



## Studies of the Static Mechanical Properties of Polypropylene-Filled Calabash Particles Bio-Composites

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### Abstract

*Production of eco-friendly bio-composites has been the new trend in composite manufacture by polymer scientists and engineers since synthetic polymers are non-biodegradable and constitute environmental hazards when disposed indiscriminately. This study utilized calabash particles (CP) as filler in Polypropylene (PP) matrix to produce bio-composites. The calabash which were grinded and sieved into 125 $\mu$ m were introduced at 10% variation to 50% filler loading followed by compounding and compression moulding. Two categories of composites were prepared, in one (CP/PP A), Maleic Anhydride (MA) as coupling agent was introduced at 3% on the weight of the filler together with an initiator Benzoyl Peroxide (BP) at 1% to enhance the interfacial adhesion between the CP and PP and in another (CP/PP B) no coupling agent was used. The result of mechanical properties i.e. tensile strength, flexural strength, impact strength, hardness and tensile modulus for composites with coupling agent showed increase from 0 to 40% filler loading representing 9.30, 24.60, 37.84, 45.78 and 31.43% increase respectively, but showed decreasing trend for composites without coupling agent from 0 to 50% with the exception of tensile modulus and hardness which increased from 0 to 50% filler loading representing 13.51 and 36.76% increase respectively. Also, SEM micrographs for fractured surfaces of tensile test samples showed signs of even dispersion of the filler particles in the composite structure with coupling agent and showed signs of uneven dispersion of the fillers in the composite structure and voids for composites without coupling agent. From the results obtained, it can be deduced that the composites produced can be used for application that require moderate strength and cheaper applications like utensils, bowls, plates etc.*

**Keywords:** Bio-composites, Calabash Particles, Composites, , Flexural Strength, Polypropylene, Tensile Strength.

### 1. Introduction

Environmental issues caused by human activities such as global warming cannot be over looked since human population is on the rise and we have limited earth's resources, this is a point of concern in almost all the fields of engineering (Umar et al., 2015). One of the activities of man which calls for environmental concern is disposal of synthetic polymer wastes which are non-biodegradable (Chaichi, et al., 2016). Therefore, both scientists and industrial researchers are now utilizing renewable materials in many products and processes (Nanda et al., 2020).

Biodegradability and availability of natural fibres from plants and animals as compared to non-biodegradable synthetic fibres (Mandal et al., 2020), has advanced the idea of substituting non-biodegradable synthetic fibres with



natural-based fillers in polymer composites and its now a popular demand (Olga and Tomasz, 2017). One good source of natural cellulosic fillers that can serve as good replacement for synthetic fibres in the production of bio-composites is calabash (Naveen et al., 2019). Calabash (*Lagenaria ciseraria*) simply put is a dried hollow seed of a gourd used for household utensils (Apollos, 2017), its fresh fruit is made up of smooth skin, light-green, white flesh and rounder varieties are called calabash gourds (Sonny and Mike, 2017). The economic importance of this fruit, its cellulosic abundance and almost totally absent in modern day applications, when fully utilised in manufacture of composites can make this fruit a promising solution to our environmental, climatic and economic, problems (Hu, et al., 2020).

As a sustainable solution to of the world's petroleum supply uncertainty, bio-composites are eco-friendly that consist of bio-plastics and agro-fillers, are of great advantage to materials and research disciplines and as a reliable solution to growing environmental threats (Umar et al., 2015). Due to their chemical resistance, sufficient sources, low production cost, light weight, great diversity, good physical properties, as well as various efficient manufacturing processes, thermoplastics form a new trend in material development, therefore, there are a great number of plastic products commercially available (Jia-Horng et al., 2015), in the other way round, due to their insufficient strength, dimensional instability and stiffness, most of these thermoplastics are not suitable for load bearing applications (Salisu et al., 2015).

