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MIXED AGRO-WASTE FILLER AS REINFORCEMENT: IMPACT ON SOME PROPERTIES OF POLYPROPYLENE

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ABSTRACT

This research studied the impact of mixed sawdust-cowpea agro-waste as filler on the properties of polypropylene (virgin and recycled). Sawdust and cowpea hull powders of mesh sizes 75µm were homogeneously mixed in a 1:1 ratio to form a hybrid filler. Different weight percentages (0, 5, 10, 15, 20) of the hybrid filler were mixed with the respective matrices to produce the composites using the injection moulding technique. The mechanical properties, tensile strength, compressive strength, shear modulus, and hardness test of the composites were studied according to the American Standard Testing and Measurement methods. The mechanical properties showed the greatest improvement in tensile strength compared to PP at 15 wt%, at 34%, and compared to rPP at 10 wt% to 15 wt%, at 38.5%. For all other tested parameters, the PP composite had the greatest improvement at 10wt%; shear strength 35%, compressive strength 10% and hardness 42%, while rPP composites observed 17% shear strength increase, a decrease followed by a steady increase in compressive strength and a decrease in hardness. The solvent imbibition after a seventy-two-hour test duration showed `zero water absorption and less than 0.5% toluene absorption for all composites. The degradation after the 180-day burial test period showed some weight loss, suggesting initiation of degradation. The micrographs of the composites obtained via Scanning Electron Microscope at 10,000 magnification showed that both PP and rPP composites have good interactions with the filler. Hence, agro-wastes such as sawdust and cowpea hulls could be used as fillers in the production of degradable plastics, where strength and a reduced absorption rate are of great importance.

Keywords: Sawdust–cowpea; Polypropylene; Mechanical; Degradation, Morphological properties.

1. Introduction

The desire for improved living conditions has led to a high dependence on polymer products, but these products have adverse effects on humans and the environment. The different polymer materials usually produced and used widely in the world include polyethylene, polypropylene, polyvinylchloride, among others. Polypropylene can be used for applications such as packaging, electrical insulation, and construction products (Shaharuddin et al., 2017). They have a long-lasting negative impact on the environment after disposal, leading to environmental pollution. To mitigate this disturbing effect on the ecosystem, scientists have reverted to using natural, renewable resources, such as biomass, to replace depleting organic materials. Biomass is organic material derived from living organisms (such as plants and animals) and is usually environmentally friendly, renewable, and biodegradable. Examples include: corn cob, rice husk, sawdust, cassava peel, cowpea hull and sugar cane bagasse. The

biomass can be used alone or in combination with other fillers as hybrid fillers in different proportions. Barbosa et al. (2017) analysed the impact and tensile properties of polypropylene, observing that virgin PP, recycled PP, and the blend exhibited similar tensile properties, but recycled PP exhibited lower energy absorption. Thus, they concluded that recycled polypropylene can be used in place of virgin polypropylene in applications that concern tension stress but not impact strength. In addition, studies on the physical and mechanical properties of polypropylene composites reinforced with rice straw lignin were carried out by Karina et al. (2017), who observed that the tensile strength of the composites decreased as the lignin content increased, and there were no significant differences between the physical and mechanical properties of recycled polypropylene and virgin polypropylene. Furthermore, Anosike-Francis et al. (2022) studied the physical-mechanical properties of wood based composite reinforced with recycled polypropylene and cowpea (*Vigna unguiculata Walp*.) husk and obtained results that the physical properties evaluated satisfied the American National Standards Institute (ANSI) A 208.1 standard and confirmation that the 20% cowpea husk (CPH), 80% wood chips (WC) and rPP optimum loading mixture can be used for the production of boards with good dimensional stability. Many studies on polypropylene matrices with different biomasses have been carried out by researchers, but none have examined a hybrid sawdust-cowpea hull biomass. Hence, this study focuses on the impact of a hybrid biomass of sawdust and cowpea hulls on the properties of polypropylene (virgin and recycled) composites. The objectives of the research include: determining the impact of hybrid fillers on the mechanical and imbibition properties of the matrices; ascertaining the degradation rate and relating the microstructural arrangement to the mechanical properties.

