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Original Research Article

Physical and Mechanical Properties of Agro-Waste Filled Recycled High Density Polyethylene Biocomposites

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Abstract: Natural plant fiber/agricultural waste materials have potential in composites due to their eco-friendliness, low cost and sustainability. Plastic and agro-wastes dumping are threatening concerns that must be settled for the protection of the worldwide ecosystem. In this study, the physical and mechanical properties of rice husk flour (RHF), corncob flour (CCF), walnut shell flour (WSF) and wood flour (WF) agro-waste filled recycled high density polyethylene biocomposites were investigated. Results showed bulk densities of RHF 350 kg/m^3, CCF 310 kg/m^3, WSF 400 kg/m^3 and WF 250 kg/m^3. Results showed moisture content of RHF 7%, WSF 6.4%, CCF 6% and WF 8%. Particle size distribution of 60-100 mesh size of the fillers was 0.295 mm to<0.125 mm. Results showed that WF composite showed higher flexural modulus of 3.0 GPa and impact strength of 60 J/m followed by RHF with flexural modulus of 2.75 GPa and WSF with impact strength of 54.4 J/m compared to the control sample of 1.75 GPa and 38 J/m. The flexural strength of WF composite was 27.4 MPa followed by RHF composite 25.4 GPa, CCF composite 20.1 GPa and WSF composite 18.1 GPa compared to the control sample of 30.5 GPa. The higher bulk densities of RHF, WSF and CCF resulted in fiber accumulation at some parts of the composite, thereby causing weak points and the resultant lower mechanical properties compared to WF composites with lower bulk density. The study has shown that agro-waste fillers could be used in composite production with good results compared to WF composites.

Keywords: Agricultural waste, Sustainability, Mechanical properties, Physical properties, Bulk density.

INTRODUCTION

Increased activity in the modern agricultural outfit generates a significant amount of waste, posing a substantial environmental concern. Plastics importance and vast spectrum of applications in human daily existence cannot be overemphasized. Meanwhile, raw material paucity is creating concern, and agricultural waste could be a viable alternative for generating value-added products such as bio-composites. As a long-term strategy for exploiting the tremendous amount of agro-waste materials that is now underutilized, natural fibers particularly agricultural waste fibers require further development. Natural fiber reinforced polymer composites are a scientific and technological innovation in the field of novel materials emphasizing the use of agricultural waste/plastic waste as a source of raw materials. As a result of their high specific strength, high modulus, reusability, green origin, large scale application, easy workability, and relatively cheap, lignocellulosic composites have gained a lot of interest. Eco-friendly composites have made significant strides since they are non-toxic, biodegradable, and safer to work with [1, 2]. The inclusion of plant fibers/agro-fillers such as corncob, rice husk, wood flour and walnut shell as natural filler is important for reinforcing polymers. The usage, reuse, and recycling of agricultural waste, as well as the addition of natural filler to the composite material, can assist to resolve environmental challenges related with their accumulation[3, 4]. With the exception of fibrous natural fillers, the loading of dispersed agricultural waste into the polymer matrix is currently receiving a lot of attention. As a result, lignocellulosic plant waste, such as shells, husks, and stems of agricultural crops can be used not only as a source of biomass but also as low-cost dispersion fillers for polymer composites.

As a result of the global request for fibrous materials, worldwide decline of trees in different countries and environmental consciousness, studies on the development of composites produced using varying waste materials is being actively investigated. Included in the possible substitutes is the development of composites using agricultural wastes such as stalks of most cereal crops, rice husks, coconut fibers, bagasse, maize cobs, peanut shells, and other wastes [5]. Lately, lignocellulosic materials like wood fiber, wood flour and agricultural wastes have been extensively utilized as reinforcing fillers in natural fiber-thermoplastic polymer biocomposites. These biocomposites which are prepared using lignocellulosic materials as reinforcing fillers are inexpensive, biodegradable, thus causing zero environmental pollution and non-toxicity to the users.

