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EVALUATION OF PHOTODEGRADATION BEHAVIOR OF POLYPROPYLENE/GRAPHITE PARTICULATE COMPOSITE FOR ROOFING APPLICATION

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ABSTRACT

Polypropylene employed in plastic roofing sheets production is vulnerable to UV light irradiation, causing it to degrade. This study investigates the effect of graphite (Foreign and Nigerian) as fillers and UV stabilizers on the mechanical properties of Polypropylene subjected to UV irradiation. Polypropylene composite was prepared with varying proportions of graphite (10% to 60%). The hardness, impact strength, and tensile strength were tested before and after 48 hours of UV exposure. The results indicate that tensile strength generally decreases with increasing filler loading and UV exposure-time, though the tensile strength of Polypropylene/FGP composites at 20% filler-loading was insignificantly affected by the UV-light with its tensile-strength slightly dropping from 32.71 MPa to 31.88 MPa. Tensile strength of Polypropylene/NGP composites at 40% slightly dropped from 41.78 MPa to 37.42 MPa, likewise that of Polypropylene/NGP composites at a 90/10 proportion dropped more (33.77 MPa to 26.62 MPa) compared with that of its counterpart. The Impact strength increases with an increase in the filler-loading up to 20%, and at 30% of the filler, the materials gained more resistance to degradation by UV light. Hardness results showed that both foreign and local graphite fillers increased the material's hardness, with PP/FGP exhibiting more consistent performance after UV exposure. The hardness value of the materials produced with 10% of the filler slightly dropped from 12.3 HRF to 11.85 after exposure. In conclusion, the composites produced offer superior resistance to degradation by UV light pertinent to Impact strength, tensile strength and Hardness of the composite compared with Polypropylene.

Keywords: UV-light, Degradation, Polypropylene, Composite

INTRODUCTION

In today's world of contemporary research, composite materials have become a major focus due to their advantageous physical and chemical properties. These materials have many applications in various industries, ranging from construction (buildings and bridges) to automotive (car bodies), aeronautics (where materials with excellent strength-to-weight ratios are required), and medical industries (Bhong *et al*., 2023). Composites are widely utilized due to their adaptability to different environments and ability to be easily integrated with other materials to meet specific functional requirements and desired properties. These materials are obtained by combining two or more constituents, leading to a novel material with unique physicochemical properties different from those of monolithic materials (Chinta, 2017). Polymer composites are materials that combine polymers with inorganic or organic additives that have specific geometries. When a filler/reinforcement of good mechanical properties (flexural strength, tensile strength, impact strength, and modulus of elasticity as the case may be) is embedded into the matrix of polymers, a composite material with improved and/or unique properties is possible (Hsissou, *et al.*, 2021). The nature of the matrix and the filler, the shape and proportion of the filler, the strength of the interfacial bond between the matrix and filler, and the method of production adopted are all parameters that significantly affect the properties of a resulting composite (Hsissou, *et al.*, 2021). The advantage of composites with a polymer matrix compared to metals is the manufacturing process which allows for the production of complex shapes along with a high strength-to-weight ratio, hence the low fuel consumption (in aviation and automotive industries), high speed in competitive sport or long-range for missiles and high payload (in Transportation) (Hsissou, *et al.*, 2021).

Particle-reinforced plastic composites (PRPCs) incorporate fillers; discrete particles to enhance or modify the properties of the matrix and/or replace some of the matrix volume with a more economical material. PRPCs are widely used in various fields, including construction, packaging, automotive tires, and medical applications. Evaluating the effective properties of these composites is a critical challenge in many engineering contexts (Danilaey *et al*., 2024). The properties of the particle-reinforced polymer are affected by the particle size, shape, characteristics, and spatial distribution of the reinforcements (Li and Li, 2025).

Severe exposure to UV light can accelerate the degradation of polymeric materials. This is because UV radiation is capable of causing photooxidative aging, which results in the breakage of polymer chains, the production of free radicals, and the reduction in the molecular weight of polymers. It also results in a loss of surface gloss and the significant deterioration of many material properties with time (Lu *et al.*, 2018).

In Polymeric Composites, Photodegradation initiates with the absorption of UV photons by chromophores i.e. hydroperoxides, catalyst residues, carbonyls, and unsaturated molecules containing double and triple bonds, and/or rings (Lu *et al.*, 2017). The activation processes initiated by UV photons excite states in macromolecules which leads to surface discoloration, yellowing, and a loss of surface gloss (Lu *et al.*, 2017). Further exposure to UV light results in the formation of a thin layer consisting of loosely adherent particles called chalking. Depending on the type of polymer, flaking of surface resin, pitting and microcracking may also occur. In addition, chemical aging such as chain scission by UV will result in a loss of low molecular weight or highly volatile products, which can vaporize very quickly at elevated temperatures. (Lu *et al.*, 2018).