Better mechanical properties and dimensional stability has recently made wood-plastic composites (WPCs) to become suitable as building materials (Adhikary et al., 2007) and has attracted much attention and significant interest as they are replacing wood in many fields of applications (Yali et al., 2012). Meanwhile the dissimilarity of the surface chemistry between natural cellulosic filler and synthetic polymer matrix usually contribute to the decrease of tenacity in filler-polymer systems (Akil et al., 2006) CP are hydrophilic while HDPE is hydrophobic. To improve interfacial adhesion between cellulosic fillers and synthetic polymer matrices coupling agents can be incorporated. Therefore, in order to reduce the non-biodegradability of synthetic polymers, biodegradable fillers can be incorporated into synthetic polymers to produce composites that are of biodegradable, low cost and eco-friendly (Muralidhara et al., 2020). This research utilised the cellulosic abundance of calabash particles and used it as filler in polypropylene matrix to produce bio-composite incorporating maleic anhydride as coupling agent to improve the interfacial adhesion. The use of calabash particles as fillers in polypropylene to produce bio-composites is the first of its kind to the best of my knowledge, also introducing calabash as filler in polypropylene matrix will aid in accelerating biodegradability of polypropylene, adding value to calabash and introducing it into modern applications.

## **2. Materials and methodology**

### **2.1 Materials**

Polypropylene (PP) (Zayo-Sigma Chemicals Ltd. Jos, Nigeria), Maleic Anhydride (MA) 98% purity (coupling agent), Benzoyl Peroxide 98% purity (initiator), Calabash Particles (CP), Retsch Sieve Shaker Machine (Emdocatt. Model: 7416), Laboratory Mill (Model 4, Arthur H. Thomas Company, PA., U.S.A.), Vacuum Oven (model: 60648,



Cole Parmer), metallic mould, Two-roll Mill (Model: 5183 North Bergen U.S.A), Compression Moulding Machine (model: 0557) Wenzhou Zhiguang shoes making machines CO., Ltd. China.

## 2.2 Methods

### 2.2.1 Collection of materials

Calabash bowls (dried seeds and inner whitish material scraped and removed) were obtained from Zurmi, in Zurmi Local Government Area of Zamfara State, Nigeria. These calabash bowls are of between 10 to 15 litres in volume. The calabash bowls were broken into smaller pieces which were then fed into a laboratory milling machine and further crushed into smaller particle sizes. The ground calabash particles from the milling machine was sieved to 125µm particle size using a standard sieve, this particle size was found to be the smallest with reasonable quantity of particles obtained. Moisture was removed from the sieved particles in an oven until a constant weight was obtained and thereafter stored in a desiccator.

### 2.2.2 Preparation of the composites

The composites were prepared from the calabash particles and PP at 10% varying filler loading up to 50% while the PP was reduced simultaneously from 100% down to 50%, during compounding using a two-roll mill, reducing the percentage of PP will aid in reducing the cost of the composite since virgin PP is costlier than CP. The composite was prepared in two forms, one with coupling agent (CP/PP A) and the other without coupling agent (CP/PP B), the coupling agent; maleic anhydride (MA) was introduced 3% on the weight of the filler during compounding followed by compression moulding and cooling of the composites for 10 minutes each.

**Table 1: Percentage composition of CP (125µm) and PP in each of the composite formulation at different filler loadings**

S/NO.	CP (%)	PP (%)	MA (%)
1	0	100	0
2	10	90	3
3	20	80	3
4	30	70	3
5	40	60	3
6	50	50	3



### **2.2.3 Tests for the composites**

#### **2.2.3.1 Tensile strength (ASTM D-638)**

Tensile strength test was carried out according to ASTM D-638 standard with multifunctional electric tensile strength tester (YG026D) at Standards Organization of Nigeria in Kaduna State. The machine load cell of 300kg capacity was used for each of the samples. The test sample dimensions were 50mm x 10mm x 4mm (length, width and thickness) respectively.

