IJARSCT
ISSN (Online) 2581-9429
International Journal of Advanced Research in Science, Communication and Technology (IJARSCT)
International Open-Access, Double-Blind, Peer-Reviewed, Refereed, Multidisciplinary Online Journal
Impact Factor: 7.53
Volume 4, Issue 2, December 2024

Utilization of Recycled Low-Density Polyethylene and Carbonized Neem Seed As A Filler in Production of House-Hold Plastic Utensil

Mustapha Basiru[1], Saidu Akun Abdallah[2] and Haruna Abubakar Danyaya[3]
Department of Science Laboratory Technology[1,2,3]
School of Science and Technology, Hussaini Adamu Federal Polytechnic Kazaure, Jigawa State, Nigeria
*Correspondence Email: abubakarharuna34y@gmail.com
ORCID ID: 0009-0006-3325-5597

Abstract*: Using natural fillers reduces the cost of production in plastic recycling. In this work, carbonized neem seed was used as filler to improve the properties of recycled low-density polyethylene. Physical and Mechanical tests conducted on the household plastic utensil produced, showed that, highest bulk density of 0.8±0.1g/cm^3 was found in 100% RLDPE, followed by 0.8±0.1 and 0.8±0.2 g/cm^3 in sample D and E with CNS content of 30g and 40g respectively. The water absorption revealed that, the quantity of water absorbed in the material increased with the increase in content of CNS. Carbonized Neem Seed absorbs water slowly due to its hydrophobic nature. The household plastic utensil with more content of CNS depicted more water absorption capacity. Considerable change in thermal properties of the household plastic utensil was also observed with the addition of CNS with the thermally stability in the order of 133±0.2, 133±0.1, 135±0.2, 140±0.3 and 137±0.2 ^{O}C respectively. The tensile strength and impact strength, also increased with the content of CNS in the household plastic utensil. The impact strength decreased as compared to that of the standard 100 %RLDPE materials used in the current study. FTIR spectrum of the activated carbon revealed the presences of O-H, C=O, P-C, and N-H, functionalities indicating the presence of alcohol, ketones, amides, organophosphorus compounds, and nitro groups. SEM images showed typical carbon morphology, retaining the original seed's structure. The carbonization process created pits with different sizes, forming micropores, mesopores, and macropores in activated carbons. SEM images of 100% RLDPE and five RLDPE-carbonized neem seed-household plastic utensil showed absence of spherulitic structure due to limited nucleation sites Filling RLDPE with more filler created more spherulites, which are linked to the crystallization of polymers from the melt and are characterized by increased density and hardness.*

Keywords: Organic waste: Plastic waste, Carbonized neem seed filler, FTIR characterization, scanning electron microscopy RLDPE-CNS.

I. INTRODUCTION

Increase in population has resulted in an increase in human activities which in turn has given rise to increase in the amount of solid waste generated (Salami et al., 2024; Salami et al., 2020; Salami et al., 2015). Researchers have stated that 1.3 billion tons of wastes are generated worldwide annually (Hoornweg and Bhada-Tata (2012) and by prediction, the volume of waste generation annually will reach about 2.2 billion tons by the year 2025 (Costa et al., 2019;Ishaq et al., 2022) and almost 4.2 billion tons by the year 2050 (Hoornweg and Bhada-Tata, 2012). Among these solid waste, plastics constitute the major component worldwide including African countries such as Nigeria. Plastic materials are consumed globally at an alarming level and this has resulted in producing massive quantities of plastic wastes daily which causes serious plastic pollution (Abdallah et al., 2021). Globally, demand for plastics continues to increase and it is estimated, that the amount of plastics in circulation will rise about 417 million ton per year by 2030 (Everard, 2020). Recycling of waste plastics is therefore essential to prevent accidental release of polymeric materials into the environment, and thus mitigate environmental pollution (Everard, 2020; Zoé, Schyns and Michael, 2020).

Low-density polyethylene (LDPE), known for its outstanding qualities such as durability, flexibility, and cost-effectiveness, is widely employed in manufacturing many utility products. LDPE is typically used in the manufacture of packaging such as sachet and bottled water, which is a significant contributor to this waste stream which is problematic due to its limited recyclability, which diminishes its quality over successive recycling cycles.

