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Effects of Natural Fibre as Biomass Additive on the Structural and Mechanical Properties of High-Density Polyethylene Bio-Composite

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ABSTRACT
The increasing global concern over plastic waste and environmental pollution has prompted growing interest in recycling and reinforcing polymeric materials with eco-friendly fillers. High-density polyethylene (HDPE), one of the most widely used thermoplastics, possesses excellent chemical resistance and processability but suffers from limited mechanical strength and wear resistance when recycled. To address these limitations, the use of agricultural by-products as sustainable bio-fillers offers an attractive alternative that simultaneously enhances material performance and promotes waste valorization. This study effects the mechanical and structural properties of recycled high-density polyethylene (HDPE) reinforced with Bambara groundnut shell ash (BGSA) as a sustainable bio-filler. Composites were developed with varying BGSA contents (0–25 wt%) and particle sizes (100–250µm) to evaluate their effects on ductility, strength, hardness, impact resistance, and wear behavior. The results show that elongation percentage decreased with increasing BGSA loading, dropping from 8.5% in neat HDPE to 4.5% at 25 wt%. Medium particle sizes, particularly 200µm, consistently maintained better ductility compared to smaller particles. Ultimate tensile strength (UTS) improved with BGSA addition, peaking at 28 MPa (12% above neat HDPE) at 15 wt% and 200µm, before declining at higher loadings. Hardness increased steadily with both higher BGSA content and smaller particle size, reaching a 51% improvement (68 BHN) at 25 wt%. Impact strength showed a decreasing trend with reinforcement, reducing from 18 kJ/m² to 12 kJ/m², though 200µm particles exhibited relatively better resistance. Wear resistance improved significantly, with wear rate reduced by 62% at optimal conditions (15–20 wt% and 200µm particle size), demonstrating potential for automotive applications such as brake pads. Scanning electron microscopy revealed improved particle dispersion with smaller BGSA sizes, while elemental analysis confirmed the presence of carbon, oxygen, silicon, and aluminum, supporting enhanced hardness at higher reinforcement levels. Overall, the results highlight that BGSA, particularly at 15 wt% and 200µm particle size, can effectively enhance the performance of recycled HDPE composites, offering an eco-friendly alternative to conventional fillers in engineering applications.

Keywords:
Recycled HDPE,
Bambara groundnut shell ash,
Bio-composites,
Tensile strength,
Hardness,
Wear resistance,
SEM/EDS.

INTRODUCTION

Growing environmental concerns associated with petroleum-based polymers have intensified the search for sustainable alternatives (Li *et al*., 2024). Agricultural wastes are increasingly attractive as reinforcements because they are inexpensive, biodegradable, lightweight, and less toxic compared to synthetic fibers (Mohanty *et al.,* 2002; Patel *et al.,* 2023). Their use also contributes to mitigating the environmental impact of synthetic reinforcements (Oladele *et al.,* 2014; Lins *et al.,* 2019; Tabie, 2025).

Bambara groundnut (VIGNA SUBTERRANEA), widely cultivated in sub-Saharan Africa, generates shells that are typically discarded but can be valorized as fillers in polymer composites (Majola *et al.,* 2021; Soumare *et al*., 2021). While other agro-waste fillers such as rice husk, coconut shell, pineapple, and palm kernel ash have been shown to improve hardness, wear resistance, and cost-effectiveness of polymer composites (Isaama et al., 2023; Adebisi *et al.,* 2021; Selmi *et al*., 2022; Danladi *et al.,* 2014)), limited studies have examined Bambara groundnut shell ash (BGSA), particularly in recycled HDPE composites with respect to particle size and composition.

Composite performance depends on the intrinsic properties of its constituents, dispersion, orientation, and matrix–reinforcement adhesion (Pothan *et al.,* 2003; May-Pat *et al.,* 2013; Idris *et al.,* 2013; Rohmat *et al.,* 2021, Tabie, 2025). Reinforcements, whether particulates, fibers, or sheets, are typically stronger than the matrix and enhance strength, hardness, and wear resistance when properly bonded (Ammisetti *et al.,* 2024). The growing demand for advanced composites, driven by applications in automotive, aerospace, and sports industries (Parveez *et al.,* 2022; Patel *et al.,* 2023), underscores the need for sustainable reinforcements from agricultural wastes.

This study aims to examine effect of Bambara groundnut shell ash (BGSA) composition (5–25 wt%) and particle size (100–250 µm) on the structural and mechanical properties of recycled high-density polyethylene (HDPE)/BGSA bio-composites.

