Investigation of Mechanical Properties of Luffa Cylindrica Particulates Reinforced Polylactic Acid Composite

MisbahuAhmed^{1,*} and Ibrahim Abdullahi²

¹National Centre for Petroleum Research and Development, Energy Commission of Nigeria, Abubakar Tafawa Balewa University Yelwa Campus Bauchi, Nigeria

> ²Faculty of Mechanical Engineering Bayero University Kano New site, Gwarzo Road Kano, Nigeria

Received: 17May2020; Revised: 20October2020; Accepted: 1December 2020; Published: 15 December 2020

ABSTRACT

The motive of this study is to lessen the dependence on non-degradable plastic by developing alternative material; luffa cylindrica particulates (LCP) reinforced polylactic acid (PLA) composites using the available plastic processing machineries. For that reason, this study focuses on the development of PLA/LCP biocomposite using compression moldingand also investigated the effect of varying the LCP weight fraction and surface treatment on the density and the mechanical properties of the developed composites. The LCP were treated with 5 wt% NaOH solution. Two batches of composites were produced using treated LCP and untreated LCP. Composites were fabricated with 10, 20, 30, 40 and 50 wt% LCP for both the treated and untreated LCP using compression molding. Characterization of the composite was made based on the evaluation of its density, tensile properties, flexural properties, impact strength and hardness. The results showed that the densities of the treated composites were slightly lower than those of the untreated. The composite with treated LCP showed desirable properties at 10, 30 and 40 wt% LCP while the composites with the untreated LCP gave better results at 20 wt% LCP. Though these properties compared favorably with some reported properties for the natural fiber reinforced polymer composites, the use of other types of surface treatment for the LCP should be studied and also the effect of varying the wt% of NaOH solution be further investigated.

Keywords: Luffa cylindrica particulates (LCP), polylactic acid (PLA), surface treatment, mechanical properties, compression molding

1.0 INTRODUCTION

Currently, plastics have increasingly become the choice materials for diverse applications owing to their long life and attractive properties but their outstanding growth in applications, makes them one of the rapidly growing sectors of the waste stream. Majority of plastic by-products are built from petroleum-based synthetic polymers.

^{*}Corresponding email: misbahuahmed@gmail.com

Synthetic polymers have displaced metals, glasses, ceramics and wood in many products, especially in the area of packaging. The commodity plastics: polyethylene (PE), poly (propylene) (PP), polystyrene (PS) and poly (vinyl chloride) (PVC) in different forms such as films, flexible bags and rigid containers have revolutionized the packaging industry. However, once disposed of, they recur in the environment. Their major disadvantage is the problem of acceptable removal of their 'waste' after useful life. They are difficult to recycle and they do not degrade in landfill site or a composite like environment [1]. These polymers therefore seem inappropriate for applications involving plastics that have short life span in terms of usage, i.e., those plastics that are used only once and then disposed. Ecological concerns have prompted a renewed enthusiasm in materials that are more natural and compostable and as a result, biodegradability and environmental safety are issues that are now of paramount importance [2].

On the basis of these, researches to alleviate pollution and litter problems have focused on developing bio-based composites consisting of biopolymers and natural fibers or agricultural residues. These products are expected to reduce our dependency on the depleting fossil fuels and decrease CO_2 emission into the atmosphere.

Natural fibers can be cultivated so that its availability is sustainable. *Luffa cylindrica*(LC) is a plant that is in abundance in most states of northern Nigeria but its use is only limited to household and medicinal purposes with majority of the fruit left to waste on farm lands. Therefore, there is the need to create awareness on the possibility of producing a completely green composite using LC as reinforcement in a biodegradable polymer matrix and thus encouraging the use of these biodegradable plastics for commodity products to help preserve the environment.

Natural fibers are degradable, their use as a substitute for their non-degradable counter parts as fillers can be beneficial to the environment. However, using natural fibers as reinforcing fillers in a composite faces several underlying factors which are related to characteristics of most lignocellulosic materials such as their irregular dimensions, stiffness, susceptibility to heat and hydrophilicity all these present a major concern which needs to be addressed in order to meet the requirements of the mechanical properties in a developed composite[3].

