IMPACT OF MIXED AGRO WASTE AS HYBRID FILLERS ON THE MECHANICAL, MORPHOLOGICAL, WATER ABSORPTION AND BIODEGRADATION PROPERTIES OF HIGH DENSITY POLYETHYLENE COMPOSITES.

Ogudo, M.C. and Chris-Okafor, P. U.

Department of Pure and Industrial Chemistry,
Nnamdi Azikiwe University, Awka, Nigeria.
ogudomarcellinus@yahoo.com
pu.chrisokafor@unizik.edu.ng

Abstract

Agricultural waste is yet an untapped source of raw materials that can, in case of proper application enhance greatly the sustainability of polymers and their composites leading to eco-friendly and environmental degradation of plastics. The impact of the mixed agro wastes; rice husk and sugarcane bagasse on the mechanical, morphological, water imbibition and degradability properties of high density polyethylene was studied. The fillers were mixed in the ratio of 50:50. The mixed fillers were incorporated into the HDPE polymer resin at different proportions (0%, 5%, 10%, 15% and 20%), and the composites were fabricated via injection moulding technique. The mechanical properties; tensile strength, %elongation at break, compressive strength, shear modulus and hardness test of the composites were analysed according to ASTM standards. Also, the morphological properties were studied with a Scanning Electron Microscope. The result of the mechanical properties test showed a reduction in tensile strength and percentage elongation at break, and an increase in hardness, with the optimum hardness observed at 10wt% filler loading. The lowest tensile strength values were observed at 5wt% and 20wt%, the lowest value for the percentage elongation at break was also observed at 20wt%. An increase in compressive strength and shear modulus of the composites were also observed, 20wt% had the optimum compressive strength and shear modulus. The result of the morphological study showed that there is good adhesion and interfacial bonding between the filler and the polymer matrix as a result of good dispersion of the fillers in the polymer matrix. Biodegradation study showed a reduction in the mass of the composites after a 3-month burial period indicating that the composite is more environmentally benign. Result from the water absorption test indicated no increase in the mass of the composites after immersion in water. This is an indication that the composite can be utilized in wet environments.

Keywords: *Agro waste, HDPE, Mechanical properties, Morphological properties, Water absorption, Degradation.*

1.0 Introduction

As a result of increased activity in the modern agricultural sector, a large amount of waste is generated, posing severe environmental concern. Plastics' ever-increasing need and wide range of applications in man's daily existence cannot be overstated. Meanwhile, the scarcity of raw resources is raising worry, and agro waste might be considered as a good alternative source for producing value-added products like bio-composites in this context. Natural fibers, particularly agricultural waste fibers, require additional development as a long-term strategy for developing the enormous abundance of natural plant fiber that is currently underutilized. Environmentally friendly composite materials made from natural fillers and polymer materials are currently gaining popularity. (Sarki *et al.*, 2011).

Lignocellulose fillers are a scientific and technological advancement in the field of novel materials, emphasizing the necessity of utilising agricultural waste as a source of raw materials. Due to their features such as high specific strength, high modulus, recyclability, renewable origin, high volume application, easy workability, and low cost, lignocellulosic composites have received a lot of interest. (Kiziltas *et al.*, 2011; Navas *et al.*, 2015; Zaaba and Ismail, 2019). To actualize the scale up manufacturing of agro waste plastic composites by the development and synthesis of these filler fibers, multiple procedures that entail mixing of filler husks at varied filler loading per weight are necessary. (Yang *et al.,* 2006). With the exception of fibrous natural fillers, the loading of dispersed agricultural waste in the polymer matrix is currently receiving a lot of attention. As a result, lignocellulosic plant waste, such as shells, husks, and stems from agricultural crops, can be used not only as a source of biomass, but also as low-cost dispersion fillers for polymer composites.

Therefore, the aim of this study is to investigate the suitability of rice husk and sugarcane bagasse as fillers for the production of High Density Polyethylene composites.