MATERIALS AND METHODS

Materials Sourcing and Preparation

Recycled high-density polyethylene (HDPE) pellets were procured, while Bambara groundnut shells were sourced locally in Jalingo, Nigeria. The shells were washed to remove sand and treated in a sodium hydroxide solution (1:15) to eliminate lignin and pectin, then rinsed with distilled water and sun-dried. The dried shells were ground using a Thomas-Willy laboratory mill (Model 4, Philadelphia) and sieved into particle sizes of 100, 150, 200, and 250 µm, following BS1377:1990 standard. Bambara groundnut shell ash (BGSA) served as reinforcement, while HDPE functioned as the polymer matrix.

Composite Production

Composites were fabricated with BGSA contents of 5, 10, 15, 20, and 25 wt% at the specified particle sizes. The mixtures were processed in a two-roll mill (Model XK-160, USA) and fed into an injection molding machine preheated to 220 °C. The molten blend was injected into molds and cooled under steam to produce the test specimens.

Mechanical Testing

TensileStrength

Tensile properties were determined using a 100 kN capacity tensometer (Model: 130812) at Cutix Cable Plc, Nnewi, Anambra State, Nigeria. Samples were clamped, loaded to fracture, and the ultimate tensile strength (UTS) and percentage elongation were calculated using:

Ultimate tensile strength (UTS)

$$= \frac{Maximum\ Force(N)}{Original\ C.S.A\ (mm^2)} = \frac{\text{Pmax}}{A} \quad (1)$$

% Elongation

$$= \frac{Final\ guage\ length - Original\ guage\ length}{Original\ guage\ length} \times 100 \quad (2)$$

Hardness Test

Brinell hardness was measured using a DHT-6 tester at the Department of Metallurgical and Material Engineering, Ahmadu Bello University, Zaria, Kaduna State, Nigeria. The indenter was applied automatically, and hardness values were read directly from the scale.

Impact Test

Impact energy absorption was determined with a pendulum impact tester (Model: U1820) at the Department of Metallurgical and Material Engineering, Ahmadu Bello University, Zaria, Kaduna State, Nigeria. Samples were positioned between anvils 55 mm apart, struck by a pendulum released from 270°, and energy values were recorded in joules.

Wear Test

Wear resistance was evaluated using a universal wear tester against a rotating gray cast iron disc (200 mm × 40 mm) at 320 rpm under a load of 440 N for 20 min. Samples (12 × 12 × 5 mm) were prepared by grinding to uniform thickness. Wear rate was calculated using equation 3.

$$Wear\ rate = \frac{\Delta W}{PS\rho} \quad (3)$$

where,

ΔW is sample weight difference before and after test (in gram), S is total sliding distance (in meter), P is the applied load and ρ is Density (g/mm^3).

Scanning Electron Microscopy (SEM) and Energy Dispersive Spectroscopy (EDS)

The Scanning Electron Microscopy model (LEO 430i) was used to examine the structural and morphological properties of the Bambara groundnut shell ash content and recycled high density polyethylene samples. The SEM was operated under high vacuum at a working distance of 10 mm. The SEM was calibrated before use to ensure consistent resolution across all samples. The Energy Dispersive Spectroscopy (EDS) was carried out on the samples and to determine the elemental composition of the composite.

RESULTS AND DISCUSSION

Effect of BGSA Content and Particle Size on Mechanical Properties

The percentage elongation (%E) and ultimate tensile strength (UTS) of recycled high density polyethylene

(RHDPE)/ Bambara groundnut shell ash bio-composite are presented in Figures 1-4.

Figures 1 and 2 show the effect of Bambara groundnut shell ash content and particle size on the percentage elongation of reinforced recycled high density polyethylene bio-composite. The data illustrated in Figures 1 and 2 show that the elongation percentage of the HDPE/BGSA bio-composite is inversely proportional to the BGSA content, regardless of the particle size, within the scope of this study. The elongation dropped from around 8.5% in the uncomposite HDPE to approximately 4.5% at 25 wt% of BGSA loading. This trend holds true for similar results found in the work of (Haque *et al.,* 2009) on natural fiber-reinforced HDPE composites, where fiber content increase from 0 to 50 wt% dictated a decrease in ductility. Ductility may be diminished in these composites because of the rigid particles. These must be BGSA, since they are the only reinforcement here. They restrict the mobility of the chains of the (presumably) ductile polymer and should thus lead to an increase in stress concentration points in the matrix. This is just what (Issam *et al.,* 2023) observed in their study of oil palm empty fruit bunch (OPEF) reinforced HDPE, where they found that both ductility and ultimate tensile strength decreased as fiber content increased. About the effects of particle size (Figure 2), Different BGSA loadings resulted in varying percentages of elongation, but the 200 µm particle size showed the highest consistency in that regard. The second most consistent was the 250 µm particle size, while the 100µm particle size yielded the lowest ductility. That optimal particle size range for maintaining BGSA reinforcement was consistent with the finding that median-sized BGSA particles are better for achieving reasonable ductility. As compared with small or large BGSA particles.