Agricultural wastes such as rice husk, sugar cane, bagasse, and so forth are very important natural materials and their ability to act as replacement for wood fiber is of great use, because of the lack of forest resources and spiking levels of environmental pollution. These natural materials are of lower density, low-cost and offer improved strength per unit weight compared to the inorganic reinforcing fillers like carbon black, calcium carbonate, talc and zinc oxide. Most of all, rice husk, an agricultural industrial residue which is easy to obtain, is one of several lignocellulosic materials, which is produced as a by-products during the rice milling process in rice-producing countries [1]. This rice husk is chiefly utilized as natural reinforcing filler in the form of rice husk ash through the burning process due to its high silica content. In the present study rice husk flour was used in order to simplify the production process. A thermoplastic polymer, a polyolefin, was used as a matrix polymer to compound with the natural fillers for the purpose of fabricating the biocomposite. The use of natural fiber-thermoplastic polymer biocomposites has been surging in the field of constructions (wood decks, window frames, and bathroom interiors) and the automotive industry (dashboard). Besides, some industries especially the marine and chemical industries have also used these composite systems.

Spontaneous increase in manufacturing industries has culminated to the need for the improvement of materials in terms of strength, stiffness, density, and lower cost with enhanced sustainability. Composite products have resulted as one of the materials having such enhancement in properties serving their purposes in various applications [6-9]. Composite materials are a combination of two or more constituents, one of which is present in the matrix phase, and another one could be in particle or fiber form. The use of natural or synthetic fibers in the manufacture of composite materials has shown vital applications in many fields such as construction, mechanical, automobile, aerospace, biomedical, and marine [10-13].

The addition of luffa fibers (LFs) as a reinforcement constituent of composite material resulted in the increase of the mechanical properties like tensile, compressive, flexural, impact strength, and water absorption characteristics [14]. Adding a 9.6 wt % of LFs in epoxy matrix caused a decrease in the density of the material by 3.12%, which further resulted in the reduction in material weight [15].

Palm fibers (PFs) showed increased fiber-matrix interfacial interaction. Also, the addition of palm fibers in low-density polyethylene (LDPE) resulted in increased Young's modulus compared to homo-polymers [16]. Friction composites are fabricated using abaca fiber (AF) reinforcement, which offers excellent wear resistance property with a wear rate of 2.864×10^{-7} cm^3/Nm at 3% of fiber content and the density decreased with increasing abaca fiber content [17].

Randomly oriented coir fiber-reinforced polypropylene composites give increasing damping properties compared to synthetic fiber-reinforced composites. High resin content show increased damping properties, therefore, lower fiber loading caused more energy absorption. The maximum damping ratio of 0.4736 was obtained at 10% of fiber content in coir–PP composite, while further increase in fiber content to 30% showed improved natural frequency of material to 20.92 Hz [18, 19].

A natural sheath forms around the grain of rice during the growth of rice seedling known as rice husk which is discarded as agricultural waste. This rice husk agro-waste is utilized as reinforcement in composite materials to investigate enhancement in material properties [20, 21]. For the improvement of the acoustic characteristics of the composite material, 5% of rice husk in polyurethane foam showed optimum sound absorption performance [22].

Polylactic acid (PLA) thermoplastic composites with kenaf fiber reinforcement gave tensile and flexural strength of 223 MPa and 254 MPa, respectively [23]. Also, before laminating, removal of absorbed water from the fibers resulted in the improvement of both flexural and tensile properties of kenaf fiber laminates [24, 25]. Formerly, polyester samples without reinforcements showed flexural strength and flexural modulus of 42.24 MPa and 3.61 GPa, while after reinforcement of 11.1% alkali-treated virgin kenaf fibers in unsaturated polyester matrix, composite material showed flexural strength and flexural modulus of 69.5 MPa and 7.11 GPa [26].