Meanwhile, in northern Nigeria, the abundance of organic waste, particularly neem seeds, presents another environmental challenge (Ezeonu & Ezeonu, 2016). Neem seeds are generated seasonally in large quantities and in most cases litter the environment, creating unsightly and hazardous conditions. However, several researchers have proved that these seeds hold enormous potential as a renewable resource (Ngulde & Yerima, 2012; Ezeonu & Ezeonu, 2016). Low density polyethylene (LDPE) and neem seeds are waste, and regarded as environmental pollutants. They are readily found in Kazaure town in Kazaure Local Government Area, Jigawa state. Effective utilization of these wastes will reduce environmental hazard. This research will explore the utilization of these waste in production of plastic RLDPE–Carbonized neem seed, household plastic utensil prepared for possible application and other purpose. When carbonized, neem seed waste can serve as an effective filler in recycled plastics, enhancing their properties, reducing production costs, and offering an eco-friendly solution to waste management challenges (Falope, Nwude & Ajayi, 2021).

II. MATERIAL AND METHODS

2.1 Materials and Equipment

The materials and equipment are of analytical grade and were used without any purification. This includes: Recycled low-density polyethylene (RLDPE), Neem seeds, Furnace, Durometer, Mosanto tensiometer, Impact tester and Metal Mould

2.2 Sample Collection

Hussaini Adamu Federal Polytechnic, Kazaure's Male and Female Hostles' garbage and waste disposal sites were the source of waste low density polyethylene (LDPE), which was hand-picked. Neem seeds were also collected within the Kazaure Local Government Area and transported to the Polymer Technology Laboratory for processing before being used in this study.

2.2.1 Sample Pretreatment

The adhesive dirt on the RLDPE was eliminated using a detergent solution followed by through rinsing under running tap water. The cleaned material was then air-dried in sun-light before being oven-dried at 40 ^{O}C to achieve a constant.

The neem seeds were manually extracted from their mesocarp. Cleaned and dried in an oven at 40 ^{O}C to a constant weight. Once dried, the seeds were ground into fine power using, a pestle and mortar. The powdered material was sieved using 20mm mesh, giving an average particle size of 0.9 mm. To remove any residual surface moisture from the sample, the processed neem was placed in a vacuum oven and dried for 24 hrs.

2.2.2 Carbonization of Neem Seed

Approximately 150 g of Neem seeds were placed in a clean porcelain crucible and introduced into a pre-cooled muffle furnace. The sample was subjected to thermal decomposing in an inert atmosphere, at 450ºC for 1 hr after the process, the crucibles were carefully removed and allowed to cool to room temperature in a desiccator to prevent absorption.

2.3 Characterization of Raw and Carbonized Neem Seed.

2.3.1. FT-IR Analysis

The raw and carbonated samples were analyzed using Fourier Transform Infrared Spectroscopy (FT-IR) to obtain a precise bio-mineralization results. A small portion of each sample was mixed with potassium bromide (KBr spectroscopy grade Merck) and ground into a uniform powder using an agate mortar. The mixture was compressed into pellets under a pressure of 7 tons. The spectral data was obtain using a Pelkin Elmer 3000 MX spectrometer over the range of 4000 to 400 cm^{-1}. Each spectrum was recorded with 32 scans the resolution of 4 cm^{-1}. The resulting IR spectra were analyzed using the Win-IR Pro software (Version 3.0), which provides a peak sensitivity of 2 cm^{-1}.

2.3.2 SEM Analysis

The scanning electron microscopy (SEM) analysis was carried out using a JOEL- Desktop SEM (JSM 7600F model). Both the raw Neem seed and carbonized samples were prepared by coating them with a thin layer of platinum to ensure conductivity. The samples were then sectioned into 6-inch (15 cm) pieces using a sputter cutting machine. The particle size and pore size distributions were examined under magnifications between X8000 -X10000. The analysis was conducted on both raw and carbonized Neem seeds and on the various RLDPE –Neem seed-based household plastic utensil produced.

2.3.2 Preparation of RLDPE –Neem seed- House Hold Plastic utensil

The RLDPE pellets was shredded into smaller particles using shredding scissors s to facilitate mixing with the carbonized Neem seed (CNS). The shredded RLDPE granules were then dried in a vacuum oven for 2 hrs, following this, the dried RLDPE granules were combined with CNS particles in accordance with the formulation specified in Table 3.3 The mixture was then fed into a mixing pot and blended until a constant torque was achieved within 10 min. Once mixed, the plastic composite was discharged into the mould from where it was shaped into the desired household plastic utensil. The mouldin procedure involved preheating at 150°C for 5 min followed by compression and cooling at the same temperature for an additional 2 min. The mould design used is this study is detailed in Appendix III.