2. Materials and Methods

2.1. Sample collection and preparation

Virgin polypropylene (vPP) produced by Exxon Mobil Nigeria and recycled polypropylene (rPP) were used for this study. Sawdust was obtained from Umuokpu Awka Timber Shop, while cowpea was obtained from the family farm at Ezeoye, Nibo, all in the Awka South LGA, Anambra State, Nigeria. The agricultural waste materials were washed to remove impurities and air-dried for 10 days before being crushed using an M6FFC-270 grain grinder and sieved locally and repeatedly with muslin cloth to a fine powder of 75 μm. This treatment method is in accordance with Chris-Okafor et al. (2018a).

2.2. Preparation of composites

The sawdust and cowpea hull powders were mixed in a 1:1 ratio with a hand mixer at room temperature for 10 minutes to form the hybrid filler (SDCO). For 0%, 5%, 10%, 15%, and 20% filler loadings, 200 g, 190 g, 180 g, 170 g, and 160 g of virgin polypropylene (vPP) or recycled polypropylene (rPP) were each mixed with 0 g, 10 g, 20 g, 30 g, and 40 g of SDCO, respectively. The homogeneous mixtures were fed into the hopper of a TU150 200 g injection moulding machine with a circular mould to produce the composites, with an average cycle time of 33 seconds per composite.

2.3. Analysis of mechanical properties of composites

The mechanical properties were measured according to the American Society for Testing and Materials (ASTM) methods.

2.3.1. Tensile Strength

This measures the stretching ability of a material under external loads. The tensile strength of the composites was determined in accordance with the ASTM D-638-14 standard using a Hounsfield Monsanto Tensometer 8889.

2.3.2. Compressive Strength

Compressive strength is the ability of a material to withstand compressive forces or strong pressure. The compressive strength was determined in accordance with ASTM D-695 using the Hounsfield Monsanto Tensometer 8889. The readings were noted, and the values were computed with the formula.

$$\text{Compressive strength } \left(\frac{N}{mm^2}\right) = \frac{\text{maximum force, P(N)}}{\text{cross sectional area, A}(mm^2)} \quad \text{Eqn 2.1}$$

2.3.3. Shear Strength

Shear strength is the measure of the ability of the material to withstand forces that cause its internal structure to slide against itself. It was determined in accordance with ASTM D-732 using the Hounsfield Monsanto Tensometer 8889. The readings were automatically recorded, and the values were computed with the formula;

$$\text{Shear strength} = \frac{\text{shear force, P (N)}}{\text{sheared area, } A_0 \ (mm^2)} \quad \text{Eqn 2.2}$$

A_o = sheared area and it is calculated as $\pi d^2/4$, d is the shear spindle diameter, while π is constant.

2.3.4. Hardness Test

Hardness measures the depth of indentation left on the surface of the composite when an indenter is pressed into it. Surface hardness was measured according to the standard ASTM D2240 using the Shore Scale Durometer Hardness Tester, and the values were automatically measured and recorded.

2.4. Imbibition of Solvents

This test is used to determine the solvent absorption rate of the composites according to ASTM D-570-98 Standard for 72 hours at room temperature.

2.5. Biodegradation Test

This test was conducted to determine the degradation rate of composites in the environment. The soil burial degradation test was used. Composites of known weights were buried 10 cm deep in the soil for six months. The weights were checked every 30 days for 180 days, and the mass reduction of the composites was used to assess degradation.

2.6. Morphological Analysis

Morphological examination was performed using a Scanning Electron Microscope (SEM) model: JEOL-JSM 7600F to determine the compatibility of the matrices and fillers.