The aim of the present study was to investigate the physical and mechanical properties of rice husk, corncob, walnut shell agro-waste and wood flour in combination with recycled high density polyethylene, in order to alleviate the environmental problems created by plastic and agro-waste litters in the environment, by converting them into useful composite products.

MATERIALS AND METHODS

Bulk density

Bulk density is the mass of many particles of a material divided by the total volume it occupies. The total volume includes particle volume, inter-particle void volume and internal pore volume. Bulk density is basically how a material will compact under various loads and can also be an indicator of flow, the higher the bulk density of the material the greater the flowability. Determinations of the bulk densities of RHF, CCF and WSF respectively were performed and then compared to that of wood flour (WF). A measuring container of known volume (0.25 ft^3) was weighed and tared. The tare of a container is its weight when it is empty, which is important to know when you can't weigh something without putting it into something else. Tare weight = unladen weight is the weight of an empty container. By subtracting tare weight from gross weight, one can determine the weight of the goods carried or contained. The container was then filled to the brim with the RHF, CCF, WSF and WF respectively. The volume of the container filled with each type filler was standardized with no tapping and reweighed. The differences in the weights of the container containing the filler and that of the empty container gave the weight of the filler.

Moisture content

The moisture contents of the RHF, CCF, WSF and WF were measured using the oven-drying method (ASTM D4442-07). An aluminum dish was weighed using a digital balance (Model PB 1502-S, Mettler Toledo Instrument, Switzerland). A 10 g sample of each agro-waste filler of 60-100 mesh were placed in a dish and the dish and samples were weighed. The dish and samples were then placed in an Isotherm drying oven (600 Series, Thermo Fischer Scientific Inc., Walthan, USA) and kept at 103±2^oC for 48 hours, until a constant weight was achieved. The dish containing each of the dried samples were cooled to room temperature in desiccators and then reweighed. The experiments were replicated thrice for reproducibility. The moisture content of the fillers was calculated on a dry weight basis as follows:

$MC = W_w – D_w/D_w \times 100$ …………………………......1

Where, MC= the moisture content (%); Ww= the wet weight of the sample and dish (g); D_w= the dry weight of the sample and dish (g).

Particle size distribution

The particle size distribution of the RHF, CCF, WSF and WF of 60-100 mesh were analyzed on oven dried (OD) fillers (100±1g) using a mechanical sieve shaker (Model Rx-86) with standard test sieves of 50, 60, 70, 80, 100, 120 mesh and pan for 10 min according to the Rotap A Method (ASTM D5644-010). The sieves were mounted on the mechanical sieve shaker powered by an electric motor operating at 28.75 to 29.17 Hz (1725-1750 rpm). A 100g sample of each agro filler was placed in sieve 1 (50 mesh), which was covered with the sieve lid. The shaker was operated at a speed of 350 rpm for 10 min. The particles collected at the bottom of each sieve were weighed and the percent weight was calculated.

Collection and preparation of plastic waste

The polyethylene plastic waste was collected from MCC dumpsite in Awka, Anambra State, Nigeria and using physical identification of plastics HDPE waste were sorted out. The polyethylene plastic waste materials (r-HDPE) were first taken to the hydraulic cutting machine to cut the plastic lumps into smaller sizes before transferring to the grinding/crushing machine for grinding/crushing and scaling. The ground/crushed plastic waste was poured into a rectangular washing bowl filled with clean water and the polyethylene (r-HDPE) that floated was collected by the use of a sieve and the ones that sank was discarded. The washed plastic waste was sun dried for 24 h and packaged for further use.

Compounding

The compounding of r-HDPE, RHF, CCF, WSF and WF and maleic acid anhydride grafted polyethylene (MAPE) was conducted according to the formulation as shown in Figure 1. The materials were mixed using an internal mixer at the Materials Engineering Workshop, Nnamdi Azikiwe University, Awka, Nigeria for 10 min at a mixing temperature of 160^oC and stirring speed of 50 rpm to achieve constant torque. After mixing the components were stored in air-tight polythene bags prior to subsequent processing.