Properties of composite with natural fibers are strongly influenced by bond strength between matrix and fibers; these can be improved with a variety of chemical treatments on the fiber such as using sodium hydroxide (NaOH), silanes, permanganate, KMnO₄ and peroxides [4].Patra et al.(2019) reported that the tensile strength and flexural strength of polylactic acid (PLA) reinforced with irradiated LC fiber increases by 79.87% and 177%, respectively[5]. Parida et al. (2015) investigated the influence of cellulose nano fibers extracted from the fruit of LC on the tensile, flexural and impact properties of composite materials using PLA processed by micro compounding and injection molding and reported that at 2 wt% LC fiber, the flexural, tensile and impact strength of the composites increases but decreases at 5wt% and 10 wt%[6].Kakar et al. (2015) fabricated biopolymer composites using PLA and heat-treated LC fiber as reinforcement processed by hot press compression molding method[7]. The composites with 15 wt% of heattreated LC fiber showed highest tensile strength while 20 wt% untreated LC fiber showed lowest tensile strength. The range of tensile strength of composites reported by them lies between 7.18 MPa and 40.20 MPa. Rice hulls filled PLA biocomposites were prepared by compression molding and reported by Srebrenkaska et al. (2014) [8]. The maximum flexural strength reported was 28.8 MPa for 30 wt% reinforcement of rice hulls which showed 3.5% decrease from neat PLA. The maximum flexural modulus was found to be 3.24 GPa at 30 wt% of fiber content which was 1.25% higher than neat PLA. Avella et al. (2009) studied the suitability of using wood fibers (WF) as reinforcements in PLA matrix and reported that the flexural strength of WF/PLA composites improved from about 100MPa for the neat PLA to about 115MPa for PLA based composites at 20-40 wt% of WF and the flexural modulus improved by about 200% at 40 wt% WF[9].

Some natural fibers such as hemp, jute, sisal, pineapple leaf, coir, flax, bamboo and palm oil has been used as reinforcement in composite development [10]. Though, the use of LCas reinforcement in the PLA matrix is reported, it is mostly in the form of short or long fiber but not as particulates. In view of this, the study is aimed at developing a biodegradable composite from LC particulates and poly lactic acid using compression molding and also to investigated the effect of varying the LCP weight fraction and the effect of surface treatment on the density and the mechanical properties of the developed composites.

2.0 EXPERIMENTAL METHOD

2.1 Materials

The materials used in the study were based on the followings: *Luffa cylindrica* sponge, PLA grade 3051D from Nature Works (USA), distilled water

2.2 Methods2.2.1 Preparation of the particulates

and NaOH (purified pellets 500g).

The LC sponge used in this study was collected locally from a farm in Bauchi state Nigeria. The outer layer (bark) and seeds of *luffa* fruit were carefully removed after which the *luffa* sponge was washed thoroughly with tap water and sun dried in the open (average temperature; 36.4°C and relative humidity; 40%) for 72 h. The dried *luffa* fiber was subjected to size reduction by grinding using local milling machine and sieved with 1000 µm aperture sieve, mesh no.18 in accordance with (ASTM E11-01). The LCparticulates (LCP) were soaked in hot water for 3 h, washed, air dried and subsequently oven dried in a hot air oven (Genlab Limited Thermal Engineers, Model No: *HAS/100/SS/DIG*) at 110°C for 3 h.

2.2.2 Chemical treatment of the particulates

The untreated fiber was divided into two halves, one half remains as untreated and the other half was treated by soaking it in 5% wt solution of NaOH maintained at room temperature for 4 h after which it was washed with distilled water until all the NaOH is eliminated [11]. After washing, the particulates were sprayed out on a mat and air dried in the open for 2 days and subsequently oven dried in a hot air oven at 110°C for 3 hours.