Table 1: Formulation of the household plastic utensil

Sample	Carbonized Neem Need filler (%)	RLDPE (%)
A	0	100
B	10	90
C	20	80
D	30	70
E	40	60
F	50	50

2.4 Physical tests of the household plastic utensil

2.4.1 Water absorption test

Water absorption tests conducted on the household plastic utensil was performed according to ASTM D570-99. Three replicate samples were employed to perform the water absorption tests and the mean values were presented. The percent increase in weight during immersion was calculated as follows:

$$\text{Increase in weight, \%} = \frac{\text{wet weight} - \text{conditioned weight}}{\text{Conditioned weight}} \times 100$$

2.4.2 Bulk density

The bulk density of the household plastic utensil was determined using the formula.

$$D = \frac{\text{Weight of the RLDPE – (CNS- household plastic utensil prepared}}{\text{(Length × Width × Height) of the utensil}}$$

2.4.3 Melting point test

A sample of the product (RLDPE – CNS-household plastic utensil) was placed in to a capillary tube which was then inserted into a melting point apparatus. The temperature at which the sample began to melt was recorded.

2.5 Mechanical test

2.5.1 Tensile strength (TS) (ASTM D 1708)

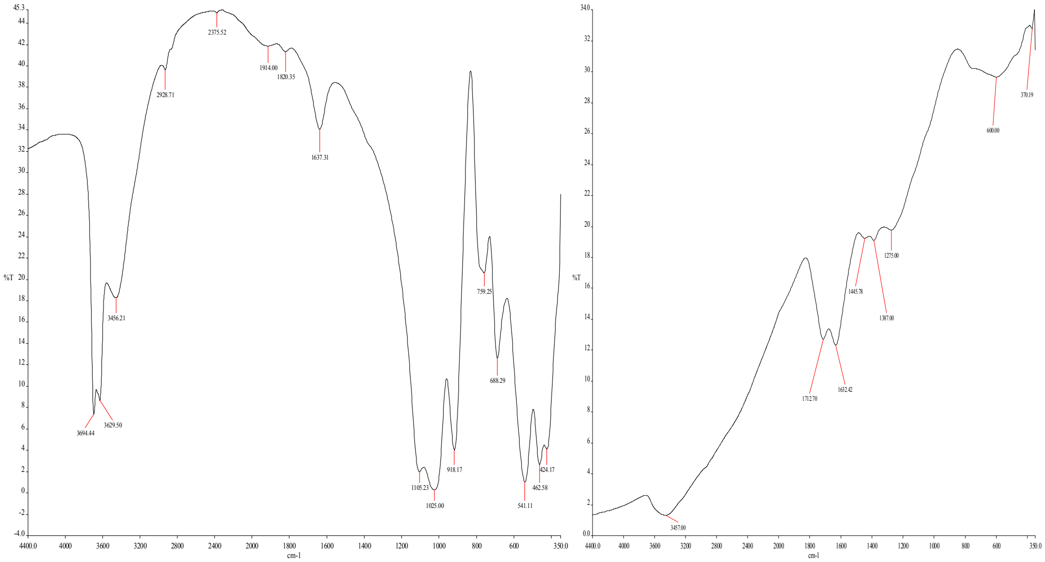
The NSP-based household plastic utensil and the standard LDPE samples were cut into one of the five ASTM D638 Specimen “dumbbell” shapes and mounted into tensile grips. An extensometer was attached to each sample and the tensile grips was operated at a constant speed of 20 inches per minute. The time taken for both the household plastic utensil and the standard LDPE to break was recorded.

2.5.2 Impact Strength (IS) ASTM D 256,

With the striking hammer (pendulum) secured in its safe position, the specimen was positioned in the impact testing machine’s vice in such a way that the notch faced the hammer with half of the specimen inside and half above the top surface of the vice. The striking hammer was raised to its maximum striking position and the indicator of the machine was set to zero. The hammer was then released allowing it to fall freely under gravity. Upon impact with the specimen, through its momentum the specimen broke. The pendulum continued to swing as the total energy was not completely absorbed by the specimen. At the highest point after breaking the specimen, the pendulum came to a stop and the indicator marked the final position. The value observed was recorded as the measurement of energy absorbed during the impact test.

III. RESULTS

The FTIR and SEM analyses of the raw and carbonized neem seeds along with that of bulk density, melting point, tensile strength (TS), impact strength (IS), of the raw LDPE and RLDPE –neem seed- household plastic utensils (B to F) are presented and discussed.