#### **2.2.3.2 Tensile modulus (ASTM E111 – 17)**

Tensile modulus was carried out according to ASTM with multifunctional electric tensile strength tester (YG026D) at Standards Organization of Nigeria in Kaduna State, Nigeria. The machine load cell of 300kg capacity was used for each of the samples. The test sample dimensions were 50mm x 10mm x 4mm (length, width and thickness) respectively.

#### **2.2.3.3 Flexural strength (ASTM D-790)**

Flexural strength test was carried out according to ASTM D-790 standard using a three point bending system with Universal Material Testing Machine (Cat Nr. Model: 26) at the Department of Mechanical Engineering, ABU, Zaria, Kaduna State, Nigeria. The machine has a maximum force of 1 kilo newton (KN), and sample dimensions were 100mm x 25mm x 8mm (length, width and thickness) respectively.

#### **2.2.3.4 Impact strength (ASTM D-256)**

Impact strength test was carried out according to ASTM D-256 standard with Charpy Impact Testing Machine at The Department of Metallurgical and Materials Engineering, ABU, Zaria, Kaduna State Nigeria. The machine load of 15J capacity was used for each of the samples. The test sample dimensions were 80mm x 10mm x 10 mm (length, width and thickness) respectively.

#### **2.2.3.5 Hardness test (ASTM E-384)**

Hardness test was carried out according to ASTM E-384 standard with a Vickers Hardness Tester Machine Model MVI-PC YADRAN-416145, INDIA, with a diamond indenter at Shell Professorial Laboratory Building, ABU, Zaria. The machine load of 500gf at 15 sec was used for each of the samples whose dimensions were 30mm x 25mm x 8mm (length, width and thickness) respectively. The test was repeated five times each and an average test result was calculated and recorded

#### **2.2.3.6 Scanning Electron Microscopy test (SEM) (ASTM E766-98)**

SEM test was carried according to ASTM E766-98 standard using fractured surface of the tensile specimens with Pro: X: Phenom world. Model number 800-07334 at the Department of Biology, Umaru Musa Yaradua University, Katsina. To impart conductivity to the polymer composites, a sputter machine used 5nm gold to coat the sample surface. The samples were inserted into the machine using the sample holder and the morphology of the samples were viewed at 15KV, and Images were obtained at 1000X magnifications

## Results

### 3.1 Tensile strength

From Fig. 1 Tensile strength of CP/PP A composite with coupling agent can be seen to be increasing with increased filler loading from 35.32 MPa of the neat polymer to 38.94 MPa at 40% filler loading i.e. 9.30% increase.

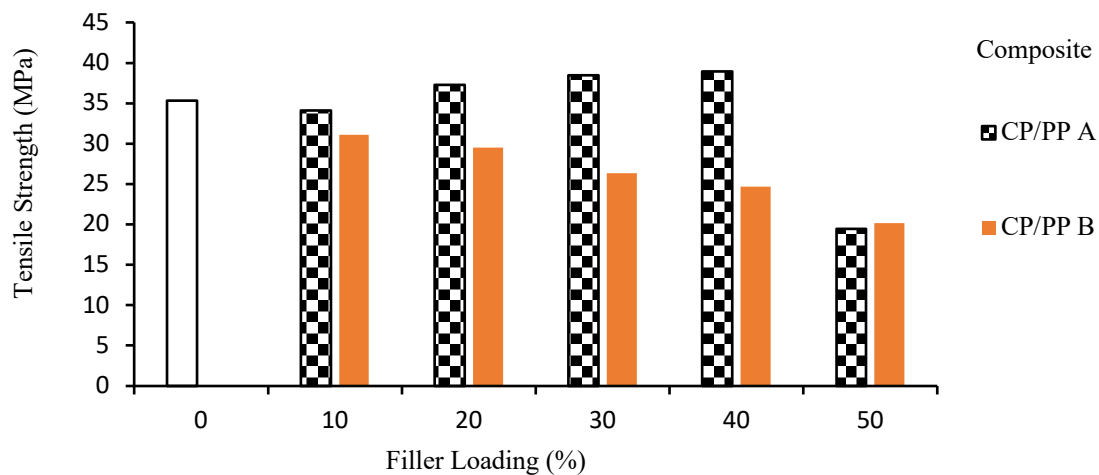


Fig. 1: Tensile strength of CP/PP composites A with coupling agent and B without coupling agent at varying filler loading