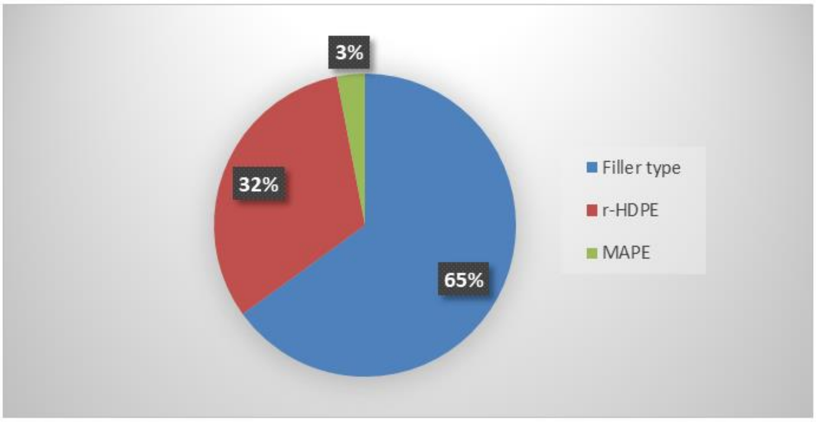

Fig 1: Composites formulation

Composite Fabrication

Fig 1 presents the formulation of the mixes used for the composite fabrication. The various mixes of the RHF, CCF, WSF and WF, r-HDPE and MAPE were poured into the hopper feeder of a co-rotating twin-screw extruder. The temperature of the extruder barrel was maintained at a temperature of 160°C at a screw speed of 50 rpm and melt pressure of 10 psi. The rectangular extrudates profiles that emerged from the extruder nozzle were cooled by immersing in a water bath, as shown in Fig 2.

Fig 2: Extruded rectangular profiles

Flexural testing

The flexural properties of the composites were determined according to ASTM D790-10. At least three replicates of each composites specimen of dimensions of 203 x 38 x 10 mm were cut from the extruded samples and stored for 48 h at 23±2°C and 50±5 relative humidity (RH) in a conditioning room. The test span was 152 mm an Instron Universal Testing Machine (Model 4466) was used for the testing. The machine was equipped with 8.5kN load cell and a support span 20 times the depth of the beam with a crosshead speed of 3.8 mm/min. The flexural strength of the composite samples was calculated using the equation:

Flexural strength (MOR) $б_f = 3P_{max}L/2bd^2$……………………….. 2

Where: $б_f$ = stress in the outer fiber at midpoint, MPa; P_{max} = maximum load at a given point on the load-deflection curve, N; L = support span, mm; b = width of beam tested, mm; d = depth of beam tested, mm.

The flexural modulus was calculated using the equation:

Flexural modulus (MOE) $E_b = L^3m/4bd^3$…………………………… 3

Where: E_b = modulus of elasticity in bending, GPa; L = support span, mm; b = width of beam tested, mm; d = depth of beam tested, mm and m = slope of tangent to initial straight line portion of the load-deflection curve, N/mm.

Un-notched Izod impact strength

The un-notched Izod impact strength was conducted according to ASTM D256, using a Pendulum Type Impact Tester. Three replicates of each composites specimen of dimensions of 63 x 12 x 3 mm were cut and stored for 48 h at 23±2°C and 50±5 RH in a conditioning room. The impact strength is calculated by dividing impact energy in J by the area under the notch. A pivoting arm is raised to a specific height and then released. The arm swings down hitting the sample and breaking it.