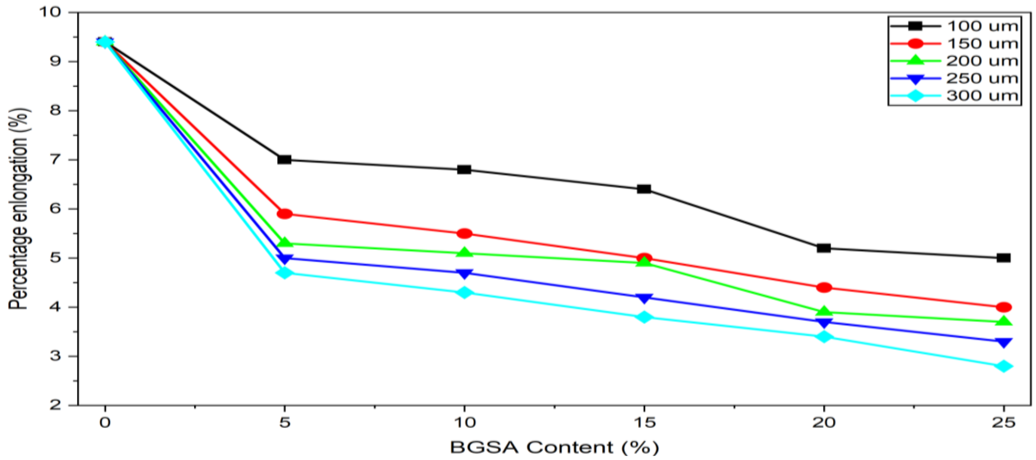

Figure 1: Effect of BGSA content on the percentage elongation of reinforced recycled high density polyethylene bio-composite

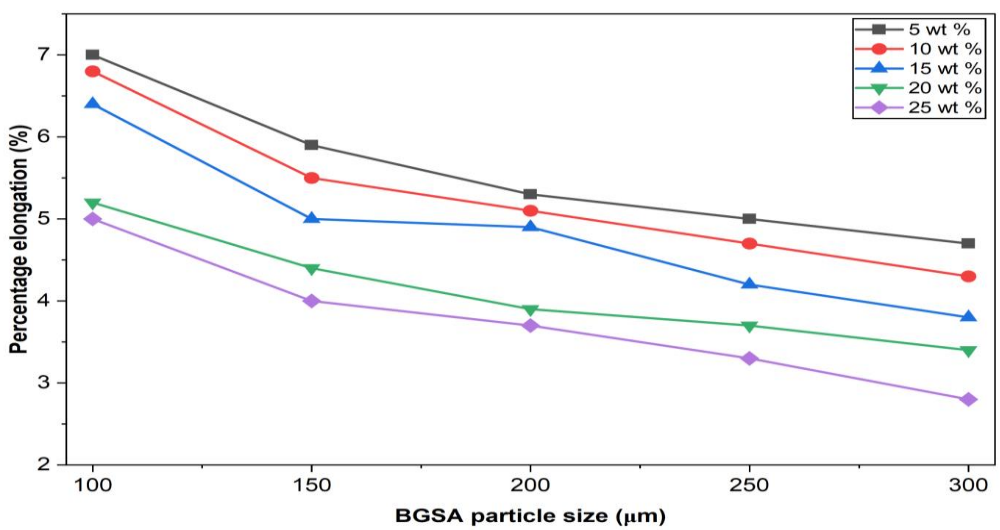

Figure 2: Effect of BGSA particle size (µm) on the percentage elongation of reinforced recycled high density polyethylene bio-composite

Ultimate Tensile Strength (UTS)