The increase might be attributed to the interfacial adhesion between the filler and the matrix due to the introduction of Maleic Anhydride at 3% on the weight of the filler, interfacial treatments serve to tailor properties of two dissimilar systems in composites by increasing bonding between filler and matrix (Silva et al., 2010). Due to variation in polarity between polar wood fibres and non-polar polyolefin, difficulty in obtaining uniform dispersion is experienced in their composites which in turn reduced the efficiency with which the fibres reinforce the polymer, but introduction of coupling agent into a composite system can improve mechanical properties (Sumit and Kamal, 2015).

Tensile strength of CP/PP A dropped by about 50% at 50% filler loading, this might be as a result of the matrix approaching its saturation level thereby losing its binding ability (Baba, et al., 2020b).

CP/PP B composites without coupling agent showed a steady decrease from 27.54 MPa of the neat polymer to 12.20MPa at 50% filler loading, about 56% decrease, the decrease might be attributed to poor interfacial adhesion between the CP and PP (Pavan, et al., 2018) since there exist no interfacial treatment in this composite system.

### 3.2 Tensile Modulus

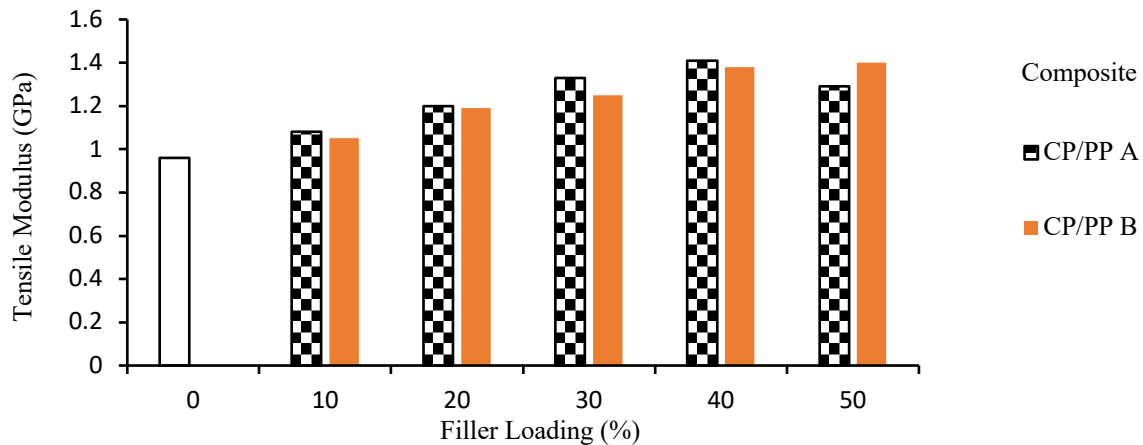


Fig. 2: Tensile modulus of CP/PP composites A with coupling agent and B without coupling agent at varying filler loading

From Fig. 2, it can be observed that tensile modulus of CP/PP composites was increasing from 0.96GPa of the neat polymer to 1.40GPa (31.43% increase) and 1.11GPa (13.51% increase) for composites A and B respectively. The increase in tensile modulus might be attributed to the stiffening effect caused by introduction of rigid fillers which might have reduced the polymer chain mobility consequently increasing modulus (Fatih and Kadir, 2012), also in CP/PP composite A, utilization of MA as coupling agent with increase in filler loading might have led improved interfacial adhesion requiring more force to deform the composite thereby increasing modulus (Jia-Hornng et al., 2015).