(b)

Figure 1: FTIR Spectrum of (a) raw neem seeds and (b) carbonized neem seed

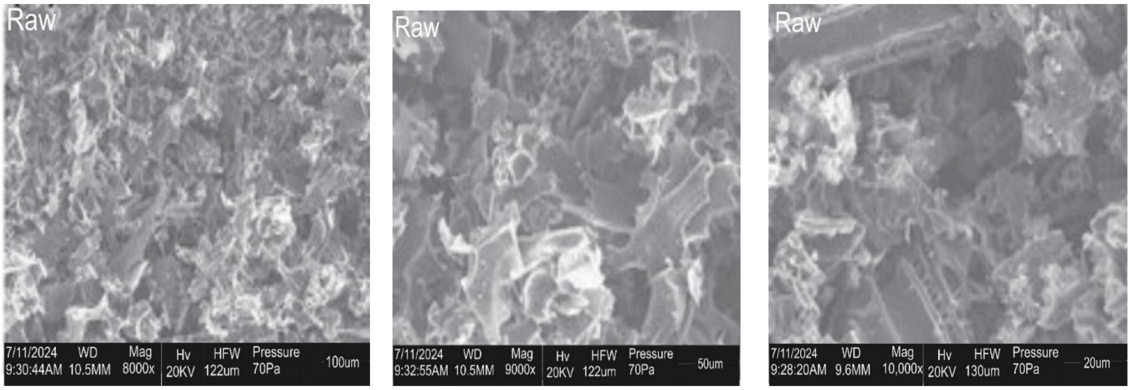


Figure 2: SEM result of raw neem seed at (a) X 8000, (b) X 9000 and (c) X 1000, respectively

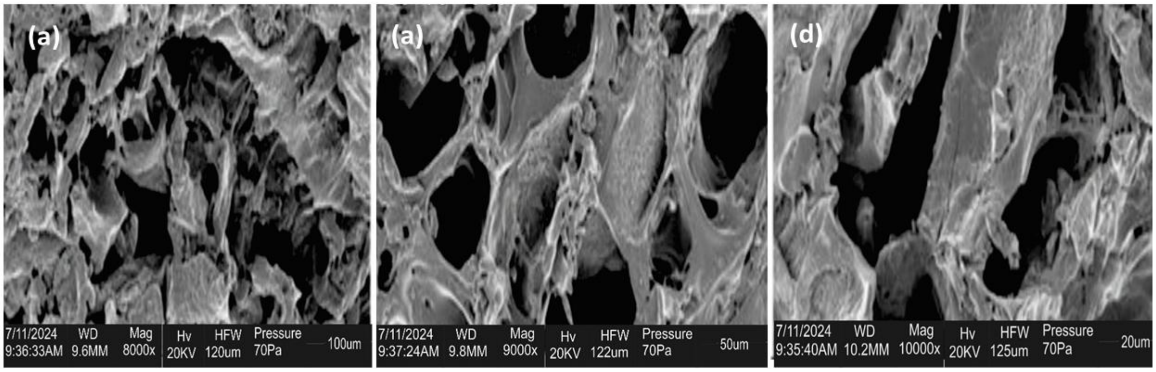


Figure 3: SEM image of carbonized neem seed at (a) X 8000, (b) X 9000 and (c) X 1000, respectively.