RESULTS AND DISCUSSIONS

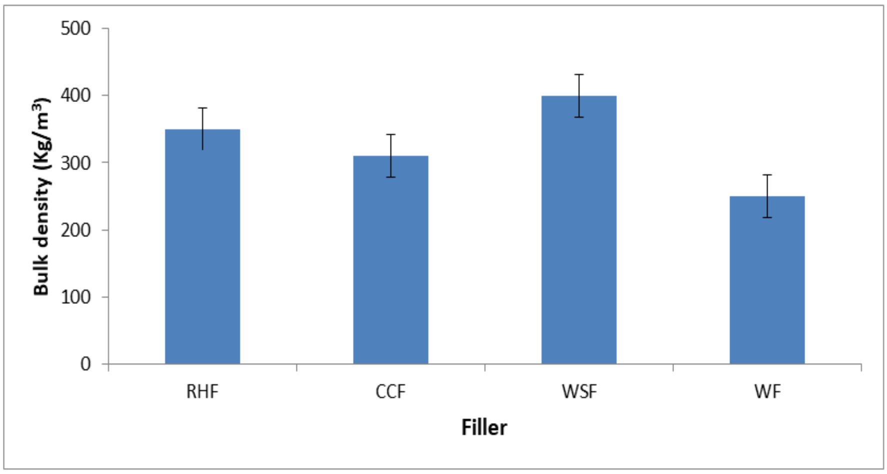

Fig 3: Bulk density against filler loading

Figure 3 shows the variation in bulk density against filler loading. It was observed that there were differences in the bulk densities with filler percentage weight. Walnut shell gave highest bulk density of 400 kg/m^3, followed by rice husk 350 kg/m^3 and corncob 310 kg/m^3, while wood flour gave the lowest bulk density of 250 kg/m^3. Low bulk density of filler may present some challenges in the feeding and mixing section of an extruder. Therefore, the increase in walnut shell, rice hull, or corn cob's filler content to 65wt%, as these fillers have bulkier fibers, resulted in fiber accumulation at some parts of the composite. Therefore, these phenomena can give rise to weak points and result in lower strength of specimens with 65wt% loading of walnut shell, rice husk and corncob compared to WF composite with a lower bulk density.

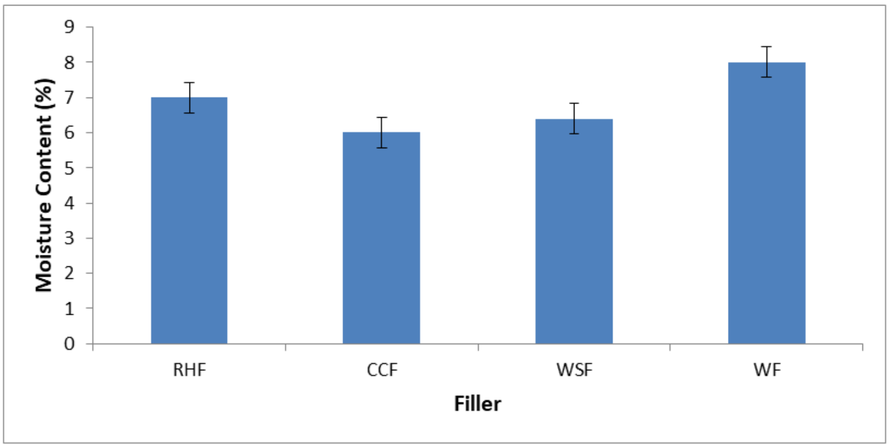

Fig 4: Moisture content of fillers

Figure 4 shows the moisture content of the fillers. From the figure, it was found that wood flour (WF) recorded the highest moisture content value of 8%, followed by rice husk flour (RHF) with moisture content of 7%, WSF with moisture content of 6.4% and corncob flour (CCF) with moisture content of 6% respectively.