2.0 MATERIALS AND METHODS

2.1 Materials

The polymeric material used in this study was HDPE (HMA014) Produced by ExxonMoil, USA of density 0.96 g/cm^3, and melt flow index (MFI) 4.0g/10 min. Rice husk was sourced locally from a rice mill in Abakaliki, Ebonyi State, while Sugarcane baggase were sourced locally from Boundary market in Ajegunle, Apapa, Lagos State, Nigeria. The agricultural waste materials were washed and sun dried for three days, after which they were pulverized using a grain mill machine M6FFC-270, and sieved locally and repeatedly to obtain a fine powder of 75µm size.

2.2 Preparation of Composites

The HDPE composites were prepared by weighing 190g, 180g, 170g and 160g of the virgin crystalline HDPE pellets and thoroughly mixing with blend of rice husk and sugarcane bagasse fillers in a 50:50 ratio. Weight of blend varied at 10g, 20g, 30g, and

40g which correspond to 5%, 10%, 15% and 20% filler loading respectively. The polymer and filler homogenous mixture was fed into the hopper of an injection molding machine TU150 200 gram with a circular shaped mould. The composites were shaped into standard dimensions for mechanical properties analysis.

2.3 Mechanical properties analysis of composites

2.3.1 Tensile strength

This is the capacity of the composites to withstand loads tending to elongate- In other words, it is measured by the maximum stress that a material can withstand while being stretched or pulled before breaking. The tensile strength of the composites was measured according to the American Standard Testing Method D-638-14, using the Hounsfield Monsanto Tensometer 8889. The test piece was measured to 160x19x3.2mm dimension.

2.3.2 Hardness test

Hardness is measured by the distance of indentation and recovery that occurs when an indentor is pressed into the surface under constant load and released. The surface hardness was calculated as Brinell Hardness Number in N/mm^2 measured according to the American Standard Testing Method E10, using the Hounsfield Monsanto Tensometer 8889. The test piece was measured to 20x20x3.2mm.

2.3.3 Shear strength

Shear strength is measured by the ability of the material to resist forces that cause its internal structure to slide against itself. The shear strength of the composites was measured according to the American Standard Testing Method D-732, using the Hounsfield Monsanto Tensometer 8889. The test piece was measured to 20x20mm dimension.

2.3.4 Compressive strength

Compressive strength is the capacity of the composites to withstand loads tending to reduce its size. It is simply the ability to resist compression.
The compressive strength of the composites was measured according to the American Standard Testing Method D-695, using the Hounsfield Monsanto Tensometer 8889. The test piece was measured to 40x40mm dimension.

2.4 Water absorption test

This test covers the method of determination of water absorption of the composites as determined by Standard ASTM D-570-98. The composites cut into 50x50mm dimension were dried and immersed in water for a period of 3 days. The moisture absorption by the composite was measured by the weight gain of the material at daily intervals. The percentage moisture absorption capacity is expressed as the ratio of increase in mass of the composite to the initial mass.

2.5 Biodegradation test

This test is done to determine the extent the composites will degrade in the environment. This was determined using soil burial degradation test. HDPE composites were buried in a soil obtained from an automobile mechanic workshop for degradation. Degradation was measured from the mass reduction of composites buried. Composites were cut into 50x50mm dimension, weighed, and buried into the soil at 15cm depth for a three-month period. The composites were weighed at interval of 30 days during the test period to determine the extent of degradation.

2.6 Surface morphological analysis

Morphological analysis was conducted using a Scanning Electron Microscope model PRO:X: 800-07334 Phenom world MVE01570775 to study the surface distribution between the polymer matrix and the filler.

3.0 RESULT AND DISCUSSION

3.1 Mechanical properties

3.1.1 Tensile strength

The effect of filler loading on the tensile strength of the composite is shown in Figure 1.