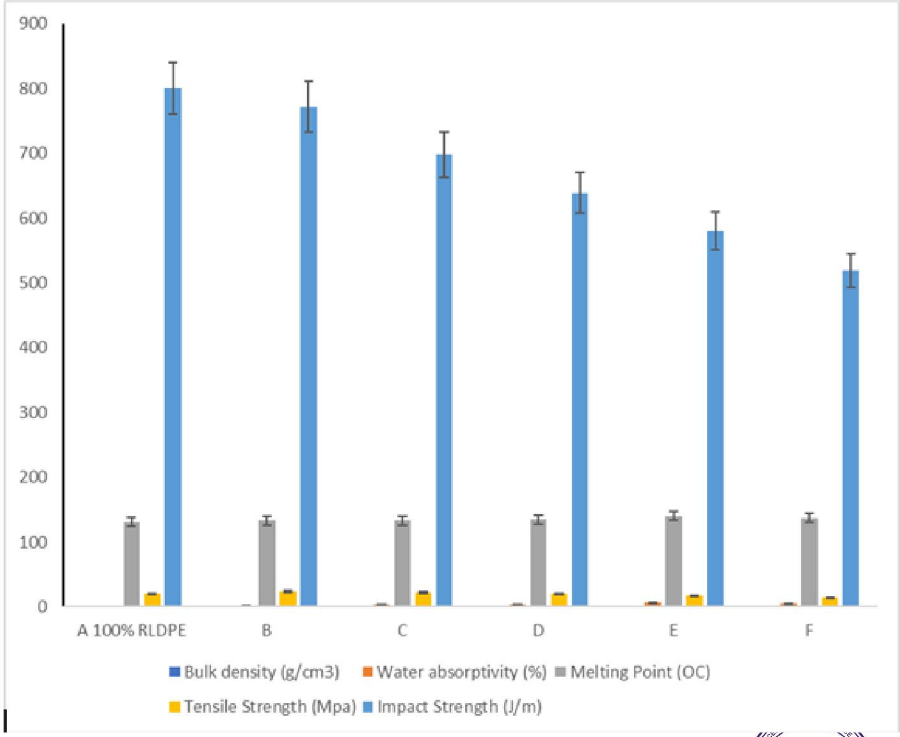


Figure 4: Physicomechanical test conducted on the RLDPE-CNS-based household plastic utensil