3. Results and Discussion

3.1. Mechanical Properties

3.1.1. Tensile Strength

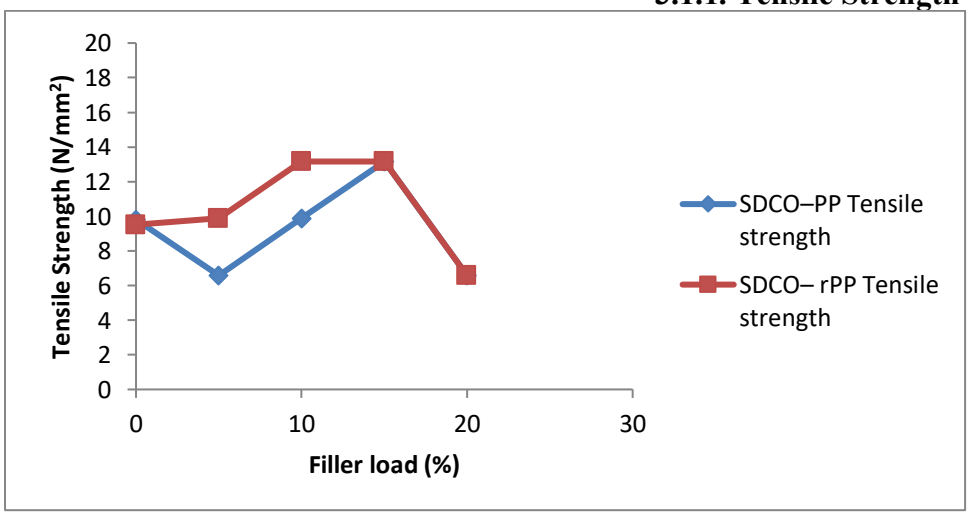

Figure 1: Impact of filler loading on the tensile strength of PP and rPP composites.

In Figure 1, SDCO-PP composites on the addition of fillers showed an initial decrease in the tensile strength before increasing, while SDCO-rPP showed an increase until the maximum point was attained. The SDCO-PP matrices observed a maximum improvement of approximately 34% increase at 15wt% filler load, while the SDCO-rPP matrices observed same improvement of about 38.5% at 10wt % and 15wt% filler load. The initial decrease observed in PP composites could be due to insufficient filler proportion in the composites or weakening of the interfacial attraction of the constituent composition, while the increase could be attributed to the cellulosic nature of the fillers and adhesion between filler- matrix interface which led to better stress transfer from the matrix to the filler at the optimum filler. vPP and rPP have been reported to exhibit comparable tensile properties; therefore, rPP can substitute vPP in applications subjected to tensile stress (Ogudo et al., 2021; Barbosa et al., 2017; Karina et al., 2017), which aligns with the findings of this study.

3.1.2. Percentage of elongation at break

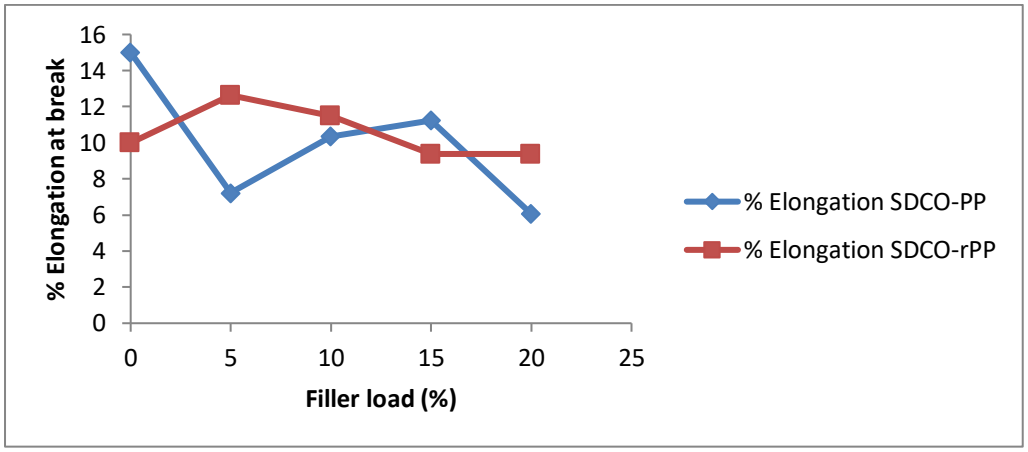

Figure 2: Impact of filler loading on the percentage elongation at break of PP and rPP composites