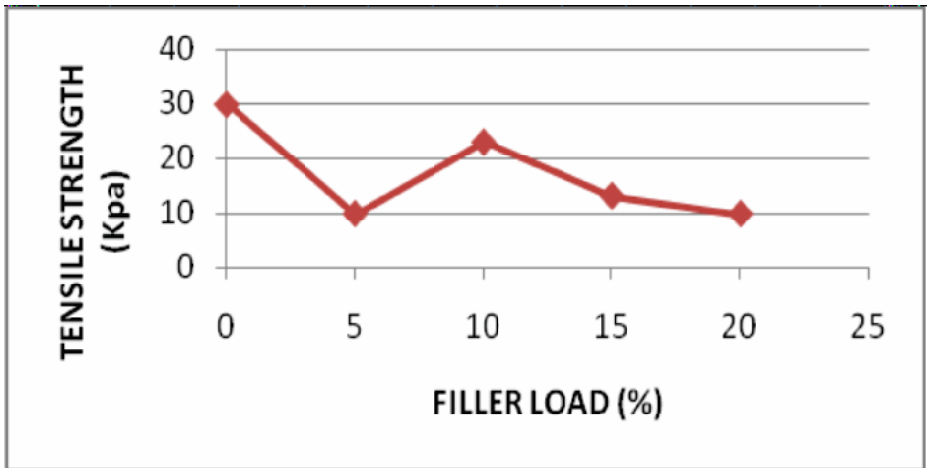

Figure 1: Effect of filler loading on the tensile strength of HDPE composites

From Figure 1 it is clearly seen that when the filler load increases, the composites' tensile strength decreases, with a 57% increase at 10wt% loading. Salmah *et al.,* (2005) found a similar result, with the loss in tensile strength due to poor filler-matrix adhesion, uneven filler dispersion in the polmer matrix, and filler particle agglomeration. This result is similar to report from Chris-Okafor *et al.,* (2018) and Onuegbu (2012).

3.1.2 %Elongation at break

This is the measure of ductility of the composite material. The effect of filler loading on the percentage elongation at break of the composite is shown in Figure 2.

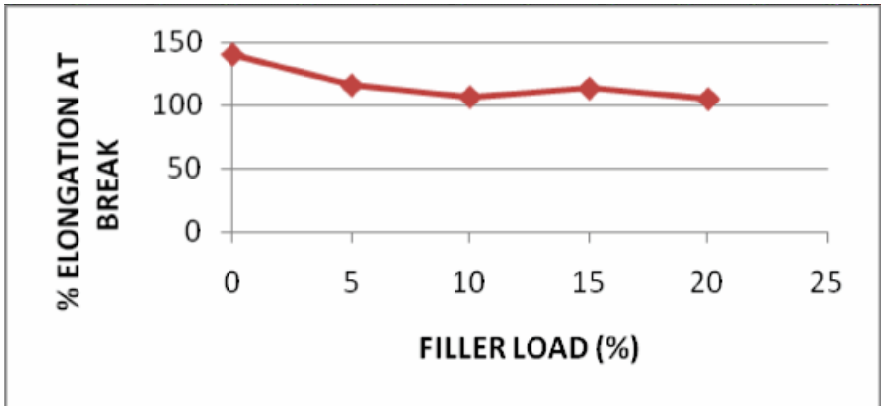

Figure 2: Effect of filler loading on %elongation at break

As seen in figure 2, the percentage elongation at break decreased with increasing filler loading. Increased filler loading in the HDPE matrix resulted in the stiffening and hardening of the composite. This lowered its resilience and toughness, leading to lower elongation at the break. (Jacob *et al.*, 2004)

With increasing filler loading, the elongation at break decreases, indicating that the filler is unable to support stress transfer from the polymer filler to the matrix. The material's ductility was reduced as the hardness of the substance increased. As a result, ductility reduces with increasing filler content. (Siti *et al.,* 2007)

3.1.3 Hardness

The hardness of the composites is its resistance to penetration and deformation. The effect of filler loading on the hardness of the composite is shown in Figure 3.