2.2.3 Preparation of composites and sample testing

A two-roll milling machine (Reliable rubber and Machinery 5183) was used for mixing the constituents. It was first set to preheat at a temperature of 180°C for an hour with the speed of the front and rear rollers set at 40 and 30 rpm, respectively. Blending was carried out by introducing the PLA directly onto the rollers and allowing it to melt for 4 mins and the addition of the varying amounts of the LCP (treated and untreated) fillers from 10-50 % wt at an increment of 10 % wt was done manually. Mixing was done between the rollers and manual turning of the mixture was done to achieve homogeneity and good dispersion within the matrix. The final mixture was transferred into a mild steel mold of dimensions 150 x 130 x 4 mm and closed. The closed mold was compressed using a hydraulic press (Carver Inc. model 12000) subjected to a pressure of 5 bar maintained for 8 mins at a temperature of 150°C. The mold was allowed to cool for 1 h at room temperature and the composite in the form of a plate was retrieved. Composites with the variation of fiber weight fraction 10, 20, 30, 40 and 50% were made for both the treated and untreated LCP.

The density of the developed composite was determined in accordance with ASTM D729. Specimen of size (100 x 10 x 4) mm was prepared and the mass of the sample was

determined using digital weighing balance (average of five weights), the corresponding volume was calculated using standard formula for volume of solid and the density was computed using the equation, $\rho_c = \frac{M_c}{V_c}$ (where ρ_c : density of composite, M_c :mass of composite and V_c :volume of composite).

Tensile testing has been evaluated using *Cusson's* Universal Materials testing machine.(model no:*P5030*) according to ASTM D638. The strain rate used was 50 mm/min.Three-point flexural testing was carried out on the same machine with spanlength of 50mm and the compression speed of 5 mm/min according to ASTM D790. Notched *Charpy* impact test has been conducted according to ASTM D256 using *Charpy* Impact Testing Machine, capacity 15J and 25J, serial no:*412-0715269C*. The hardness test was performed in accordance to ASTM D2240 using *Indente* Universal hardness testing machine. For all the mechanical testing, an average has been taken on five specimens.

3.0 RESULTS AND DISCUSSIONS

3.1 Density

The density values of the PLA and the PLA/LCP composites with variation of LCP weight fractions are presented graphically in Figure 1.

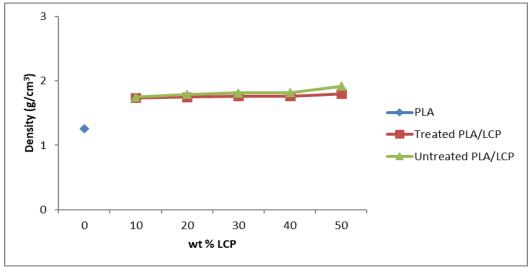
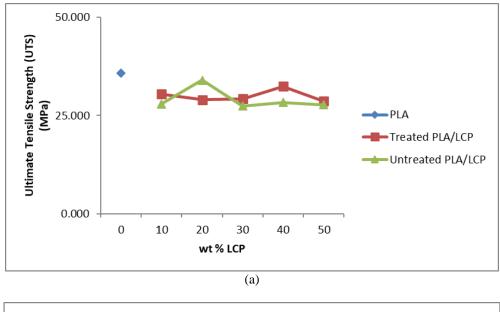


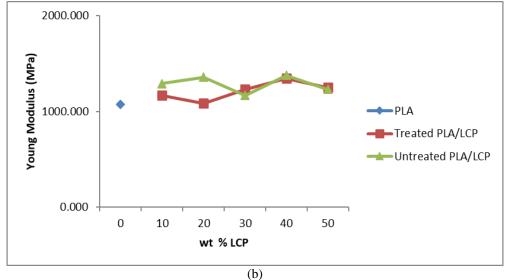
Figure 1: Density of PLA and PLA/LCP composites against filler content

The results showed that the neat PLA matrix has a density value of 1.259 g/cm³. The reported density values of PLA from other studies were 1.24 g/cm³ [12] and 1.25 g/cm³ [13]. The density values obtained for PLA and its composites from this study showed that they lied within the range of 1.2 and 1.9 g/cm³. Surface treatment led to a slight decrease in density values of the treated composites and this could be attributed to the removal of pectin, lignin and hemicelluloses from the filler. The removal of these constituents improves adhesion of the filler with the matrix [14]. Fillers generally have low density which makes it possible to produce composite of low weights at low cost [15] and low weights are among the advantages of composite materials.

