5:5	0.25	0.23	8.00	0.20	20.00



**Fig. 10** effect of Sorbitol on weight loss of bioplastic film

### Conclusion

Cassava waste peels can be harnessed into viable raw material as biodegradable plastic. Improve physiological properties in this study was accounted for through the addition of moderate amount of MCC as an additive to minimize the plastic water absorption. A superior water absorption was obtained with a composition of 5:5 ratio of Starch : MCC with a 40 % sorbitol. The results also reflect a clear indication that addition of MCC triggers lower density, thereby making the film a good packaging biomaterial. Furthermore, the study revealed a decreasing solubility with increase MCC, thereby confirming their weaker polar property. The biodegradability test, unveiled that the bioplastic without the reinforcement with MCC, showed a higher percentage of degradation (55.46 %) after two weeks, indicating a higher weight loss. Consequently upon the addition of filler (MCC), there was a decline in the biodegradability. Therefore It can be concluded that MCC influenced the physical, biodegradability properties of starch-based bioplastics. Increase hydrophobicity was also evident with increase MCC. Moreover, MCC addition triggered the enzymatic degradation more efficiently. These results indicated that MCC performed an important role to enhance the physical, and biodegradability properties.

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