Particle size distribution

Figure 5 shows the particle size distribution of CCF, RHF, WSF and WF using 50, 60, 70, 80, 100, and 120 mesh size (0.295mm to < 0.125 mm) and the pan. About 72 % of RHF was distributed (very fine particles) in the pan (< 0.125 mm). About 20-35 % of WSF was distributed (medium particles) in the 70, 80,100 mesh size (0.211 to < 0.152 mm). About 55 % of WF was distributed (high particles) in 50 mesh size (0.295 mm). About 10-30 % of CCF was distributed in (medium to very fine particles) in the 70-120 mesh size (0.211 to < 0.125 mm) and pan.

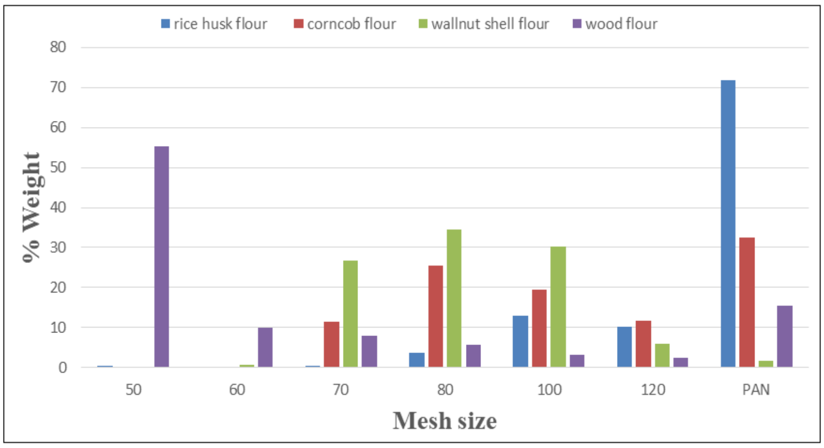

Fig 5: Particle size distribution of fillers

Flexural strength

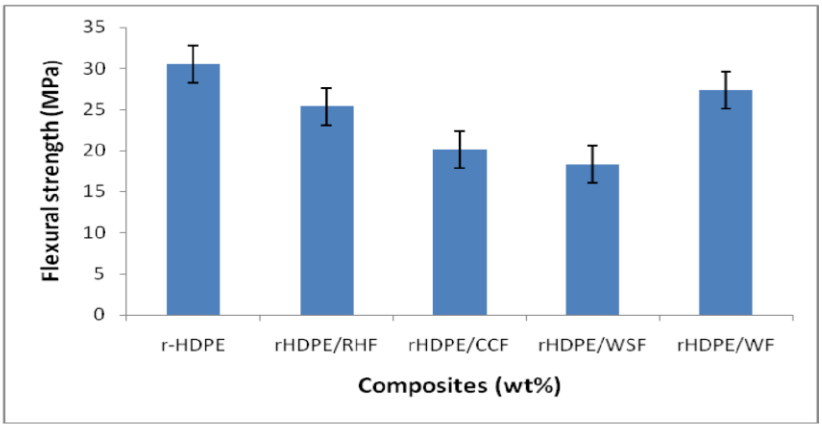

Fig 6: Flexural strength of composites

Figure 6 represents the flexural strength of the composites at filler loading of 65wt%. It was observed that the flexural strength of the four different types of agro filler/rHDPE composites decreased with 65wt% agro-filler compared to the control sample of rHDPE. It was found that the flexural strength of the agro fiber/rHDPE composites were not better than that of the neat rHDPE. The WF composites gave the highest flexural strength of 27.35MPa, followed by RHF composites 25.35GPa, CCF composites 20.1GPa and WSF composite 18.1GPa respectively. This variation in flexural strength could be related to the differences in particle size distribution of the fillers and as well as to the differences in the type of filler used.