From Figure 2, the percentage of elongation at the break of the rPP initially increased before decreasing as the filler load increased. This decrease indicates that the filler increased the matrix's rigidity and reduced its ductility, thereby producing a harder, more brittle material. The PP composites showed a decrease in percentage elongation at break across the entire filler loading range, indicating improved rigidity and reduced ductility. This agrees with Chris-Okafor et al. (2018) and Nwokoye et al. (2024b), where a decrease in the percentage elongation at break was observed with increasing filler loading due to the stiffening and hardening of the composite, which reduced its resilience and toughness.

3.1.3 Compressive Strength

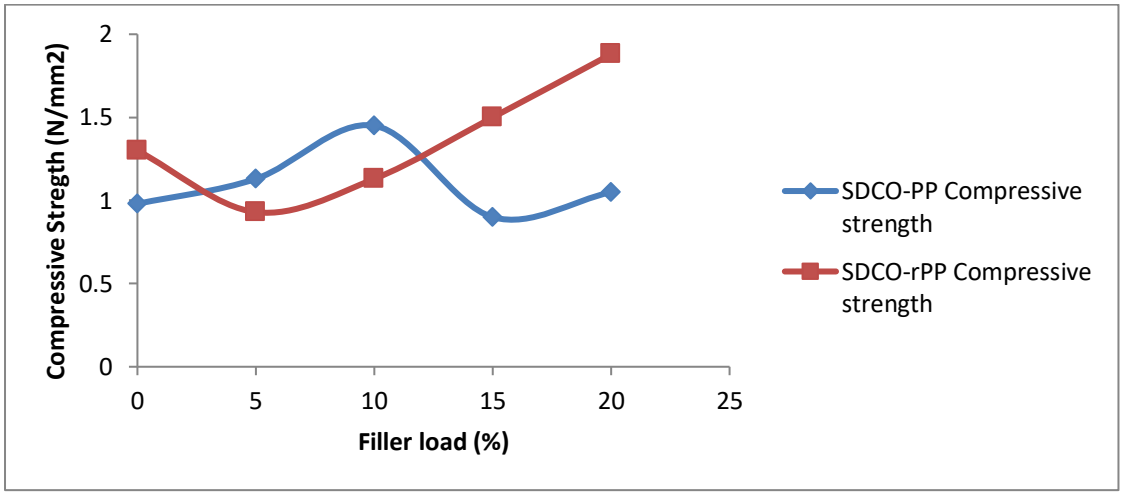

Figure 3: Impact of filler loading on the compressive strength of PP and rPP composites

Figure 3 shows that the compressive strength of the PP composites increased in a fluctuating manner, with a maximum increase of 48% at a filler load of 10 wt%. According to the work of Chris-Okafor et al. (2017), where an increase in compressive strength was observed, but in an irregular manner, the compression depends on the interaction of the matrix and the filler and not on filler load quantity. On the other hand, the rPP composites showed an initial 28% decrease before a continuous increase upon the addition of filler. The decrease could be attributed to insufficiency of the filler leading to poor reinforcement, while the increase in the composite's compressive strength could be attributed to the filler's reinforcing characteristics, which improved the composite's mechanical properties due to its cellulosic nature, which is in agreement with the works of Ogudo et al. (2021) and Nwokoye et al. (2024).