### 3.3 Flexural strength

Fig. 3 shows similar trend with tensile strength, the flexural strength of CP/PP composites A was increasing with increase in filler loading from 65.50MPa of the neat polymer to 86.87MPa at 40% filler loading representing 24.60% increase. The increase in flexural strength might be attributed to incorporation of maleic anhydride as coupling agent which might have improved the bonding between the CP and PP. Increase in interfacial bonding between filler and matrix improves mechanical properties (Igwe and Onuegbu, 2012), because improved filler matrix interaction provides effective transfer of stress from matrix to filler (Adamu et al., 2020). Also, there was a sharp drop of flexural strength from 87.87MPa at 40% filler loading to 62.45MPa at 50% filler loading, this might be as a result of saturation level of the composite being reached reducing binding ability of the matrix on the filler (Kai et al., 2018).

Meanwhile, CP/PP composites B shows a decreasing trend from 65.50MPa of the neat polymer to 47.82MPa (26.99% decrease) at 50% filler loading, the decrease might be attributed to poor interfacial adhesion between the filler and matrix (Igwe and Onuegbu, 2012).

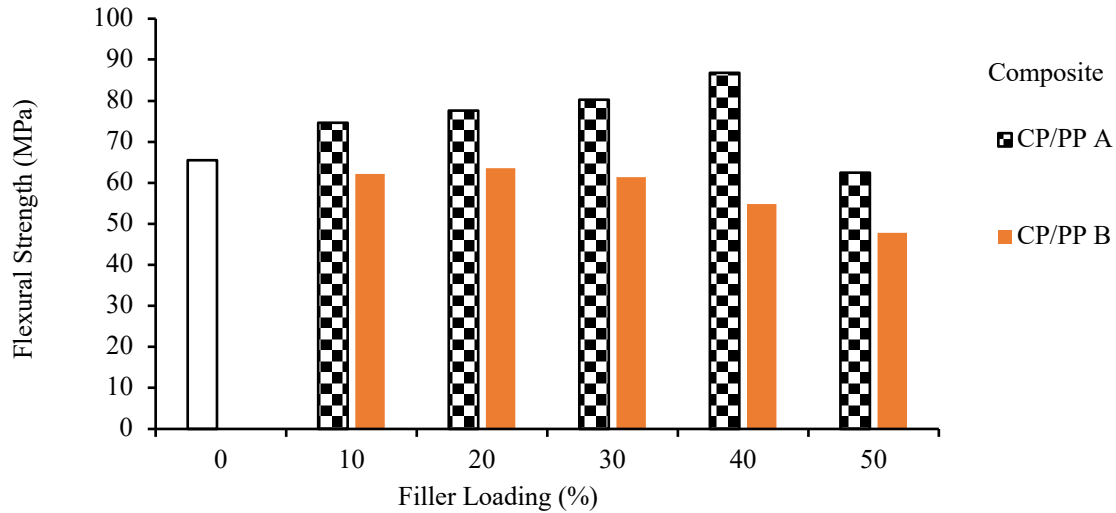


Fig. 3: Flexural strength of CP/PP composites A with coupling agent and B without coupling agent at varying filler loading