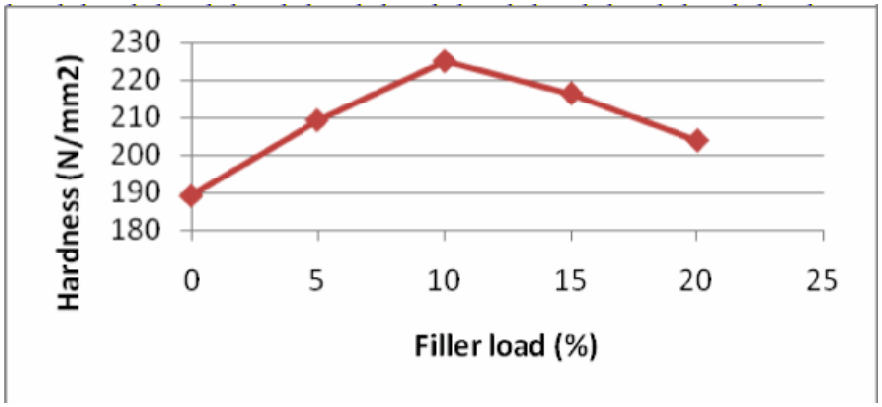

Figure 3: Effect of filler loading on the hardness of HDPE composites.

As seen in Figure 3, the hardness increased steadily as filler loading increased from 5 to 10wt% but had a 9% decrease at 20wt%. The optimum hardness was observed at 10wt%. This is evident in the composites percentage elongation at break (10wt% has the least value).

The increase in hardness may be attributed to the strengthening effect of the filler incorporated into the polymer matrix. Fillers are usually added to polymeric materials to improve their rigidity and strength. This is attributed to the nature of the filler. The higher the percentage of the fillers incorporated, the harder the material, and the more rigid it becomes. This is evident even from the results obtained for elongation at break, a direct opposite of hardness, which was noticed to decrease with increase in filler content.

3.1.4 Compressive strength

This is the resistance of the composite material to breaking under compression. The effect of filler loading on the compressive strength of the composite is shown in Figure 4.

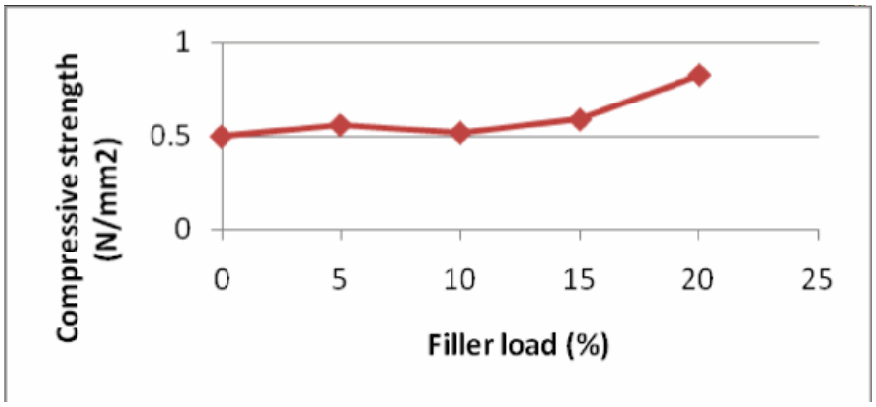

Figure 4: Effect of filler loading on the compressive strength of HDPE composites

Figure 4 shows a corresponding increase in compressive strength with increasing filler loading. This suggests that the composites would regain its original form after a heavy weight is lifted from it. This enhanced property is due to the reinforcing nature of the hybrid fillers. This result is similar to those of Onuegbu *et al.,* (2010), Ruijun *et al.,* (2013), Chris-Okafor *et al.,* (2019).

3.1.5 Shear modulus

The shear modulus is the modulus of rigidity of the composites. It is the ratio of the shear strength and the shear strain. The effect of filler loading on the Shear modulus of the composite is shown in Figure 5. It is clearly seen in Figure 5 that the shear modulus increased with an increase in filler loading. This observation is as a result of the reinforcing nature of the fillers. It increased the rigidity of the composites.

3.2 Morphological properties

The morphological study of the composites was carried out using the Scanning Electron Microscope (SEM). This study is done to observe the distribution of the filler in the polymer matrix. Figure 6(a), 6(b), 6(c) and 6(d) is the micrograph of RHSB-HDPE 0wt%, 5wt%, 10wt%, and 15wt%, respectively. The variations in mechanical properties of the composites are related to the SEM micrograph.