3.2 Tensile Properties

The tensile properties of the PLA and the PLA/LCP composites with variation of LCP weight fractions are presented graphically in Figures 2(a)-(c).





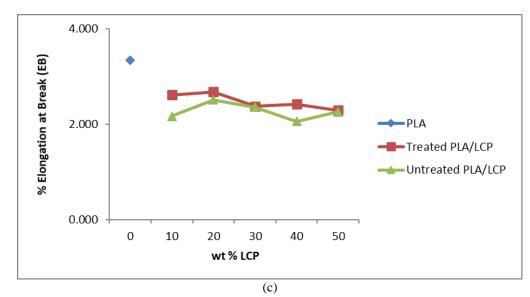


Figure 2: (a) Ultimate tensile strength (UTS) (b) tensile modulus (c)percentage(%) elongation at break of PLA and PLA/LCP composites against filler content

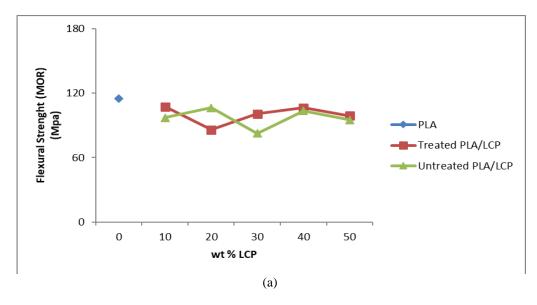
From the tensile strength results in Figure 2(a), a general decrease in tensile strength with filler loading was observed for the treated and untreated composites as compared to the neat PLA. The reason for the general decrease of tensile strength with filler loading is attributed to the irregular shape exhibited by the fillers which may not be able to maximize the interfacial surface area and are rather poor in their ability to support the stress transfer from the matrix leading to weak bond between the filler and matrix and this subsequently leads to reduction in tensile strength. The result also indicated that the surface treated composites have better UTS; this is because the alkali treatment resulted in an improvement in the interfacial bonding by giving rise to additional sites of mechanical interlocking, thereby promoting resin-filler interpenetration at the interface and also helps to improve the dispersion of fillers in the matrix, thereby reducing the filler agglomeration. This observation is in agreement with studies of Finkenstadt *et al.* (2007a) [16], Clarizio and Tatara (2012) [17], Ahmad *et al.* (2007) [18], Ewulonu and Igwe (2011) [19], Ibrahim *et al.*(2012) [20], Eze *et al.*(2013) [21] and Koutsomitopoulou *et al.*(2014) [22].

As seen in Figure 2(b), an increase in tensile modulus with LCP loading was observed. The tensile modulus of the composites with or without surface treatment is higher than that of the neat PLA matrix. Also, the tensile modulus of the untreated composites in most cases is higher in comparison to the treated ones thus, the treatment did not improve the tensile modulus. The general increase in tensile modulus with filler loading is commonly attributed to the inherent rigidity of fillers which exhibits higher stiffness than the polymer matrix. This rigidity tends to restrict polymer chain mobility which results in stiffening effect in polymer/filler composites, thereby increasing the rigidity of the composites. It could be inferred that the increase in tensile modulus of the composites with LCP loading is an indication that lignocellulosic fillers have the ability to impart greater stiffness to the matrix and since the increase in filler content resulted in the corresponding increase in the tensile modulus; it means that the tensile modulus is dependent on the filler content rather than the matrix [18].

Figure 2(c) presents the % elongation at break of PLA and PLA/LCP composites against the filler content. A general decrease in % elongation with increase in the LCP loading was observed for the treated and untreated composites as compared to the neat PLA. The reduction is attributed to the fact that fillers cause polymer matrices to lose their elastic properties due to stiffening effect which in turn leads to the restriction of the polymer chain mobility [18]. Also, with the increase in filler content, the matrix reduces in quantity which consequently decreases the effect of the matrix as compared to that of the filler and this can lead to an increase in modulus of the composites but a reduction of EL.