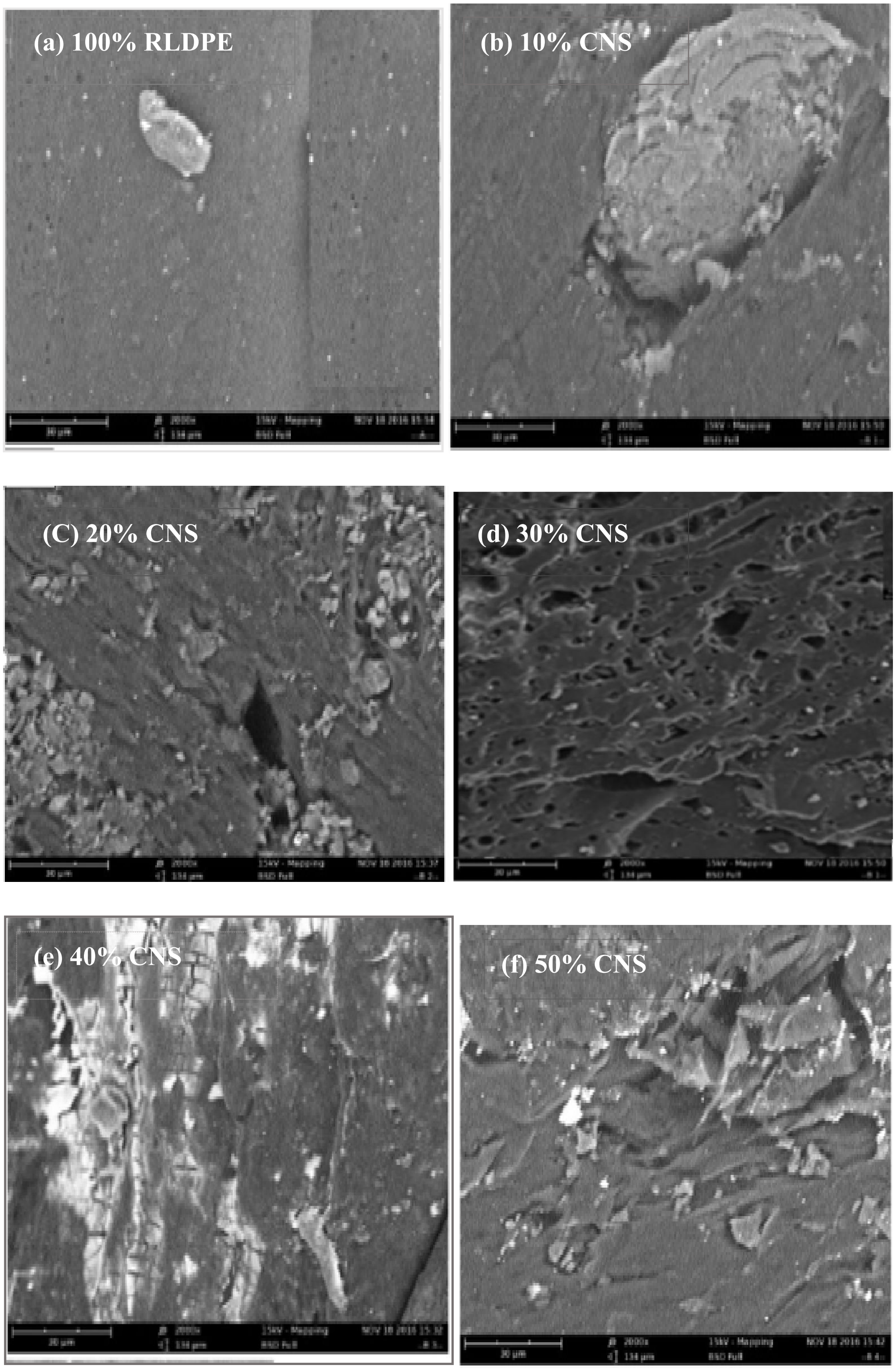


Figure 5. SEM images of RLDPE – CNS-based household plastic utensil prepared with (a) 100% RLDPE, (b) 10%CNS, (c) 20% CNS, (d) 30% CNS (e) 40% CNS and (f) 50% CNS. Key: CNS= Carbonized neem seed

IV. DISCUSSION

Figure 4 (a) depicts the impact of Carbonized Neem Seed (CNS) on the bulk density of the RLDPE –CNS- household plastic utensils. The data indicates that the highest bulk density of 0.8±0.1g/cm^3 was observed in 100% RLDPE, followed by 0.8±0.1 and 0.8±0.2 g/cm^3 observed in samples D and E which contained 30g and 40g of CNS, respectively. On the other hand, samples B, C and F with CNS containing 10, 20 and 50 g of CNS, showed lower bulk densities of 0.7±0.2, 0.7±0.1 and 0.6±0.0 g/cm^{-3}, respectively. The observed reduction in bulk density in sample F is attributed to CNS content. The trend where bulk density decreases with the increase in reinforce content was observed in jute-reinforced LDPE composites as reported in Jahan *et al.,* (2012).

The effect of different content of CNS on water absorption of the house plastic utensil is shown in figure 4(b). The result reveals that, the amount of water absorbed by the composite increased with the increase in content of CNS. The CNS absorbs water slowly due to its hydrophobic nature. The household plastic utensil with containing more CNS exhibited higher water absorption. Therefore, sample C and E household plastic utensils depicting 3.5±0.1 and 6.3±0.2 % water absorption showed highest quantity of water absorption compared to samples D and sample F with 2.9±0.2 and 5.2±0.1 respectively.

Figure 4 (c) reveals that the household plastic utensils show higher thermal stability compared to standard RLDPE which recorded melting point of 131±0.2 ^{O}C. The results indicate that there, was a significant change in thermal properties of the household plastic utensil due to the addition of CNS material. The thermal stability of the household plastic utensil followed the order: 133±0.2, 133±0.1, 135±0.2, 140±0.3 and 137±0.2 ^{O}C for B, C, D, E and F respectively. The result of this finding also agrees with the work of Afroza *et al.,* (2018). The high thermal stability of these household plastic utensil is attributed to the high-water absorption capacity with sample E showing the highest water absorption and thermal stability, respectively.

The Figure also shows the tensile strength and impact strength of the 100 % RLDPE, standard and the RLDPE-CNS household plastic utensil at varying CNS content. The figure clearly shows that, as the content of CNS increases in the household plastic utensil, the tensile strength increased especially in sample B and C and decreases from sample D to F. In contrast, the impact strength on the other hand decreases as compared to that of the standard 100 %RLDPE materials used in the current study. The result of the tensile strength is consistent with the findings of Fares *et al.,* (2010), while that of the impact strength agrees with the work of Afroza *et al.,* (2018).

As shown in Figure 1, the FTIR spectrum of the generated activated carbon reveals the presence of functional groups including: O-H, C=O, P-C, and N-H respectively, which correspond to the presence of alcohol, ketones, amides, organophosphorus compounds, and nitro groups, respectively. Similarly, the FTIR analysis of the raw neem seed (figure .1) shows the presence of C-H and C=C for monosubstituted alkenes, C-O of esters, and N-H for primary amine, which is also present in the carbonized seed. The RLDPE-neem seed-household plastic utensil' enhanced changing characteristics and thermal stability may be ascribed to the functional group on the activated carbon, which acts as a free active site for binding. The result of this analysis aligns with the findings of Priyanto *et al.,* (2017).

The SEM images of both the raw and carbonized neem seeds at x 8000, x 9000, and x 10,000 illustrated in Figures 2 and 3 show a typical morphology of carbons derived from biomass. These images indicates that part of the samples viewed images at various magnifications retained the original cellular structure of the seed. The carbonization process results in the production of pits of different sizes, creating unique micropores, mesopores, and macropores in the carbonized samples. Furthermore, the SEM images demonstrate that the carbon matrix of neem seed-based carbonized contains pits and fissures.