Figures 3 and 4 indicate the variation of ultimate tensile strength of recycled high density polyethylene matrix with BGSA content and particle size respectively. The UTS results (Figures 3 and 4) show a curious trend in which the tensile strength first seems to increase with the amount of BGSA, reaching a kind of max level at around 15 wt%. Then, at higher loadings, the strength drops. The best UTS was very close to that 15 wt% level: a sample reinforced with 15wt% BGSA (200µm size) in our matrix reached an ultimate tensile strength of 28MPa, nearly 12% better than the unreinforced version, which was at 25 MPa. This drop-off business in tensile strength, after it seems to peak, is puzzling. The enhancement of UTS observed up to 15 wt% can be linked to the efficient transfer of load from the matrix to the reinforcement particles. This observation is in line with that of (Jonathan *et al*. 2021). The decrease in UTS at higher loadings (20-25wt%) cannot be ascribed to lack of matrix-material compatibility, as evidenced by the retention of approximately 80% of the compressive strength of the matrix in the composite. This trend contrasts with that reported by (Jonathan *et al.* 2021), who found that the properties of HDPE continuously improved with reinforcement from multiwalled carbon nanotubes, with a 42.4% increase in the material's UTS at a 3wt% addition. (Kumar *et al.,* 2021) attributed the difference in behavior to the comparative aspect ratio and surface area of the carbon nanotubes and BGSA in the research. Figure 4 depicts the particle size effect, demonstrating that UTS consistently ranks highest with 200µm particles across most BGSA loadings, followed closely by 150 µm particles. This notion of optimal performance with medium-sized particles aligns with (Shubhashini, 2011; Sarki *et al.,* 2011) reported improved toughness in HDPE composites at 63 µm marble sludge particle size, emphasizing the importance of particle size optimization.

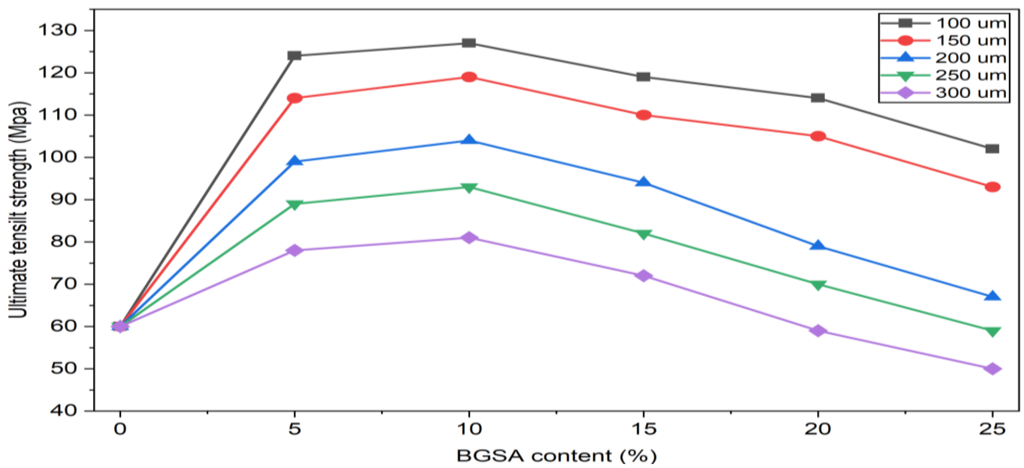

Figure 3: Effect of BGSA content (%) on the ultimate tensile strength (Mpa) of reinforced recycled high density polyethylene bio-composite

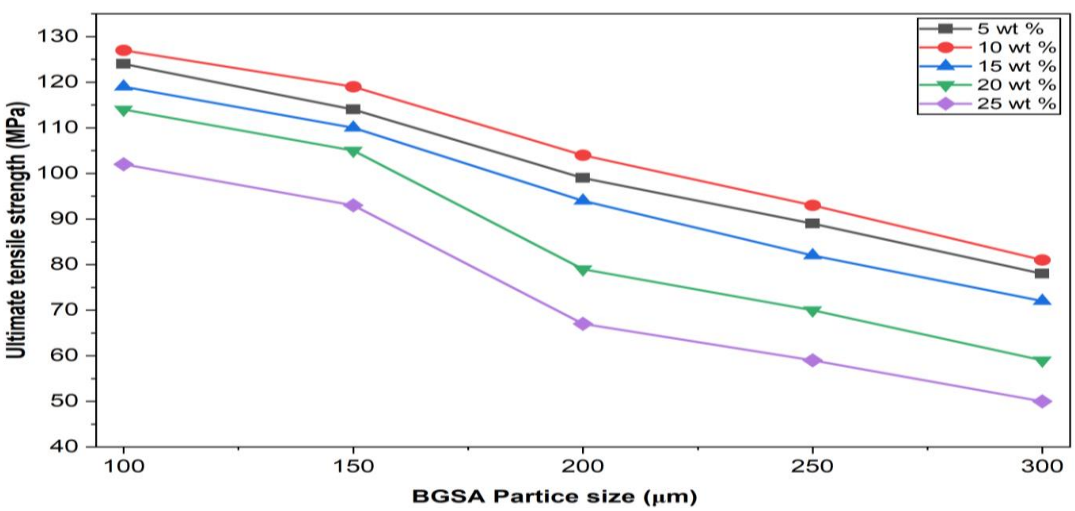

Figure 4: Effect of BGSA particle size (µm) on the ultimate tensile strength (Mpa) of reinforced recycled high density polyethylene bio-composite