3.1.4. Shear Strength

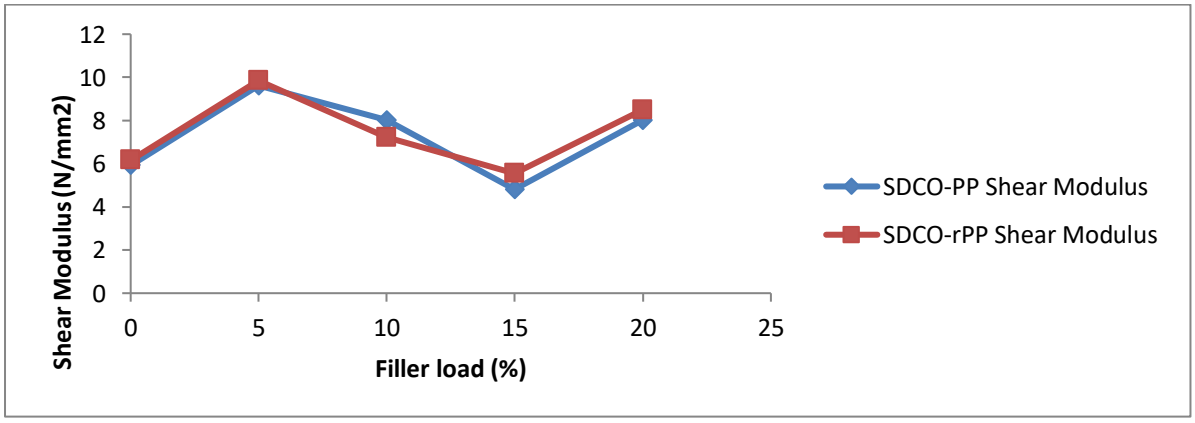

Figure 4: Impact of filler loading on the shear strength of PP and rPP composites

In Figure 4, shear strength generally increases for all composites except at 15 wt% filler for both PP and rPP. At 5 wt% filler, maximum increases of 62.5% (PP) and 60% (rPP) were obtained, indicating that the hybrid filler increases rigidity, so a greater force is needed to deform the composites in the loading direction. This agrees with Ogudo et al. (2021) and Nwokoye et al. (2024b). At 15 wt% filler, PP and rPP show decreases of 18.75% and 10%, respectively, indicating reduced rigidity.

3.1.5. Hardness

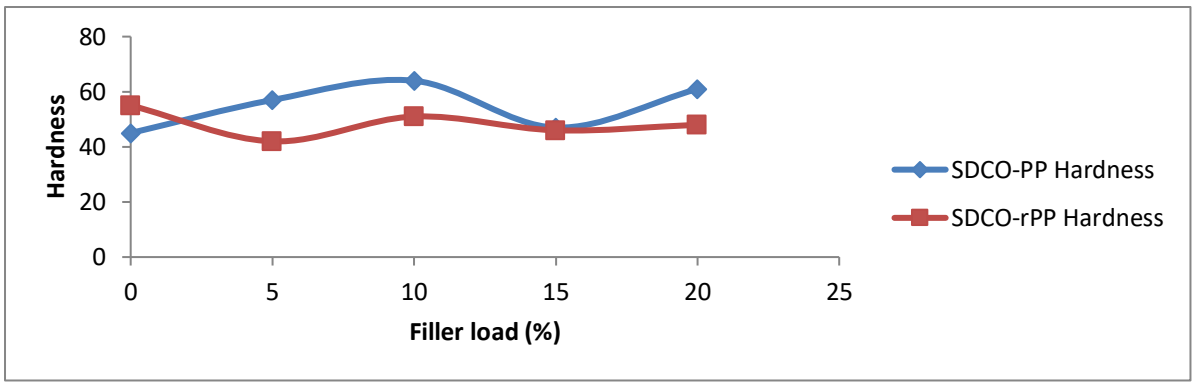

Figure 5: Impact of filler loading on the hardness of PP and rPP composites

Figure 5 reflects an overall increase in hardness as the filler load increases for all PP composites, with an optimum hardness of 29.7% at 10wt% filler load. The hybrid filler improved the hardness of the composites by reducing the elasticity and increasing the surface resistance to indentation. This aligns with the findings of Chris-Okafor et al. (2018), Ogudo et al. (2021), and Nwokoye et al. (2024), who reported increased hardness with higher filler loads. In contrast, the SDCO-rPP composites showed an overall decrease, with a minimum of -23.6% at 5 wt% filler load, indicating that the blend did not enhance the stiffness of the rPP matrix.