### 3.4 Impact strength

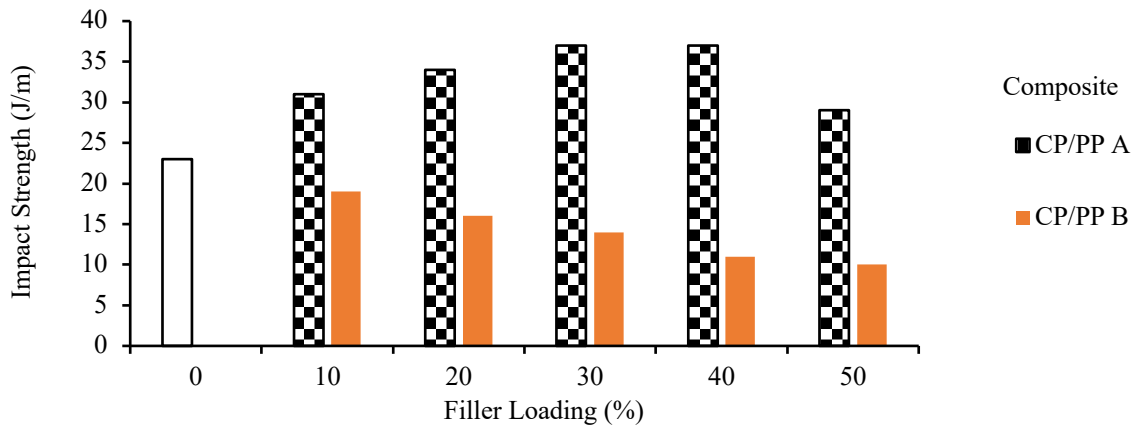


Fig. 4: Impact strength of CP/PP composites A with coupling agent and B without coupling agent at varying filler loading

Fig. 4 shows the result of impact strength increasing with increase in filler loading of the composite for CP/PP composites A from 23J/m of the neat polymer to 37J/m at 40% filler loading representing 37.84% increase. This might be attributed to improved interfacial interaction between the filler and matrix when coupling agent was introduced in the composite system, maleic anhydride has the property of covalent bonding on to another polymer

chain (Maldas and Kokta, 1991). Introduction of coupling agent into a composite system with dissimilar surface chemistry between the filler and matrix improves interfacial adhesion and consequently improves mechanical properties, increase in impact strength has been reported by several researchers due to the incorporation of coupling agent into a composite system (Shunmin, et al., 2017). In contrast with CP/PP composites A, in CP/PP composites B without coupling agent, impact strength was found to be decreasing with increased filler loading from 23J/m of the neat polymer to 10J/m (56.52% decrease) at 50% filler loading. This might be attributed to dissimilarity between the surface chemistry of filler and matrix bringing about reduction in the tenacity of the composites (Mohanty et al., 2002), decrease in impact strength may result from non-inclusion of coupling agent in the composite.

### 3.5 Hardness

Hardness which is a measure of material resistance to indentation of CP/PP composites A and B was found to be increasing with increased filler loading from 49.5Hv of the neat polymer to 91.3Hv (45.78% increase) and 78.27Hv (36.76%) increase for composites with and without coupling agent respectively. Hardness of a material is dependent on stiffness of that material, introduction of rigid fillers might have impart stiffness to PP thereby disrupting polymer chain mobility when force is applied consequently increasing hardness (Musa, et al., 2019). Since 40% filler loading gave the best and least mechanical property before reaching saturation, fractured surface of the tensile test specimen was used for SEM determination. SEM image of CP/PP composite A at 40% filler loading can be seen in Fig. 5, it can be observed that there is location of homogenous dispersion of the CP in the composite matrix with coupling agent. Coupling agents aid full embedment of reinforcing fillers in the matrix thereby preventing de-bonding and de-bonding leads to plastic deformation of composites (Baba et al., 2020c). Meanwhile, in Fig. 6, the SEM image of CP/PP composite B without coupling agent showed areas of agglomeration and uneven dispersion of the filler in the composite which might be the reason why there is reduction in the mechanical properties of composites without coupling agent. (Baba et al., 2020a).