Flexural modulus

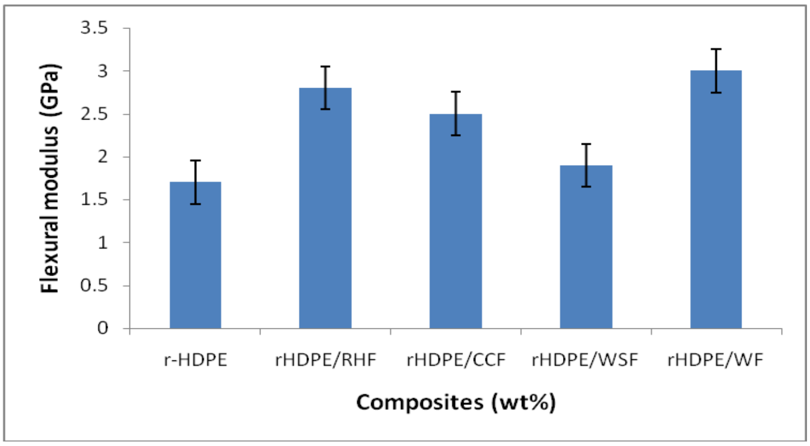

Fig 7: Flexural modulus of composites

Figure 7 shows the flexural modulus of the agro-filler composites with 65wt% filler loading. It was found that the flexural modulus of the composites was better than that of the neat rHDPE. Results showed that the WF composites exhibited the highest flexural modulus of 3.0GPa, followed by the RHF composites 2.75GPa, CCF composites 2.5 GPa, and WSF composite 2.0GPa compared to the control sample of 1.75GPa respectively. The increased flexural modulus of WF composites is due to the lower bulk density and particle size distribution. Increase in bulk density will give rise to weak points which will cause filler accumulation and agglomeration resulting in reduced mechanical strength.

Un-notched Izod Impact strength

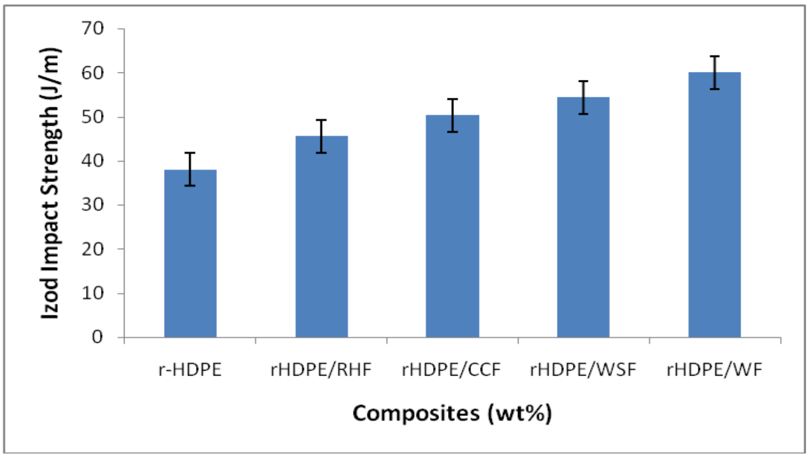

Fig 8: Un-notched Izod Impact Strength of composites

Figure 8 reports the un-notched izod impact strength of the agro-filler composites at 65wt% filler loading. The incorporation of the fillers into the recycled HDPE brought about increment in the impact strength of the composites compared to the neat rHDPE. The WF/rHDPE composites showed superior impact strength of 60 J/m, followed by WSF/rHDPE of 54.4 J/m, CCF/rHDPE 50.3 J/m and RHF/rHDPE composite 45.6 J/m compared to the neat rHDPE of 38 J/m. This is due to bulk density, fiber type and to the differences in the particle size distribution of the agro fibers.

CONCLUSION

Agro fibers are excellent alternative materials, because they are plenteous, widespread, and easily available. Besides, their abundance and renewability, utilization of agro fiber has advantages in economy, environment, and technology. The main objective of this research was to study the potential of agro wastes as reinforcements for thermoplastics. Incorporation of these agro wastes materials at high filler loading resulted in improvements on the flexural and impact properties of the composite system, which varied according to the fiber type. WF-filled composite exhibited superior flexural and impact properties compared to that of the other agro-filler composites due to its lower bulk density and broad particle size distribution.

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