Brinell Hardness

The effects of BGSA particle sizes and concentration on the hardness and impact strength of recycled high density polyethylene (RPE)/Bambara groundnut shell ash bio-composite are presented in Figures 5 to 6.

Figures 5 and 6 depict the variation of Brinell hardness of recycled high density polyethylene matrix with Bambara groundnut shell ash content and particle size respectively. The hardness results (Figures 5 and 6) demonstrate a consistent increasing trend with both BGSA content and decreasing particle size. The hardness increased from approximately 45 BHN for unreinforced HDPE to about 68 BHN at 25 wt% BGSA loading, representing a 51% improvement. This trend is consistent with the findings of (Chinonso & Isaac, 2012), who reported significant increases in hardness with reduced particle size and increasing fiber content in OPEF-reinforced HDPE composites.

The continuous improvement in hardness with increasing BGSA content can be attributed to the inherent hardness of the ceramic-like BGSA particles, which resist indentation more effectively than the polymer matrix. The inverse relationship between particle size and hardness (Figure 6) suggests that smaller particles provide better reinforcement efficiency for hardness improvement, possibly due to increased particle-matrix interfacial area and more uniform stress distribution.

These results align with the observations of (Boukfessa *et al.,* 2021), who reported increasing hardness trends with reinforcement content in carbon black and natural reinforced composites, attributing this behavior to the rigid nature of the reinforcing particles.

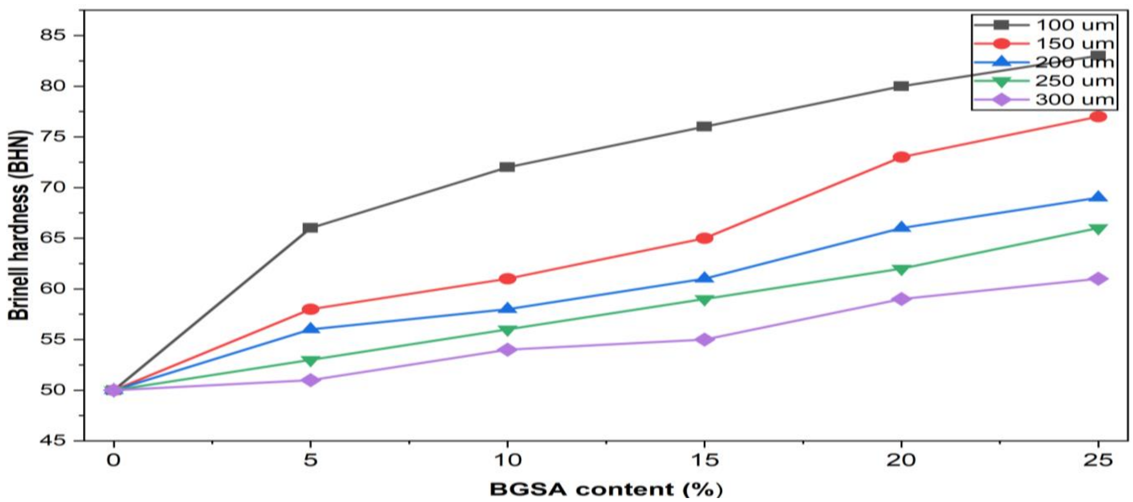

Figure 5: Effect of BGSA content (%) on the Brinell hardness (BHN) of reinforced recycled high density polyethylene bio-composite