Based on the literature survey, Nigerian graphite has not been utilized appropriately in composite formulations, especially in polymer matrix composites (PMCs). Meanwhile, plastic materials used for outdoor applications such as house roofing sheets are vulnerable to UV degradation from sunlight and other weathering conditions, which significantly reduces their lifespan. Thus, there is a need to enhance polymers' resistance to photodegradation to improve the durability and performance of these materials.

MATERIALS AND METHODS

Mold Preparation

The mold was constructed according to the required dimension of the material. For this work, a square mold of 140 mm x 120 mm x 3.2 mm dimension was adopted. The mold was produced with the use of a 3 mm heavy gauge iron sheet so that it would not be affected by the high temperature during composite production. Hence, the effect of mold bending was eliminated.

Preparation of Composite

Two different graphite (local and foreign) were utilized as reinforcements in the composite production. The Nigerian Locally sourced graphite and foreign graphite were blended in a two-roll mill with Polypropylene to produce Polypropylene/local graphite and Polypropylene/Foreign graphite composites respectively. The composites were produced by mixing the Polypropylene and the respective fillers using a two-roll mill at a temperature of about 190ºC for five minutes with the rolls of the two-roll mill machine in counterclockwise motion at a speed of 45rpm. The Polypropylene was first melted to allow for adequate flow of the molten polymer before pouring the respective filler (graphite). Upon achieving a band and bank formation of the polymer on the front roll, the prepared fillers were introduced gradually to the bank, crossed, and allowed to mix for 5 minutes for homogeneity. The composite was sheeted out and labeled accordingly. The respective mixtures obtained from the mill were then placed into a metal mold of dimensions 140 mm x 120 mm x 3.2 mm and afterward placed on a hydraulic hot press (Compression Molding Machine) for shaping at a temperature of 160ºC and pressure of 2.5 MPa for 5mins. The respective mixture was then cooled, removed from the mold, and labeled accordingly. The NGP and FGP fillers were utilized at a percent weight ratio ranging from 10%-60% of the total weight.

The composites formulation is shown in Table 1 containing six specimens for each sample (FGP1, FGP2, FGP3, FGP4, FGP5, FGP6 and NGP1, NGP2, NGP3, NGP4, NGP5, NGP6)

Table 1: Composite Formulation

Samples	Fillers		Polypropylene (%)
	Processed Nigerian Graphite (NGP) (%)	Foreign Graphite Powder (FGP) (%)	
Control	0	0	100
NGP1/FGP1	10	10	90
NGP2/FGP2	20	20	80
NGP3/FGP3	30	30	70
NGP4/FGP4	40	40	60
NGP5/FGP5	50	50	50
NGP6/FGP6	60	60	40

UV Light Aging Test procedure

This tester can isolate 18 pieces of standard panels (size 150x70mm). During the test, the 12 samples (FGP1, FGP2, FGP3, FGP4, FGP5, FGP6, and NGP1, NGP2, NGP3, NGP4, NGP5, NGP6) are installed in a column form rotating sample track. They are fixed with tension rings, the sample rack rotates uniformly and ensures every sample gets the same irradiance energy, increasing the comparability and repeatability of testing results. The water inlet pipe was connected to the external water supply (water transfer into the cabinet aims to spray and heat water), while the outlet pipe is also connected to the outdoor. When the machine is turned ON, it automatically controls the inlet and outlet of the water. The workroom door was closed and rotated 180 degrees to fasten.

The machine was turned ON by rotating the red power switch to ‘‘1’’, the objective is to simulate the natural environment for 48 hours by subjecting the samples to the following set of parameters. For the first 24 hours, the test samples were exposed to UV light accelerated aging at a set temperature of 37 ºC as the average/normal day temperature experienced by outdoor materials with a spraying time of 2 minutes after every 2 hours. Then, for the second 24 hours the test samples were exposed to UV light accelerated aging at a set temperature of 50 ºC to simulate extreme weather conditions just like in the hot season and the spraying interval was extended to 4 hours. An accumulated time of 48 hours on exposure to UV light accelerated aging was achieved on the samples, the 12 UV-exposed samples were then carried to the material testing lab to investigate the effect of photodegradation on the mechanical properties of the Polypropylene/NGP and Polypropylene/FGP composites.