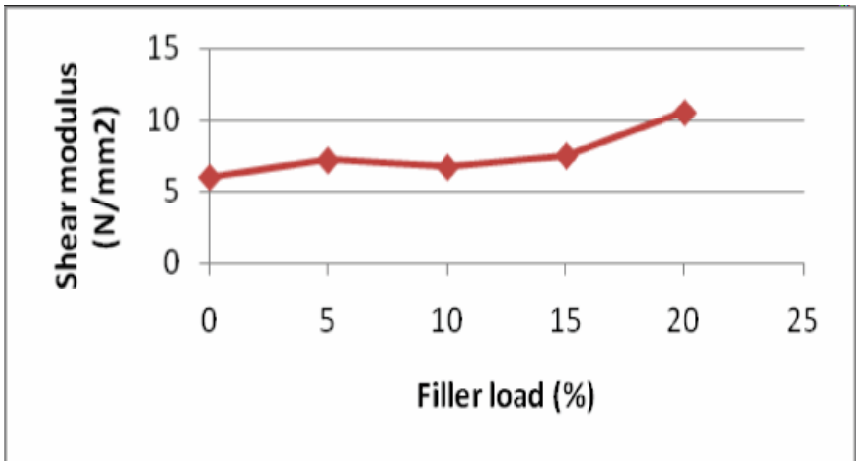

Figure 5: Effect of filler loading on the Shear modulus of HDPE composites

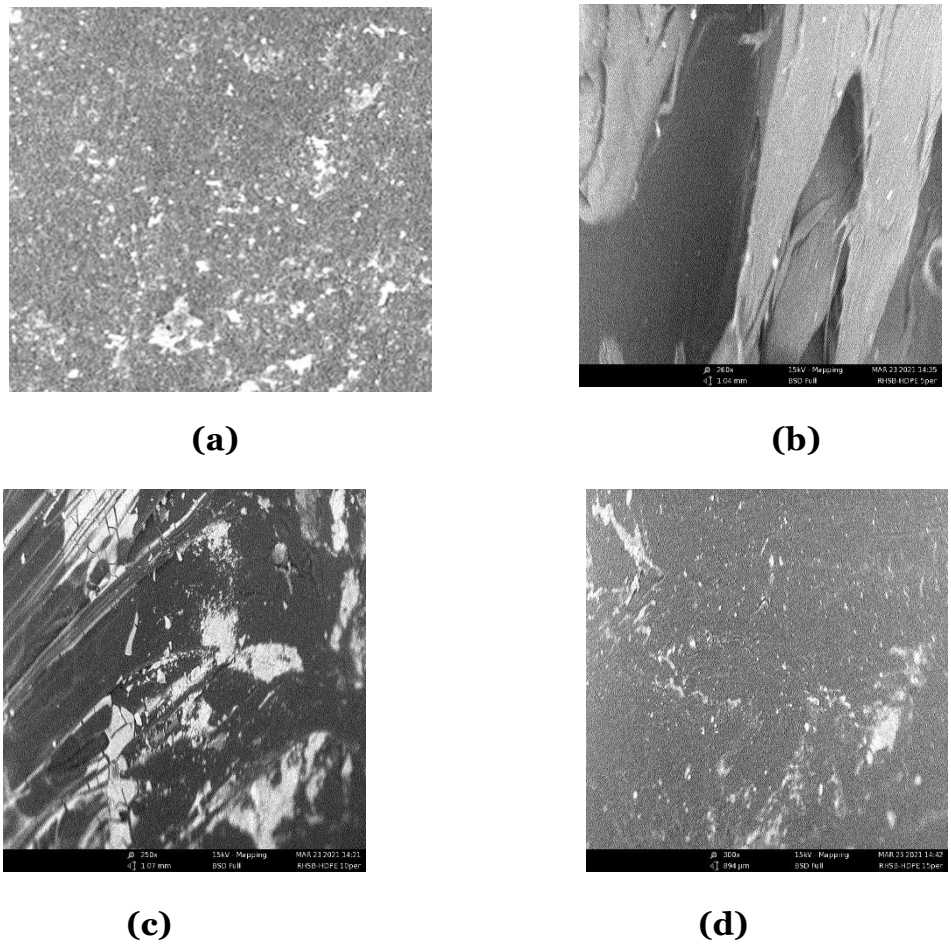

Figure 6: SEM micrograph of RHSB-HDPE (a) 0wt% (b) 5wt% (c) 10wt% (d) 15wt%

From Figure 6 it is clearly observed that the filler dispersion improved with increasing filler content. The high value for hardness, compressive strength and shear modulus exhibited by RHSB-HDPE 15wt% could be as a result of the dispersion of the filler in the polymer matrix.

The SEM micrographs for fig. 6(b), 6(c) and 6(d) shows that there are no clear gaps between RHSB hybrid fillers and HDPE matrix, indicating good interface bonding. The fillers are well attached to the matrix which is as a result of good interfacial interaction between the filler and the matrix.

3.3 Water absorption

The water absorption test results of the composites showed no increase to the final weight of the composites after complete immersion in water for a period of 3 days. This is an indication that the composites is incapable of absorbing water and can be employed in water storage, bathroom interior, and other uses in wet environments.

3.4 Biodegradability test

Figure 7 reports the biodegradation test results for the hybrid blends of RHSB-HDPE composite.

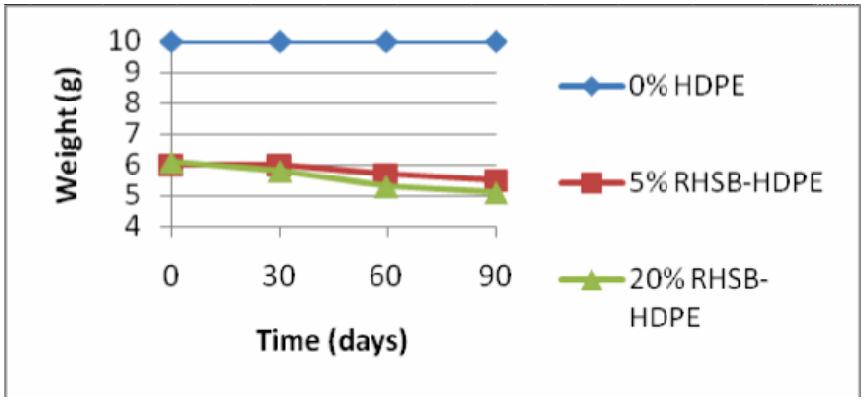

Figure 7: Effect of filler loading on the biodegradability of HDPE composites.