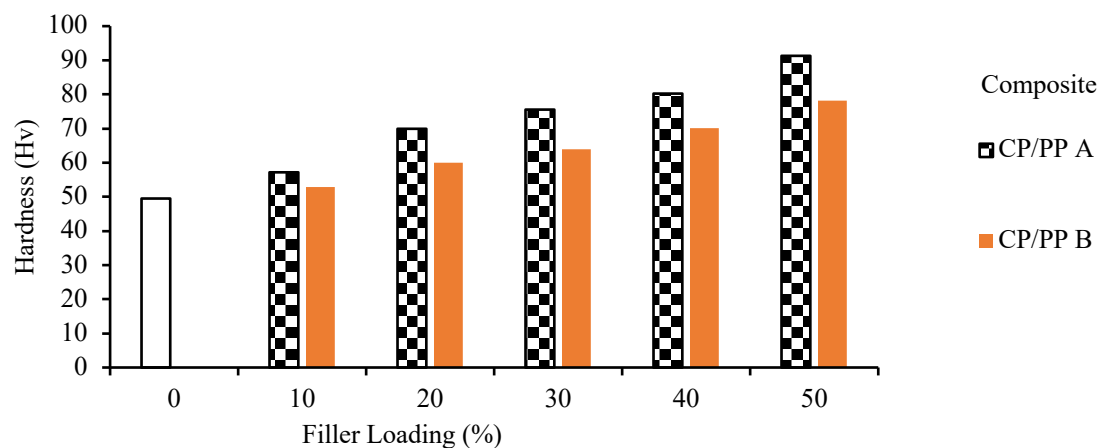


Fig. 5: Hardness of CP/PP composites A with coupling agent and B without coupling agent at varying filler loading