Figure 5 (a-f) presents the SEM images of 100% RLDPE and the five RLDPE-carbonized neem seed-household plastic utensil made. Figure 5(a) shows the identification of filler particles in 100% RLDPE (i.e., unfiltered sample A) at x 2000 magnification. Identifying the filler particles is challenging since the image lacks spherulitic structure due to absence of many nucleation sites as a result of melt solidification. On the other hand, Figure 5 (b) displays the image at x 2000 magnification for the RLDPE-carbonized neem seed-household plastic utensil filled with 10% filler loading (Sample B). The addition of the carbonized neem seed as a filler introduced more nucleation sites leading to more spherulites in the structure. As a result, the filled RLDPE exhibits more spherulites than the unfilled.

Once more, the CNS's darkening effect makes the filled RLDPE image appear darker than the 100% RLDPE image. Similarly, RLDPE filled with 20%, 30%, 40%, and 50% CNS filler loading is shown in Figure 5 (c) to (f) (x 2000). The addition of fillers to the RLDPE matrix significantly increased the number of nucleation sites resulting in a higher density of spherulites in the structure. However, the quantity of the spherulites involved restricts their growth as their expansion stops upon coming into contact with their neighbouring growing spherulite. The RLDPE with 20% filler has more spherulites than the one filled with 10% filler. Again, the 30% RLDPE filled sample exhibits more spherulites than that filled up to 20% or 10%. At the highest filler loading of 40%, the structure displayed the greatest number of spherulites, which are tiny in size as shown in Figure 5 (F). This observed trend continues up to 50% filled RLDPE lower filler levels as already described.

The crystallization and properties including density, hardness etc., of polymers depend on the formation of spherulites. Non-branched linear polymers have these spherical semicrystalline structures.

The number of nucleation sites, the structure of the polymer molecules, the rate of cooling, and other factors all influence the formation of spherulites, which are linked to the crystallization of polymers from the melt and are characterized by increased density, hardness, and brittleness when composed of highly ordered lamellae as opposed to the disordered polymer. When compared to the unfilled or control samples, the images of the filled RLDPE samples showed improved spherulization because of the nucleation sites provided by the fillers.

V. CONCLUSIONS AND RECOMMENDATION

In this study, hybrid household plastic utensil exhibited notable improvements in tensile strength at B and C filler content followed by a decline in performance from D to F. As a result, the B and C content showed superior mechanical qualities. All the hybrid household plastic utensils were found to have superior thermal stability over 100% RLDPE, indicating a significant change in thermal characteristics. Comparing the hybrid household plastic utensil to the 100% RLDPE, it was discovered that the F hybrid was the lightest in density, followed by B and C. In terms of water absorption capacity, samples, B and D showed the lowest absorption rates. In the overall, sample B was observed to have exceptional performance in this study. It is suggested that further research be conducted on various agricultural wastes to determine which ones are best suited for use as reinforcement in the creation of polymeric materials, as these materials are found to have numerous applications in both home and industrial settings. Government support for agricultural extensions and encouragement of farmers, businesses, and the general public to recycle their waste into valuable materials for the community should continue.

ACKNOWLEDGEMENT

The researcher sincerely thanks Almighty Allah for giving him the capacity and allowing him to conduct this study. The management of Hussaini Adamu Federal Polytechnic, Kazaure is highly appreciated for providing the conducive environment for the study. Special appreciation goes to the Tertiary Education Trust Fund (TETFund for providing the financial support to conduct the research.

REFERENCES

[1]. Abdallah, S. A., Tay, K. S., & Low, K. H. (2021). Removal of Hg (II) ions with CD/DVD waste-derived aminated polycarbonate: adsorption and optimization studies. *Desalination and Water Treatment*, *231*, 229-243.

[2]. Abdel-Shafy, H. I., & Mansour, M. S. (2018). Solid waste issue: Sources, composition, disposal, recycling, and valorization. *Egyptian journal of petroleum*, *27*(4), 1275-1290.

[3]. Al-Maaded, M., Madi, N. K., Kahraman, R., Hodzic, A., & Ozerkan, N. G. (2012). An overview of solid waste management and plastic recycling in Qatar. *Journal of Polymers and the Environment*, *20*, 186-194.