Determination of Mechanical Properties of the Prepared Polypropylene Composites

Tensile Test

Tensile strength is the maximum stress a material can withstand before fracturing when it is subjected to uniaxial (stretching) loading. The tensile strength of the materials was determined before and after exposure to the UV light using a Universal Testing Machine. The test was carried out following ASTM D-638 standard. A dumbbell-shaped samples with dimensions 100 mm x 15 mm x 3 mm and a gauge length of 40mm were subjected to uniaxial (stretching) loading. Tensile modulus and percentage elongation at break for each of the samples were calculated and recorded automatically by the machine.

Impact Test

The impact strength of materials is directly related to the overall toughness, defined as the material's ability to absorb energy before fracture. The impact test was carried out based on the ASTM D-156 standard. The samples were cut into 80 mm x 13 mm x 3 mm dimensions with 45º notches. The test was carried out using an Izod Impact Testing machine. The samples were individually clamped vertically on the jaw of the machine, and a hammer of weight 1500 N was released from an inclined angle of 150°. The impact energy for the

corresponding tested sample was taken and recorded. The impact strength was calculated and recorded accordingly using equations 1 and 2

Average Impact Energy = (J) (1)

Impact Strength = Average Impact Energy / (width x thickness) mm (2)

Where Sample thickness = 3.0 mm, Width 13 mm

Hardness Test

Hardness is the resistance of materials to indentation or plastic deformation. The hardness test measures the depth of penetration of an indentor. The test was conducted using a digital Rockwell INDECTEC testing machine HRF (model8187.5LKB), using an F scale with a steel ball indentor of 1/16 inch. A minor load of 10kgf was first applied to the respective samples followed by a major load of 60kgf. The hardness was determined by the penetration of the indentor onto the respective samples for 5 seconds while the hardness value was read from the digital scale. Three readings were taken for each sample with the average value taken and recorded as the hardness value of the respective sample.

RESULTS AND DISCUSSION

Tensile Test

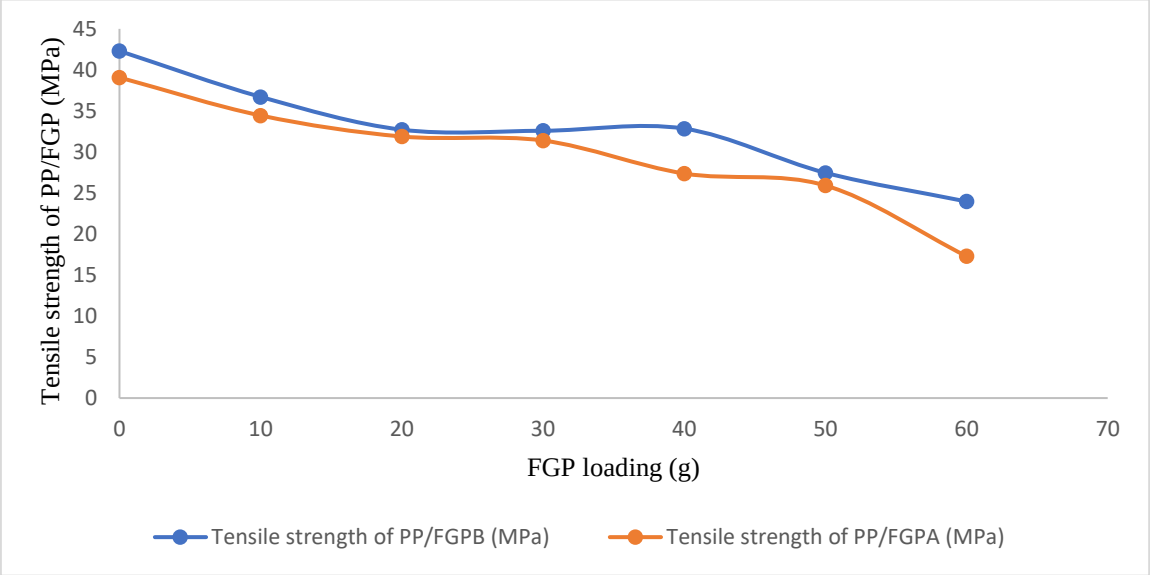

Figure 1: A graph of tensile strength against the % composition of the foreign graphite composites before and after UV exposure for 48 hours