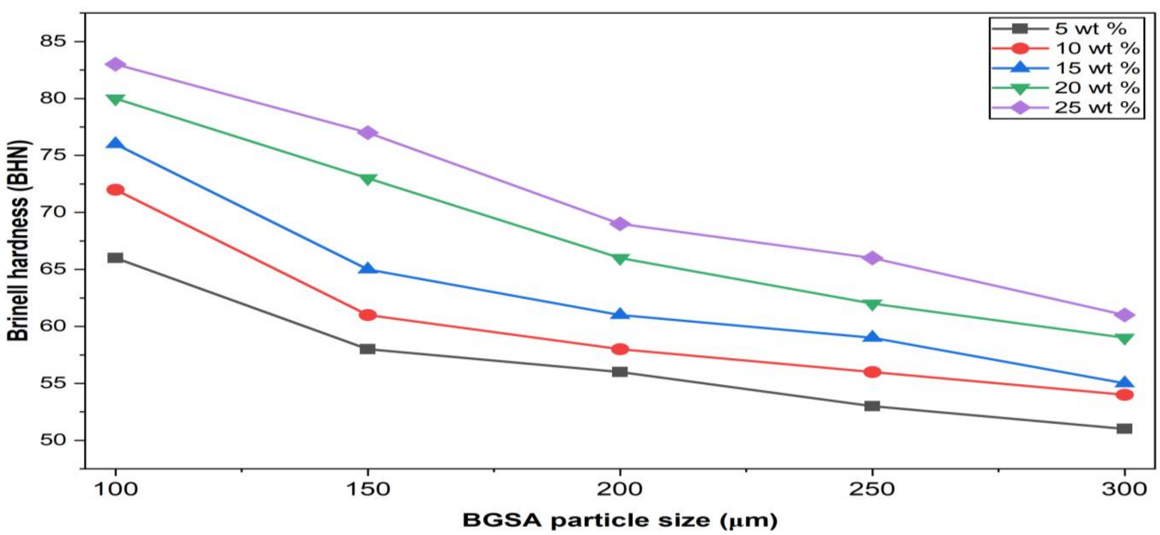

Figure 6: Effect of BGSA particle size (μm) on the Brinell hardness (BHN) of reinforced recycled high density polyethylene bio-composite

Impact Strength

The variations of impact strength of recycled high density polyethylene matrix with Bambara groundnut shell ash content and particle sizes are presented in Figures 7 and 8. The impact strength results show a general decreasing trend with increasing BGSA content, with some fluctuations depending on particle size. The impact strength decreased from approximately 18kJ/m² for unreinforced HDPE to about 12kJ/m² at 25wt% BGSA loading. However, certain combinations, particularly with 200μm particles at 10-15wt% loading, maintained relatively higher impact strength values. This decreasing trend is consistent with the brittleness typically associated with particulate-reinforced composites, as reported by (Bashar *et al*., 2012) in coconut shell-reinforced brake pad materials. The rigid BGSA particles act as stress concentrators, leading to crack initiation and propagation under impact loading conditions.

Interestingly, the 200μm particle size showed the best impact performance across most loadings, which correlates with its superior performance in tensile and ductility tests. This suggests that this particle size provides the best balance between reinforcement and matrix compatibility.

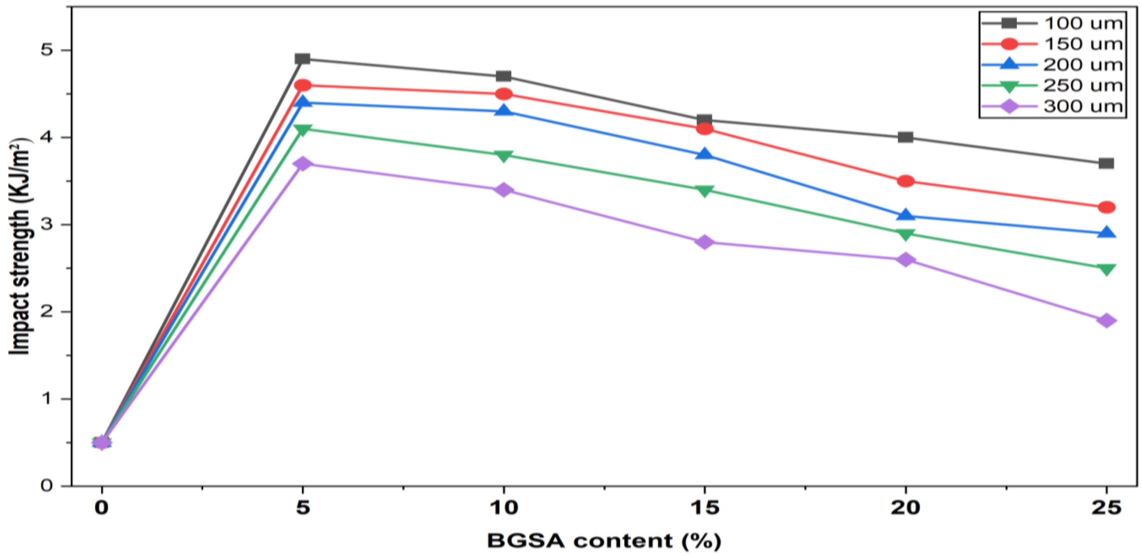

Figure 7: Effect of BGSA content (%) on the impact strength (KJ/m^2) of reinforced recycled high density polyethylene bio-composite