3.3 Flexural Properties

The flexural properties of the PLA and the PLA/LCP composites with variation of LCP weight fractions are presented graphically in Figures 3(a) and (b).



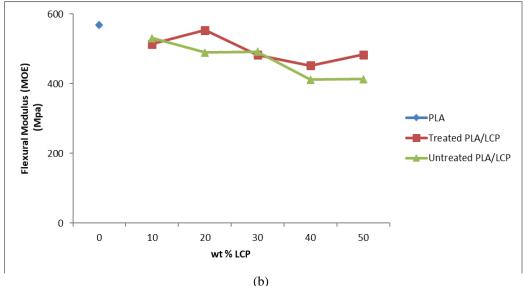


Figure 3: (a) Flexural strength (b) flexural modulus of PLA and PLA/LCP composites against filler content

From the results as shown in Figure 3(a), the flexural strength of the PLA composites showed a general decrease with LCP loading as compared to the neat PLA. This is in agreement with the observations of Yussuf *et al*(2010) [23]. This decrease could be attributed to the inability of the fillers, which are irregular in shape, to support stresses transferred from the polymer matrix as observed from the results, surface treated LCP composites had higher flexural strength than the untreated ones [24].

The results of the flexural modulus are as presented in Figure 3(b) which indicated that the flexural modulus of both the treated and untreated composites decreases with an increase in the LCP loading. Subramonian *et al.*(2016) while working on the effect of fiber loading on the mechanical properties of bagasse fiber–reinforced polypropylene composites reported a decrease in the flexural modulus of the composites with an increase in fiber loading but noted however, even with the decrease, the composites still have higher flexural modulus than the neat PP matrix[25].

In the current study, the flexural modulus also decreases with increase in fiber loading but, the neat PLA matrix has higher values of flexural modulus than the composites this could be as a result of poor surface adhesion between the matrix and the fiber.

3.4 Impact Strength

The impact strength results are presented in Figure 4.

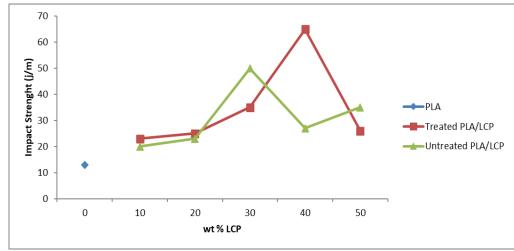


Figure 4: Charpy-V impact strength of PLA and PLA/LCP composites against filler content

A general increase in impact strengths with filler loading was observed for the treated and untreated composites as compared to the neat PLA matrix. This observation suggests that the LCP added to the PLA matrix acts like a solid "additive" which stiffened the flexibility of the polymer and improves its ability to absorb and dissipate energy, thereby enabling the composite to possess high impact energy to fracture. This is in agreement with the observations of Nwanonenyi *et al.*(2013) [26].

3.5 Hardness Test

The results for the hardness of the produced composites are presented in Figure 5.

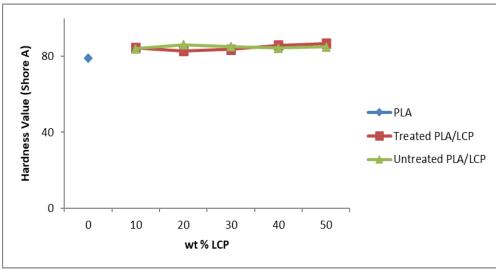


Figure 5: Shore A hardness values of PLA and PLA/LCP composites against filler content

The results showed a general increase in hardness with LCP loading for the composites as compared to the neat PLA. The increase in hardness with filler loading could be as a result of a more compact or rigid structure on the surface of the composite which leads to generation of greater resistance to penetration and consequently, higher

hardness values are obtained for the composites with filler loading [27]. Imoisili *et al.*(2012) also made a similar observation[28].