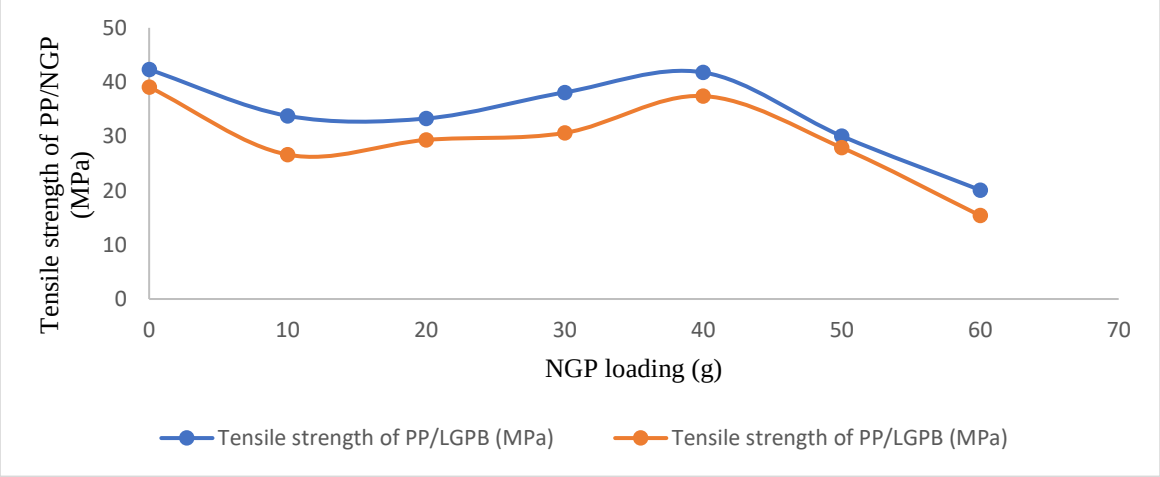

Figure 2: A graph of tensile strength against the % composition of the Nigerian graphite composite before and after UV exposure for 48 hours

Figure 1-2 shows that; the tensile strength generally decreases with increasing filler loading and UV exposure time, though the tensile strength (Figure 1) of Polypropylene/FGP composites at 20% filler loading was insignificantly affected by the UV light with its tensile strength slightly dropping from 32.71 MPa to 31.88 MPa, and this was maintained up to 30% filler loading beyond which it dropped. The tensile strength (Figure 2) of Polypropylene/NGP composites at 40% slightly dropped from 41.78 MPa to 37.42 MPa. The tensile strength (Figure 1) of Polypropylene/NGP composites at a 10/90 proportion dropped more (33.77 MPa to 26.62 MPa) compared with that of the Polypropylene/FGP composites (Figure 1) at the same filler proportion of 10/90, dropping from 36.71 MPa to 34.44 MPa. This suggests that both the foreign and Nigerian graphite-reinforced polypropylene composites are affected by the UV light, however, the Nigerian graphite-reinforced composite is relatively more

affected by the UV light. A similar observation was made by (Azeem *et al.*, 2022).

The irradiated UV light led to PP chain scission thus, making the composite brittle and reducing its tensile strength. For the sample with 40/60 composition, a notable observation is the improvement in tensile strength (Figure 2) for the NGP composite 41.78 MPa before UV irradiation) compared to the foreign (Figure 1) graphite sample (32.84 MPa). After UV exposure, the NGP maintains relatively higher tensile strength (Figure 1) at 37.42 MPa, while the foreign graphite (Figure 1) experiences a significant drop to 27.37 MPa. This could suggest that Nigerian graphite provides better UV resistance at this composition. Sample 50/50 of foreign and Nigerian graphite samples show a decrease in tensile strength (Figure 1-2) with increasing filler loading. The foreign graphite composite (Figure 1) sees a reduction from 27.46 MPa to 25.91 MPa after UV irradiation, while the processed Nigerian graphite composite (Figure 1) drops from 30.04 MPa to 27.91 MPa, maintaining slightly better post-UV performance. Furthermore, in the composite filled with 60g of both FGP and NGP, the tensile strength significantly decreased, with the foreign graphite (Figure 1) dropping from 23.96 MPa to 17.29 MPa, and the Nigerian graphite (Figure 1) dropping from 20.09 MPa to 15.38 MPa after UV irradiation this could be as a result of poor stress transfer between the PP and either of the two graphite powder used as the reinforcing agent. It could be suggested here, that a higher filler loading weakens the composite's tensile properties, and UV exposure impairs this effect similar trend was reported by (Ghasemi and Farshchi 2020).