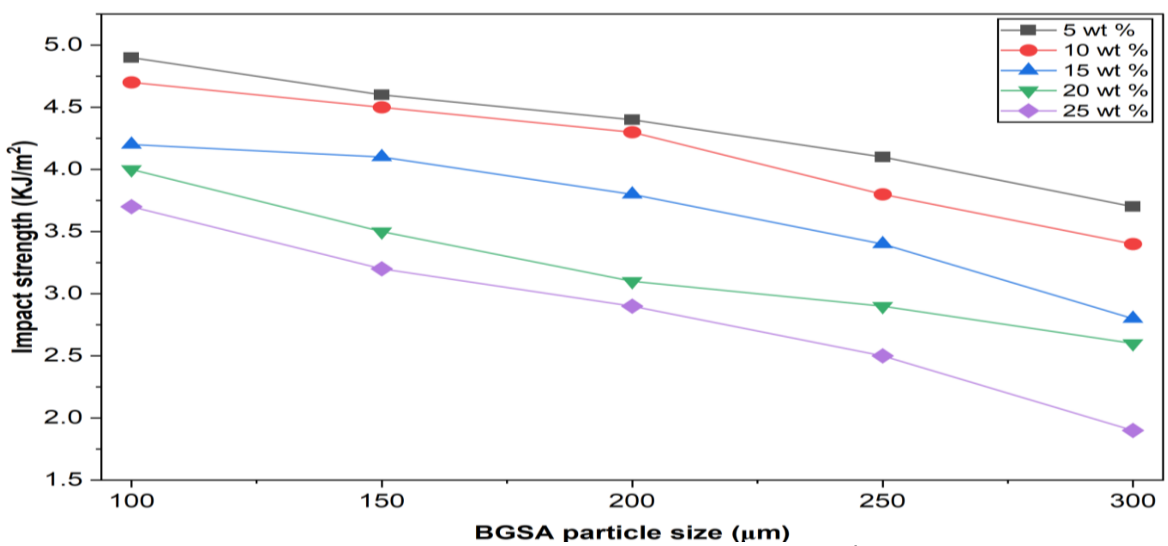

Figure 8: Effect of BGSA particle size (%) on the impact strength (KJ/m^2) of reinforced recycled high density polyethylene bio-composite

Wear Rate analysis

The effects of BGSA content and particle size on the wear rate of Bambara groundnut shell ash reinforced recycled high density polyethylene bio-composite are presented in Figures 9 and 10. It can be observed in the Figures 9 and 10 that BGSA addition significantly improves the wear resistance of HDPE, with wear rate decreasing from approximately 0.65mm²/Nm for unreinforced HDPE to about 0.25mm²/Nm at optimal loadings. This represents a 62% improvement in wear resistance, which is crucial for automotive applications like brake pads.

The optimal wear performance was achieved at 15-20wt% BGSA loading with 200µm particle size. This improvement can be attributed to the abrasive resistance provided by the hard BGSA particles, which protect the polymer matrix from wear. The results support the findings of previous researchers who investigated various agricultural wastes for brake pad applications (Edokpia *et al*., 2014; Guojun *et al*., 2014).

The mechanical properties achieved in this study compare favorably with other natural fiber-reinforced HDPE composites reported in literature. The 12% improvement in UTS at optimal loading is comparable to the results reported by (Adebisi *et al*., 2021), who found enhanced mechanical properties in HDPE composites reinforced with Bambara groundnut shell particles and palm kernel shell ash.

The significant improvement in wear resistance makes these bio-composites particularly suitable for brake pad applications, addressing the environmental and health concerns associated with asbestos-based materials as highlighted by (Idris *et al*., 2013).