[4]. American Energy Council (2019). Plastics in Automotive Fact Sheet – AEC, Transport Team. Retrieved from https://plastics.americanchemistry. com/fact-sheets-and-infographics/plastics-in-automotive.pdf

[5]. Barclay, G. F. (2015). Anatomy and morphology of seed plants. *eLS*.

[6]. Dey, S., Veerendra, G. T. N., Babu, P. A., Manoj, A. P., & Nagarjuna, K. (2024). Degradation of plastics waste and its effects on biological ecosystems: A scientific analysis and comprehensive review. *Biomedical Materials & Devices*, *2*(1), 70-112.

[7]. Everard, M. (2020). Twenty years of the polyvinyl chloride sustainability challenges. *Journal of Vinyl and Additive Technology*, *26*(3), 390-402.

[8]. Ezeonu, C. S., & Ezeonu, N. C. (2016). Alternative sources of petrochemicals from readily available biomass and agro-products in Africa: A review. *J Pet Environ Biotechnol*, *7*(301), 2.

[9]. Falope, F. Y., Nwude, D. O., & Ajayi, F. T. (2021). Use of Neem (Azadirachta Indica) Seeds as Filler in Natural Rubber Compounding. *ChemSearch Journal*, *12*(1), 78-84.

[10]. Fares, D.; Alsewailem and Yazeed A. Binkhder, (2010). Petroleum and Petrochemicals Research Institute, King Abdualziz City for Science and Technology, P.O. Box 6086 Riyadh 11442, Saudi Arabia *Journal of Reinforced Plastics and composites l*, Vol. 29, No. 11/2010 1743 0731-6844/10/11 1743–7 $10.00/0 DOI: 10.1177/0731684409337339

[11]. Farzadkia, M., Mahvi, A. H., Norouzian Baghani, A., Sorooshian, A., Delikhoon, M., Sheikhi, R., & Ashournejad, Q. (2021). Municipal solid waste recycling: Impacts on energy savings and air pollution. *Journal of the Air & Waste Management Association*, *71*(6), 737-753.

[12]. Ganesh, K. S., Sridhar, A., & Vishali, S. (2022). Utilization of fruit and vegetable waste to produce value-added products: Conventional utilization and emerging opportunities-A review. *Chemosphere*, *287*, 132221.

[13]. Ghanta, M., Fahey, D., & Subramaniam, B. (2014). Environmental impacts of ethylene production from diverse feedstocks and energy sources. *Applied Petrochemical Research*, *4*, 167-179.

[14]. Jahan, A,; Rahman, MM,; Kabir H, Kabir, MA,; Ahmed F, et al. (2012) Comparative study of physical and elastic properties of jute and glass ber reinforced LDPE composites. *International Journal of Scientic & Technology Research* 1: 68-72.

[15]. Lettieri, P.; and S.M. Al-Salem; book for Management, T.M. Vallero and D.A. Letcher, Editors. 2011, *Academic Press*: Boston. p. 233-242

[16]. Ligon, S. C., Liska, R., Stampfl, J., Gurr, M., & Mülhaupt, R. (2017). Polymers for 3D printing and customized additive manufacturing. *Chemical reviews*, *117*(15), 10212-10290.

[17]. Murthy, K. S. R., Pullaiah, T., Bahadur, B., & Krishnamurthy, K. V. (2017). Ethnobotany of the Neem Tree (Azadirachta Indica a. Juss): A Review. *Ethnobotany of India, Volume 5*, 573-599.

[18]. Ngulde, Y. M., & Yerima, I. (2012). IMPROVEMENT OF THE THERMAL EFFICIENCY OF BIOGAS PRODUCED FROM NEEM LITTER.

[19]. Nwanegbo, I. M., Ajagba, P. A., & Egbebu, J. A. (2024). Trace Elements Content of Neem Leaves and Mechanical Extraction of Neem Oil (Azadirachta indica). *International Journal of Pure and Applied Science*.

[20]. Priyanto, A., Fuadah, A., & Aji, M. P. (2017). Performance of porous composite from waste glass on salt purification process. *Procedia engineering*, *170*, 41-46.

[21]. Schyns, Z. O., & Shaver, M. P. (2021). Mechanical recycling of packaging plastics: A review. *Macromolecular rapid communications*, *42*(3), 2000415.

[22]. Sultana, S., Parvin N.; H.and Sarwaruddin C. AM.; (2018). Physical, mechanical, thermal and morphological analysis of date palm mat (DPM) and palmyra palm fruit (PPF) ber reinforced high density polyethylene hybrid composites (pdf/AMS-4-153.pdf) DOI: 10.15761/AMS.1000153