3.2. Study of solvent imbibition

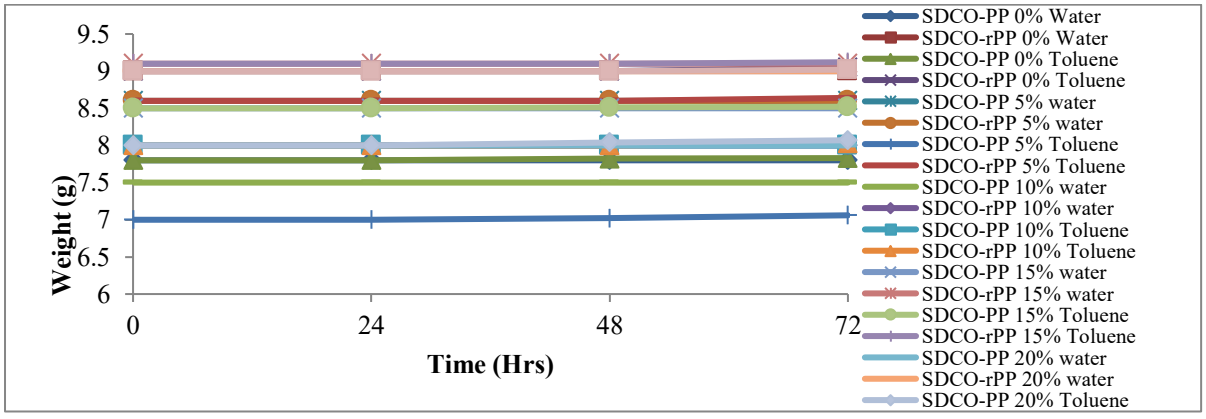

Figure 6: Impact of filler loading on the solvent imbibition properties of PP and rPP composites.

Figure 6 shows no weight change for any composite after 72 hours in room-temperature water. After 72 hours in toluene, the weight gain was only 0.2–0.5%. This minimal uptake is likely due to the non-polar, hydrophobic nature of the polymer matrix and lignin in the filler, the low filler loading, or temperature effects. These findings agree with Ogudo et al. (2021) and Nwokoye et al. (2024) and indicate the composites are suitable for applications requiring water resistance.

3.3. Biodegradation Test

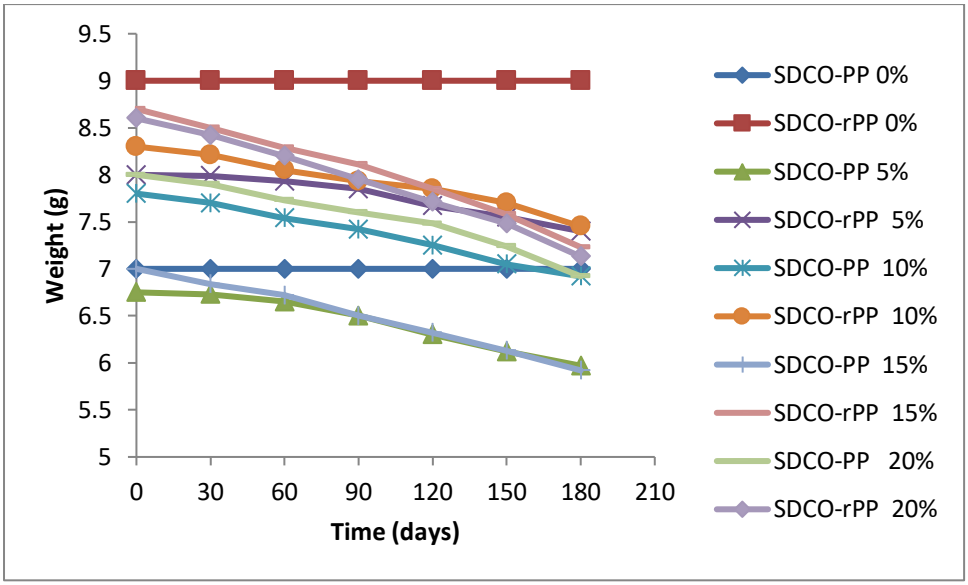

Figure 7: Impact of filler load on the degradation rate of PP and rPP composites.