### 3.6 Scanning Electron Microscopy

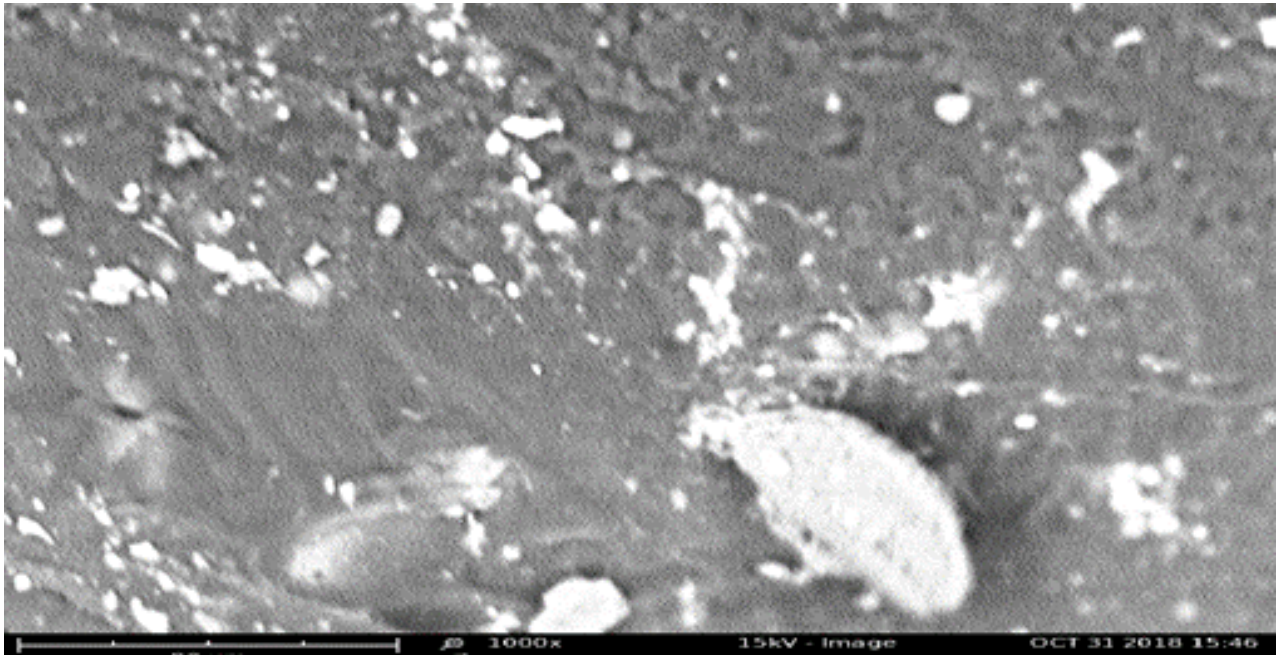


Fig. 5: SEM image of CP/PP composite A at 40% filler loading with coupling agent

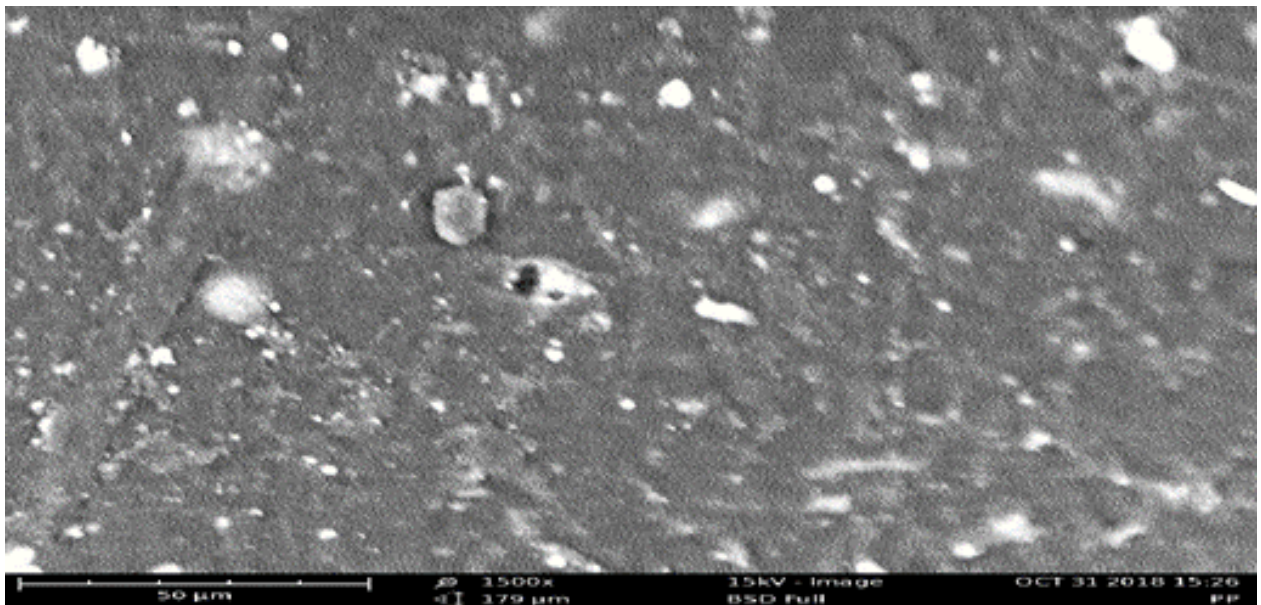


Fig. 6: SEM image of CP/PP composite B at 40% filler loading without coupling agent



#### 4. Conclusion

Studies of the static mechanical properties of CP/PP composites was conducted successfully and the results obtained showed general increasing trend of mechanical properties i.e. tensile strength, tensile modulus, flexural strength, impact strength and hardness for CP/PP composite A showed increasing trend from 0 to 40% filler loading which might be due to incorporation of maleic anhydride as coupling agent. Meanwhile, CP/PP composite B showed decreasing trend from 0 to 50% filler loading, but tensile modulus and hardness where increasing with increased filler loading. The SEM image of CP/PP composite A at 40% filler loading showed even distribution of fillers in the matrix which is a sign of proper bonding between the filler and matrix preventing debonding, while, CP/PP composite B showed uneven dispersion of fillers with areas of filler sprout. From the result of the mechanical properties obtained it can be deduced that the composites can be used for applications where low to moderate mechanical properties are required such as local utensils, buckets, plates, furnitures etc.

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