From figure 7, as anticipated, there was no reduction in weight for 0% HDPE during the 90 days test period. For 5wt% RHSB-HDPE composites, weight loss was noticed after 30 days during test period, and there was a general reduction in weight with as the burial time increased. It is also observed from the figure that increased filler content led to higher degree of degradation of the composites. The 5wt% and 20wt% filler loading gave an 8% and 16% weight reduction after 90 days, respectively. This observation corroborates the research findings of Amir *et al.,* (2013) and Oldak *et al.,* (2006). These authors discovered that biodegradability of composites is well pronounced and enhanced if the composite contains at least 30% of the natural fillers.

Conclusion

Rice husk and sugarcane bagasse were successfully incorporated as hybrid fillers into high density polyethylene (HDPE) polymer matrix to make polymer composites. The incorporation of this filler improved the hardness, compressive strength, and shear modulus of the composite. However, the tensile strength and percentage elongation at break both decreased. The foregoing findings, as well as those from other studies, show that this is a common occurrence with lignocellulosic fillers.

The mechanical properties of the composites formed were found to be influenced by the interaction of the polymer matrix with the filler, as well as the particle size and distribution of the fillers within the matrix. During the test period, the composites were found to resist water absorption. This indicates that the composite is well suited for water storage, drainage pipes, and other wet-environment applications.