Impact Test

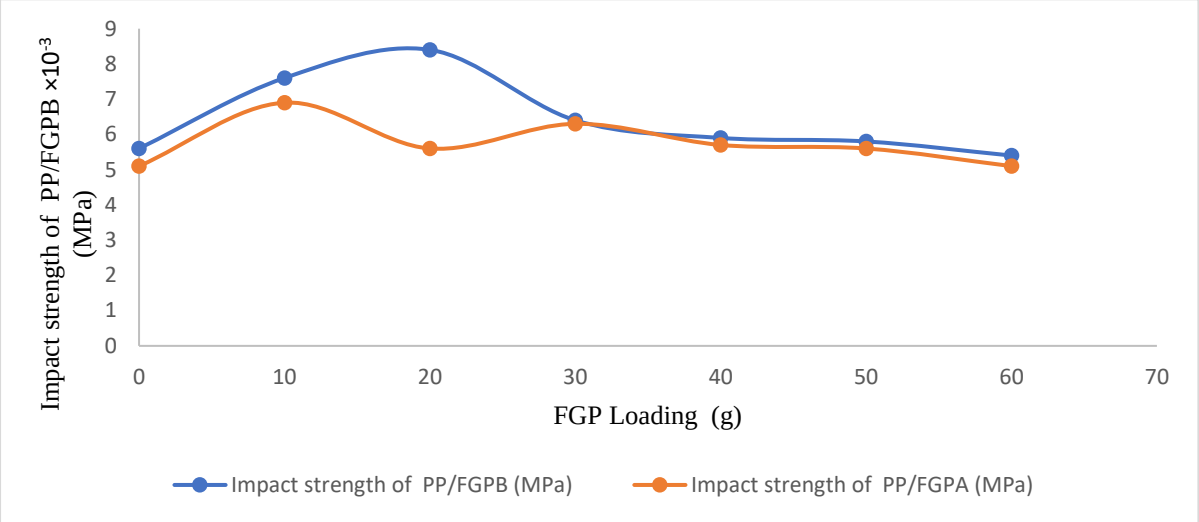

Figure 3: A graph of Impact strength against the % composition of the Foreign graphite composite before and after UV exposure for 48 hours

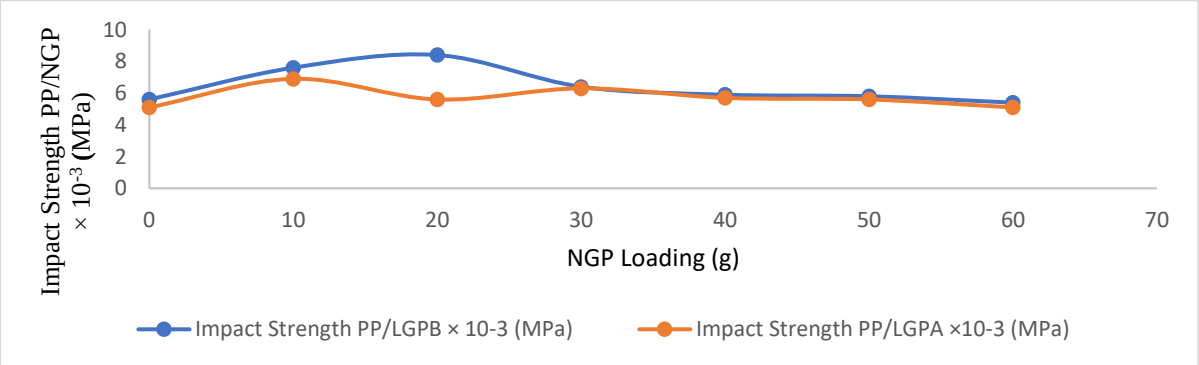

Figure 4: A graph of Impact strength against the % composition of the Nigerian graphite composite before and after UV exposure for 48 hours

The impact strength of polypropylene (PP) reinforced with foreign graphite (FGP) and Nigerian graphite (NGP) before and after 48 hours of UV light exposure was presented in Figure 3 – 4 accordingly. Impact strength measures a material's ability to resist a sudden force or shock without rupturing, this reflects the material's toughness.