Figure 7 shows that there were no weight reductions for the composites without fillers, i.e., 0% SDCO-PP and 0% SDCO-rPP, during the 180-day test period. Other composites with biomass fillers exhibited weight loss after a 30-day test period, with the loss continuing as burial time increased. At the end of the 180-day test period, the composites SDCO-PP and SDCO-rPP at the 5wt% filler load had -11.6% and -7.5% weight loss, respectively, while at the 20wt% filler load they had -13.5% and -17.1% weight loss, respectively. Thus, degradation, as indicated by weight loss, increases with filler load for all filler-containing composites. This observation is in line with the research findings of Chamas et al. (2020); Ogudo et al. (2021); Nwokoye et al. (2024) and Nwokoye et al. (2024b). These authors observed that degradation of composites improves with increasing filler content and degradation time under favourable conditions.

3.4. Microcrystalline Analysis

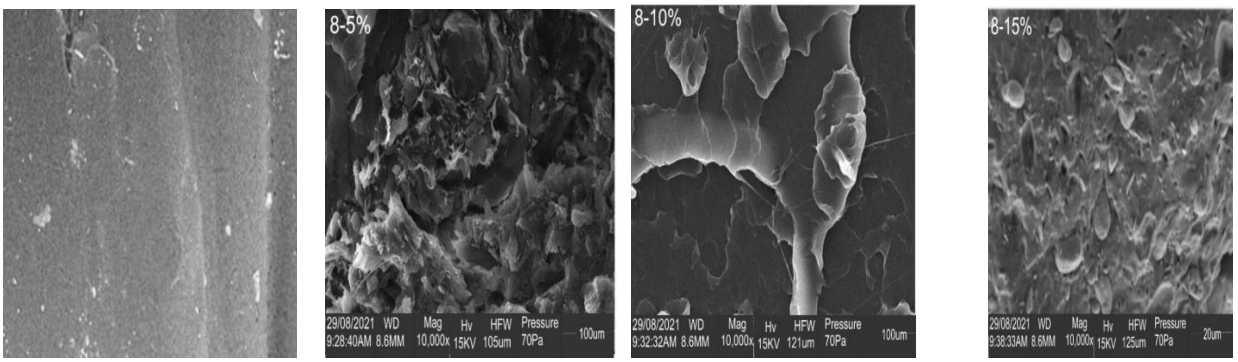

Fig. 8a(i) 100% PP (ii) 5% SDCO – PP (iii) 10% SDCO-PP (iv) 15% SDCO-PP

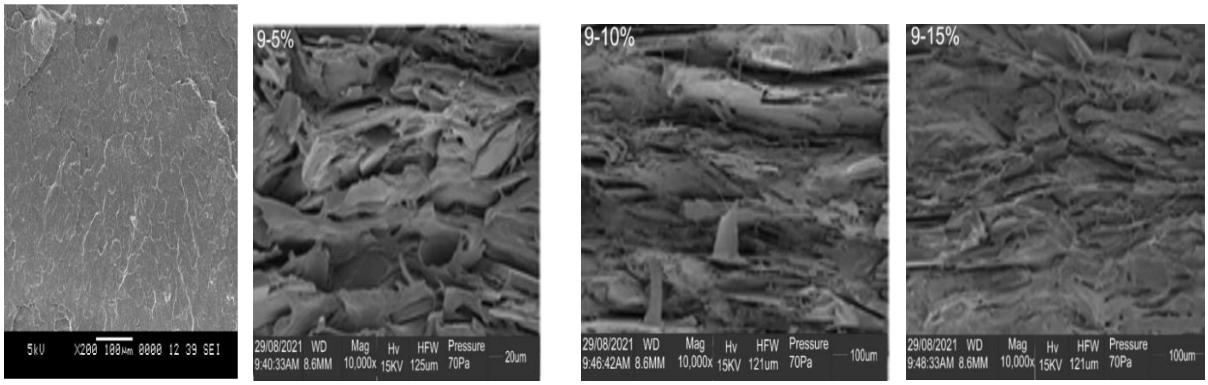

Fig. 8b: (i) 100% rPP (ii) 5% SDCO –rPP (iii) 10% SDCO- rPP (iv) 15% SDCO– rPP

Figure 8ai showed the morphology of PP as a homogeneous surface with cleavages, while Figures 8aii to iv showed the morphology of the composites (SDCO-PP) as the filler load increased from 5wt% to 15wt%. From the Figures above, at 5 wt% filler loading, there was interaction between the filler and the matrix that was not too strong, thus resulting in the presence of voids. As the filler load increased to 10 wt%, compatibility increased, with observed cleavages and a dark surface. The observed cleavage indicated stronger intermolecular bonding that can resist deformation under stress. At 15wt% filler load, tiny stony agglomerates with few voids were obtained, indicating collapse of the interaction between the filler and the matrix. Fig. 8bi showed the morphology of rPP as a homogeneous surface with cleavages, while Fig. 8bii to iv showed the morphology of the composites as the filler load increased from 5wt% to 15wt%. From the Figures, the filler was consistently and uniformly distributed within the matrix, and there was the presence of large folds with breaks at 5wt% filler loading, which means that the interfacial bonding between the filler and the matrix polymer was strong, but due to