4.0 CONCLUSION

In the present study, *luffa cylindrica* particulates (LCP) reinforced PLA composites have been successfully fabricated using compression molding. The effect of LCP weight fraction and surface treatment on the density and mechanical properties of the composites were studied and the following conclusions were drawn from the study:

- The density of the composites increases with an increase in LCP wt% with the treated composites having lower densities than the untreated.
- Tensile strength, % elongation at break, flexural strength, flexural modulus all decreased with an increase in LCP wt% while the tensile modulus, hardness and impact strength increases as the LCP wt% is increased.
- Composites properties were highest at 20 wt% untreated LCP loading while composites with the treated LCP showed desirable properties at 10, 30 and 40 wt% LCP. 40 wt% treated composites exhibited most of the better result and this is an indication that the surface treatment has indeed improved some of the mechanical properties of the composites.

Though, the results compared favorably with other reported works, it is recommended that the use of other types of surface treatment for the LCP should be studied and also the effect of varying the wt% of NaOH solution be further investigated.

REFERENCES

- Umar S., 2015. Development and Characterization of Polylactic acid (PLA)/ Guinea corn (Sorghum Bicolor) Husk Particulate (GHP) Composites, Doctor of Philosophy Thesis. Department of Metallurgical and Material Science, Ahmadu Bello University (ABU) Zaria, Kaduna State, Nigeria.
- 2. Ramamoorthy S.K., Skrifvars M. and Persson A., 2015. A Review of Natural Fibers Used in Biocomposites: Plant, Animal and Regenerated Cellulose Fibers, *Polymer Reviews*, 55(1): 107-162.
- 3. Wayan S., Kade S. and Krissanti A., 2014. Mechanical Properties of Rice Husk Fiber Reinforced Polyester Composites, *International Journal of Materials, Mechanics and Manufacturing*, 2(2): 165-168.
- 4. Wambua P., Ivens J. and Verpoest I., 2003. Natural Fibres: Can They Replace Glass in Fibre Reinforced Plastics? *Composite Science and Technology*, 63: 1259 1264.
- 5. Patra S., Mohanta K.L and Parida C., 2019. Mechanical Properties of Bio-fiber Composites Reinforced with *Luffa Cylindrica* Irradiated by Electron Beam, *International Journal of Modern Physics B*, 33(26): 195-305.
- 6. Parida C., Sarat K.D., Pinaki C., 2015. Mechanical Properties of Injection Molded Poly(lactic) Acid—*Luffa* Fiber Composites, *Soft Nanoscience Letters*, 5: 65–72.
- 7. Kakar A., Jayamani E., Heng S.K., Bakri M.K.B. and Hamdan S., 2015 Optimization of Hot Press Compression Molding and Fabrication of Poly Lactic Acid (PLA) Luffa Biocomposites for Biomedical Applications, *Australian Journal of Basic and Applied Sciences*, 9:105-112.
- 8. Srebrenkoska V., Gaceva G.B. and Dimesk D., 2014. Biocomposites Based on Polylactic Acid and Their Thermal Behavior after Recycling, *Macedonian Journal of Chemistry and Chemical Engineering*, 33: 277-285.
- 9. Avella M., Buzarovska A., Errico M.E., Gentile G. and Grozdanov A., 2009. Eco-Challenges of Bio-Based Polymer Composites, *Materials*, 2: 911-925.
- 10. Mishra S.M., Drzal L.T., Parija S.N. and Tripathy S.S., 2003. Studies on Mechanical Performance of Biofibre/Glass Reinforced Polyester Hybrid Composites. *Composites Science and Technology*, 63(10): 1377–1385.