The impact strength (Figures 3 & 4) was observed to be increasing with filler loading. That is, as the proportion of graphite (both foreign and Nigerian) increases. The impact strength of the composite (Figures 3 & 4) initially rises until it reaches it peak at 80/20 (PP/Graphite) composition and then drops with a further increase in filler. For instance, in the case of PP/FGP_b, the impact strength (Figure 3) increases from 5.6 MPa to 7.9 MPa at 80/20 (80 g PP/20 g FGP_B) this is before exposing the samples to the UV light irradiation. Correspondingly, for PP/NGP_B (Figure 4), the trend is similar, with the impact strength rising from 5.6 MPa at 100/0 (control) to 8.4 MPa at 80/20 (80 g PP/20 g NGP_B), a similar result was reported by (Khairul, *et al.*, 2016).

The increase in impact strength with filler addition could be attributed to the reinforcement effect which absorbs and distributes the applied impact energy more efficiently. This may be due to the good interfacial interaction between the polypropylene (PP) and the graphite (FGP or NGP). This

could also be a result of the stress transfer mechanism at the interface of the matrix (PP) and filler. At moderate filler loading (20-30), the graphite particles are well-dispersed within the polypropylene matrix, leading to better energy absorption and impact resistance parallel results reported by (Masudur Rahman and Khan, 2018).

Decrease in Impact Strength beyond 70/30 composite composition (such as 60/40), the impact strength decreases for both types of graphite fillers (foreign and Nigerian graphite). For example, PP/FGP_B (Figure 3) decreases from 7.9 MPa at 80/20 to 6.8 MPa at 60/40 and PP/NGP_B (Figure 4) decreases from 8.4 MPa at 80/20 to 6.5 MPa at 60/40 composites. Further, at higher FGP or NGP loadings, graphite particles tend to agglomerate, which leads to non-uniform dispersion. These agglomerates act as stress concentrators, making the material more prone to brittle failure under impact stress equivalent observation was made by (Haijun *et al.,* 2023). The polypropylene matrix also becomes more restricted in its ability to deform, which reduces its toughness (Haijun *et al.,* 2023). As a result, the composite becomes less effective at absorbing and dissipating impact energy. Comparison between FGP and NGP, Nigerian graphite composites (PP/NGP_B) (Figure 4) shows slightly higher impact strength than foreign graphite composites (PP/FGP_B) (Figures 3) before UV light exposure of the test samples, particularly at higher filler loadings. This observation could be due to geomorphological differences of the individual graphite deposits. Alternatively, it might suggest that Nigerian graphite particles have a more favorable surface area or bonding characteristics with the polypropylene matrix compared to the foreign graphite filler.

After exposing the prepared composites at different FGP and NGP loadings to UV light for 48 hours, It was observed that the UV light exposure leads to photo-oxidation of the polypropylene matrix, breaking polymer chains and forming brittle regions. This results in a loss of flexibility and toughness, leading to a decrease in the impact strength of the samples (Figures 3 & 4) from tough and hard to soft and brittle, a similar observation was reported by (Zhao, 2021). On the other hand, PP/FGP_A (80/20) (Figure 3) decreases from 7.9 MPa (before UV light) to 6.4 MPa (after UV light irradiation) while PP/NGP_A (80/20) (Figure 4) decreases from 8.4 MPa (before UV-light irradiation) to 6.9 MPa (after UV-light irradiation). This could be attributed to the UV light radiation causing polypropylene chain scission and embrittlement of the polypropylene matrix, which may reduce its ability to absorb high-impact energy and dissipate impact energy. It could be established here that, Graphite fillers help to slow down the degradation of polypropylene with and without UV light radiation. Thus, these results explain why composites with moderate graphite content retain more of their impact strength after UV exposure compared to those with higher or lower of both the foreign and Nigerian graphite filler content the corresponding result was reported by (Haijun *et al.,* 2023).

Moreover, at the 60/40 composition, the impact strength of PP/FGP_A (Figure 3) decreases from 6.8 MPa (before UV light exposure) to 6.1 MPa (after UV light exposure), a slight reduction compared to the virgin polypropylene (100/0), which also drops from 5.6 MPa to 5.1 MPa. This observation agreed with the results reported by (Boubakri *et al.,* 2010; Wypych, G. 2015) which established that UV radiation degrades polymers by breaking down their molecular structure, leading to a decrease in some of their mechanical properties like impact strength. Unlike the composites containing both the foreign and locally soured graphite which absorbed impact energy and dissipated UV radiation, thereby protecting the polypropylene matrix from extensive degradation. This leads to a better retention of impact strength in the composites containing moderate amounts of graphite. Foreign graphite (FGP) composite (Figure 3) generally shows less reduction in impact strength after UV exposure compared to Nigerian graphite (NGP) composites (Figure 4).