unsaturation of the matrix by the filler, the breaks occurred. As the filler loading increased to 10 wt%, the compatibility between the filler and the matrix improved, yielding a better structure with stronger bonding and fewer breaks. The best morphology, with a near-homogeneous structure and no apparent phase separation, was obtained at 15 wt%. Thus, the SDCO hybrid filler showed great interaction with the matrices, as observed with filler dispersion, leading to good filler-matrix compatibility. The SDCO-PP and SDCO-rPP composites that showed higher tensile strength at 10 wt% and 15 wt%,

respectively, were due to the filler particles being well dispersed in the polymer matrix. Chan et al. (2016) and Jumaidin et al. (2017) stated that when filler particles were well dispersed in the polymer matrix, there was excellent adhesion between them, leading to increased interfacial strength and, thus, higher tensile strength. This result aligns with the authors' report.

4. Conclusions

Sawdust and cowpea hull hybrid fillers were successfully incorporated into virgin polypropylene (PP) and recycled polypropylene (rPP) to produce composites. The fillers increased tensile strength, compressive strength, and shear modulus for both PP and rPP at different loadings. Hardness increased in PP but generally decreased in rPP. Elongation at break decreased at all filler loadings for both matrices.

After 72 hours, all composites were water resistant and showed only 0.2–0.5% toluene uptake, indicating suitability for wet environments. A 180-day soil burial test showed some weight loss, suggesting the onset of degradation. Micrographs at 10,000× revealed that SDCO-PP (10 wt%) and SDCO-rPP (15 wt%) had the best filler–matrix interaction, consistent with their higher tensile strength.

The properties were influenced by the synergistic effect of the hybrid fillers, particle size, and filler dispersion. Overall, agro-wastes such as sawdust and cowpea hulls are viable, low-cost, eco-friendly, and readily available fillers for degradable plastics like packaging materials.

Recommendations

1. A study on the use of sawdust-cowpea hull hybrid filler with other matrices is recommended.
2. Also, a study on varying the percentage composition of the respective fillers during hybrid formation is recommended to determine the most reactive and best biomass.

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