- 11. Faludi G., Dora G., Renner K., Moczo J. and Pukanszky B., 2013a. Biocomposite from Polylactic Acid and Lignocellulosic Fibers: Structure-property Correlations, *Carbohydrate Polymers*, 92(2): 1767-1775.
- 12. Fortunati E., Armentano I., Iannoni A. and Kenny J., 2010. Development and Thermal Behaviour of Ternary PLA Matrix Composites, *Polymer Degradation and Stability*, 95(11): 2200-2206.
- 13. Ghali L., Msahli S., Zidi M. and Sakli F., 2009. Effect of Pre-treatment of Luffa Fibres on the Structural Property, *Material Letters*, 63(1): 61-63.
- 14. Castellano M., Gandini A., Fabbri P. and Belgacem M., 2004. Modification of Cellulose Fibres with Organosilanes: Under What Conditions Does Coupling Occur? *Journal of Colloid and Interface Science*, 273(2): 505-511.
- 15. Kim H-S., Lee B-H., Choi S-W., Kim S. and Kim H-J., 2007. The Effect of Types of Maleic Anhydride-grafted Polypropylene (MAPP) on the Interfacial Adhesion Properties of Bioflour-filled Polypropylene Composites, *Composites Part A: Applied Science and Manufacturing*, 38(6): 1473 1482.
- 16. Finkenstadt V., Liu L. and Willett J.L., 2007a. Evaluation of Poly(lactic acid) and Sugar Beet Pulp Green Composites, *Journal of Polymers and the Environment*, 15: 1-6.
- 17. Clarizio S.C. and Tatara R.A., 2012. Tensile Strength, Elongation, Hardness, and Tensile and Flexural Moduli of PLA Filled with Glycerol-plasticized DDGS, *Journal of Polymers and the Environment*, 20: 638-646.
- 18. Ahmad I., Abu-Bakar D., Mokhilas S. and Raml A., 2007. Direct Usage of Products of Poly(ethylene terephthalate) Glycolysis for Manufacturing of Rice Husk/Unsaturated Polyester Composite, *Iranian Polymer Journal*, 16(4): 233-239.
- 19. Ewulonu C.M. and Igwe I.O., 2011. Properties of Oil Palm Empty Fruit Bunch Fibre Filled High Density Polyethylene, *International Journal of Engineering and Technology*, 3(6): 458-471.
- 20. Ibrahim M., Sapuan S. and Faieza A., 2012. Mechanical and Thermal Properties of Composites from Unsaturated Polyester Filled with Oil Palm Ash, *Journal of Mechanical Engineering and Sciences*, 2: 133-147.
- 21. Eze I.O., Obidiegwu M.U. and Eyarefe S.O., 2013. Some Mechanical and End-use Properties of Bamboo Powder Filled Low Density Polyethylene Composites, *Natural and Applied Sciences: Part-I*, 4(2): 199-207.
- 22. Koutsomitopoulou A., Bénézet J., Bergeret A. and Papanicolaou G., 2014. Preparation and Characterization of Olive Pit Powder as a Filler to PLA-matrix Biocomposites, *Powder Technology*, 255: 10-16.
- 23. Yussuf A.A., Massoumi I. and Hassan A., 2010. Comparison of Polylactic Acid/Kenaf and Polylactic Acid/Rice Husk Composites: The Influence of the Natural Fiberson the Mechanical, Thermal and Biodegradability Properties, *Journal of Polymers and the Environment*, 18: 422–429.
- 24. Huda M.S., Drzal L.T., Misra M. and Mohanty A.K., 2006. Wood-Fiber-Reinforced Poly(lactic acid) Composites: Evaluation of the Physicomechanical and Morphological Properties, *Journal of Applied Polymer Science*, 102(5): 4856–4869.
- 25. Subramonian S., Ali A., Amran M., Sivakumar L., Salleh S. and Rajaizam A., 2016. Effect of Fiber Loading on the Mechanical Properties of Bagasse Fiber–reinforced Polypropylene Composites, *Advances in Mechanical Engineering*, 8(8).DOI: 10.1177/1687814016664258.
- 26. Nwanonenyi S.C., Obidiegwu M.U. and Onuegbu G.C., 2013. Effects of Particle Sizes, Filler Contents and Compatibilization On the Properties of Linear Low-Density Polyethylene Filled Periwinkle Shell Powder, *International Journal of Engineering and Science*, 2(2): 1–8.
- 27. Părpărită E., Darie R., Popescu C-M., Uddin M.A. and Vasile C., 2014. Structure—morphology—mechanical Properties Relationship of Some Polypropylene/lignocellulosic Composites, *Materials and Design*, 56: 763-772.
- 28. Imoisili P., Olunlade B. and Tomori W., 2012. Effect of Silane Coupling Agent on the Tensile Properties of Rice Husk Flour (RHF) Polyester Composite, *The Pacific Journal of Science and Technology*, 13(1): 457-462.