Comparing the results of the impact strength of the virgin PP, PP/FGP PP/NGP before and after the UV-light irradiation, there was increase in the impact strength from the PP to PP/FGP and to PP/NGP (Figure 4) composites, for instance, PP/FGP_a (80/20) (Figure 3) decreases from 7.9 MPa to 6.4 MPa, whereas PP/NGP_a (80/20) (Figure 3) decreases from 8.4 MPa to 6.9 MPa, Foreign graphite might have a better UV shielding effect, possibly due to its superior purity or more uniform dispersion within the matrix, which allows it to protect the polymer better from UV-induced degradation.

Hardness Test

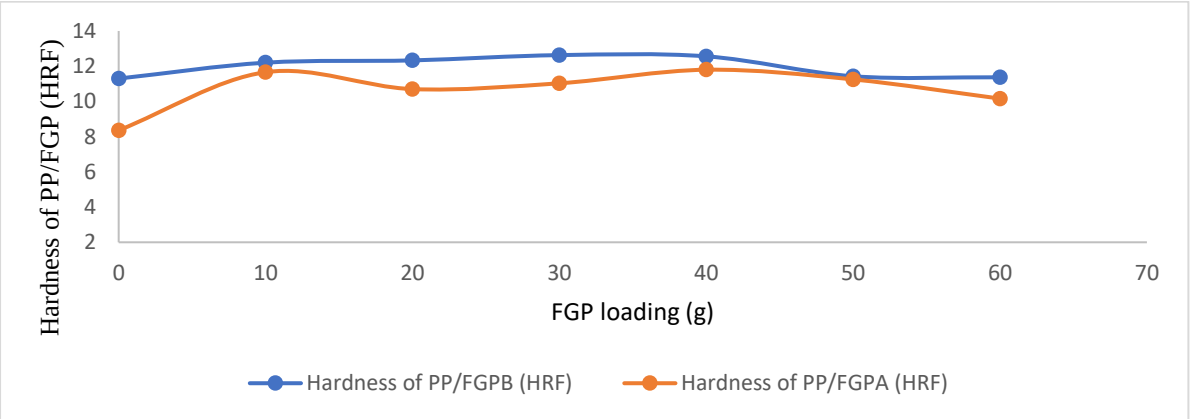

Figure 5: A graph of Hardness against the respective % composition of the Foreign graphite composite before and after UV exposure for 48 hours

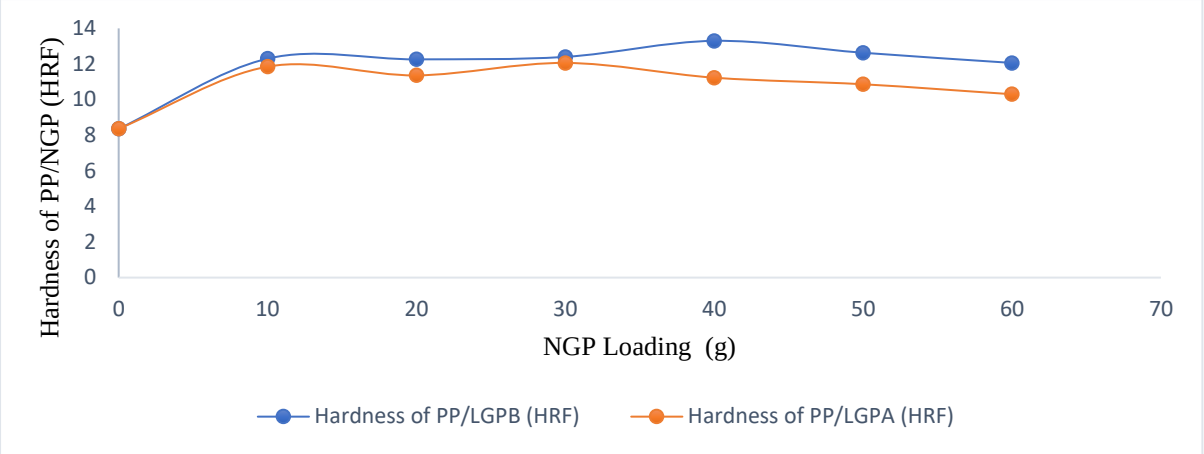

Figure 6: A graph of Hardness against the respective % composition of the Nigerian graphite composite before and after UV exposure for 48 hours

Figure 5 – 6 above illustrates the result of the hardness test with the respective hardness values of polypropylene (PP) and its composites before and after exposure to UV light. The composite with 100 % virgin polypropylene (100/0) has the lowest hardness value, particularly before UV exposure with hardness values of 11.30 HRF and 8.36 HRF for PP/FGP_B (Figure 5) and PP/FGP_A (Figure 6) respectively. This is expected, as unfilled polymers typically have low mechanical properties compared to filled composites (Cheng *et al.*, 2020).