The composites were also found to degrade which was evident in its weight reduction over a three-month test period. More filler content in the composites would greatly enhance biodegradability of the composites. However, this may deteriorate the mechanical properties of the composites. As a result of the findings, it is advised that agro wastes such as rice husk and sugarcane bagasse, which are inexpensive, readily available, and environmentally benign, be utilized as fillers in the production of plastics since they are more cost-effective, and feasible. When disposed of, the plastics produced would easily disintegrate.

References

Amir, N., Ashori, A., Kazemi T., Ali, (2014). Characterization and biodegradability of polypropylene composites using agricultural residues and waste fish. *Composites Part B: Engineering,* 56(2014):279–283.

Chris-Okafor, P.U., Okonkwo, C., Ohaeke, M., (2018). Reinforcement of High Density Polyethylene with Snail Shell Powder, *American Journal of Polymer Science*, 8(1): 17-21.

Chris-Okafor, P.U., Arinze R. U., Iloamaeke, I.M., Izundu, B. C., (2019). Investigation of the Mechanical Properties of Flexible Polyether Foam Filled with Eggshell and Groundnut Husk Powder. *Organic Polymer Material Research*, 1(2): 1-6.

Jacob, M., Thomas, S. & Varughese, k.T. (2004). Mechanical properties of sisal oil palm hybrid fiber reinforced natural rubber composites. *Composite, Science and Technology*, 64: 55–965.

Kiziltas, A., Gardner, D.J., Han, Y., Yang, H.S., (2011). Thermal properties of microcrystalline cellulose-filled PET –PTT blend polymer composites. *Journal of Thermal Analysis and Calorimetry,* 103(1):163-170.

Navas, C.S., Reboredo, M.M., Granados, D.L., (2015). Comparative study of agro industrial wastes for their use in polymer matrix composites. *Procedia Materials Science*, 8:778-785.

Oldak, D., Kaczmarek, H., Buffeteau, T., Sourisseau, C., (2006). Photo and bio-degradation processes in polyethylene, cellulose and their blends studied by ATR-FTIR and Raman spectroscopies. *Journal of Material Science;* 40(16):4189–4198.

Onuegbu, T.U., Obianuko, N., Mbachu, G.U., Iloamaeke, I.M.. Effects of Animal Waste (Goat Femur) as a Filler in Flexible Polyether Foam, Journal of Basic Physical Research, 2010, 1(1): 5-8.

Onuegbu, T.U., (2012) A Study On the Effects of Coconut and Palm Kernel Shells On Density, Porosity Index and Tensile Properties of Flexible Polyether Foam,ARPN Journal of science and Technology, 2012, 2(6): 529532.

Ruijun, G., Mohini, M.S, Samir, K.K.. A Feasibility Study of Polyurethane Composite Foam with Added Hardwood Pulp, Industrial Crops and Products, Checkmate press, New York, 2013, 42(2): 73-279.

Salmah, H. Ismail, A. & Bakar A., (2005). Dynamic vulcanization of paper sludge filled polypropylene (PP)/ethylene propylene diene terpolymer (EPDM). *Malaysian Journal of Microscopy*, 2: 15–22.

Sarki, J., Hassan, S.B., Aigbodion, V.S., Oghenevwet a, J.E., (2011). Potential of using coconut shell particle fillers in ecocomposite materials. *Journal of Alloys and Compounds*, 509(3):2381-2385.

Siti Shuhadah, M., Supri, A.G. & Kamaruddin, H. (2007). Mechanical properties of egg-shell powder (ESP) filled low density polyethylene composites. Proceedings of VIIth National Symposium on Polymeric Materials, UniKL, Kuala Lumpur, 27–28 Nov 2007.

Yang, H.S., Kim, H.G., Son, J., Lee, B.J., and Twang, T.S., (2006) Rice Husk Flour Filled with Polypropylene Composites, Mechanical and Morphological Studies, *Journal of Composite Structure*. 63(3-4):305-312.

Zaaba, N.F., and Ismail, H., (2019). Thermoplastic/natural filler composites: A short review. *Journal of Physical Science*, 30(1):81-99.