As the proportion of the foreign graphite increases, there is a noticeable improvement in hardness value. For instance, the 90/10 composition shows a significant increase in hardness to 12.20 HRF for PP/FGP_B (Figure 5), indicating that the FGP improves the load-bearing capacity of the polypropylene matrix. Furthermore, for the composite filled with the Nigerian graphite (NGP) (Figure 6) a similar trend was observed with an increase in hardness from 8.36 HRF for pure PP to 13.30 HRF in the 60/40 composition (PP/NGP_B), illustrating the effect of the filler material. However, the effect of FGP seems to be more consistent across the compositions compared to NGP, which may be due to differences in particle size or dispersion (Li *et al.*, 2021).

With respect to the Effect of UV light Exposure on the Hardness of the prepared specimen: After 24 hours of UV light exposure, the composite shows varying degrees of hardness. For pure PP (100/0), hardness remains constant before and after exposure, likely due to the absence of additives that would be degraded or oxidized under UV light. The Composite with FGP (90/10, 80/20, 70/30) (Figure 5) shows a slight reduction in hardness after UV light exposure, particularly for PP/FGP_A where hardness drops from 11.66 HRF (90/10) to 11.03 HRF (70/30). This indicates that although FGP enhances hardness, it offers limited UV light protection, possibly due to poor UV light absorption properties (Dhanorkar and Mallick, 2023).

On the other hand, composite with NGP (PP/NGP_B) (Figure 6) demonstrates a more substantial reduction in hardness, especially for the 60/40 composition where hardness decreases from 13.30 HRF to 11.23 HRF after UV light exposure. This suggests that NGP may undergo more pronounced UV light degradation, possibly because Nigerian graphite lacks certain surface treatments that could improve UV light stability (Singh and Das, 2022).

As for the comparison between Foreign and Nigerian graphite Fillers on the PP matrix: Before UV light exposure, both FGP and NGP show similar trends in hardness improvement, but after UV light exposure, the NGP-filled composite appears to degrade more than their FGP counterparts. This may point to a difference in the interaction between the fillers and the PP matrix. FGP may have better dispersion or higher interfacial interaction with the PP matrix, providing better overall mechanical performance and UV light resistance (Wang *et al.*, 2020).

Overall, both foreign and Nigerian graphite fillers improve the hardness of polypropylene composite, but the degree of UV light degradation varies. Foreign graphite provides more consistent mechanical performance both before and after UV light exposure while Nigerian graphite shows higher initial hardness but more significant degradation after UV light irradiation.

CONCLUSION

The present study has evaluated the effect of Foreign and Nigerian graphite on some of the mechanical properties of polypropylene in response to UV light irradiation for 48 hours. Hence the following conclusions were drawn. Foreign graphite composite (PP/FGP) tend to offer better UV light resistance at lower filler loading (90/10, 80/20), as the tensile strength of the material is not severely affected compared to Nigerian graphite composite which seems to favor applications where initial tensile strength is more critical as Nigerian graphite filler consistently provides higher initial tensile strength compared to foreign graphite filler across most compositions (70/30, 60/40, 50/50). While foreign graphite may be better suited for environments with higher UV exposure, particularly at lower filler loadings. The UV light affects both the PP/FGP and PP/NGP composites, however foreign graphite composite (PP/FGP_a) generally experience less reduction in tensile strength compared to the Nigerian graphite composite (PP/NGP_a). For instance, after UV exposure, PP/FGP_a at (80/20) composition experiences a decrease in tensile strength from 7.9 MPa to 7.6 MPa, while PP/NGP_a at (80/20) composition experiences a decrease in tensile strength from 8.4 MPa to 5.6 MPa. This indicates that FGP may provide slightly better protection against UV-induced degradation, possibly due to its higher purity or better UV shielding properties. PP/NGP composites show a greater decline in hardness compared to PP/FGP composites. For instance, in the 60/40 composition, the hardness of NGP decreases from 13.30 HRF to 11.23 HRF, while FGP decreases from 11.66 HRF to 11.03 HRF. This suggests that NGP is more susceptible to UV degradation, possibly due to its lesser ability to absorb or shield the polymer matrix from UV light.

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