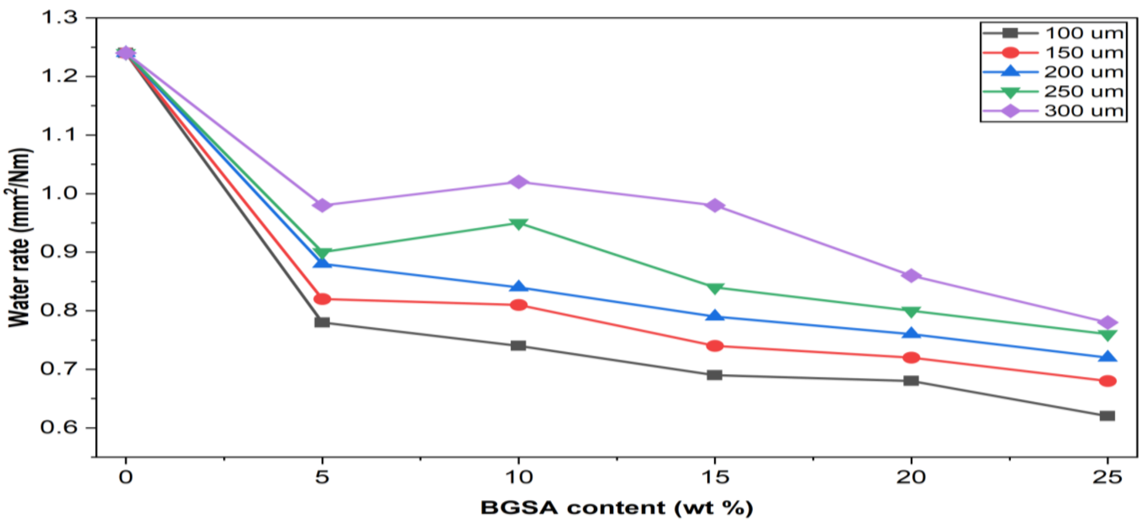

Figure 9: Effect of BGSA content (wt%) on the water rate (mm^2 /Nm) of reinforced recycled high density polyethylene bio-composite

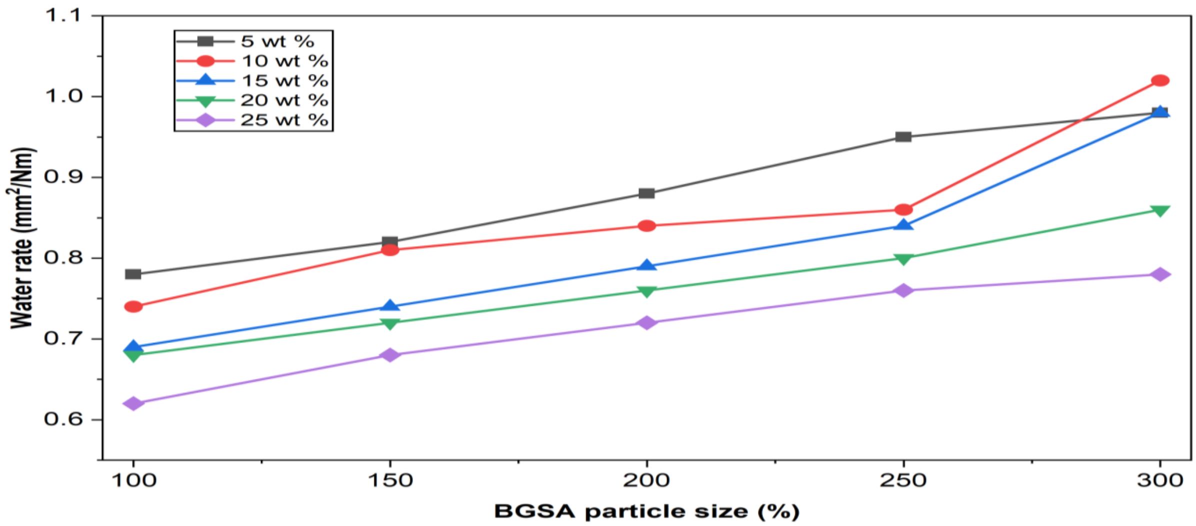

Figure 10: Effect of BGSA particle size (%) on the water rate (mm^2 /Nm) of reinforced recycled high density polyethylene bio-composite

Scanning Electron Microscopy (SEM) and Energy Dispersive Spectroscopy (EDS) Analyses of the Developed Composite

Figures 11- 15 show the scanning electron microscopy analysis of recycled high density polyethylene (RPE) matrix reinforced with different concentrations of Bambara groundnut shell ash (BGSA) of different particle sizes. Figures 11(a)-15(a) revealed dispersion of BGSA in the polymer matrix which was more in the composite of small Bambara groundnut shell ash particle size (100µm). Figures 11 (b) to 15 (b) show the elemental analysis of the developed composites. The spectrums reveal the presence of four (4) major elements such as carbon, oxygen, silicon and aluminum. It is also revealed that the carbon content of the developed composite increased as the reinforcement composition increased. This account for the increased hardness of the composite as the bambara groundnut shell content increased in Figures 5 and 6.

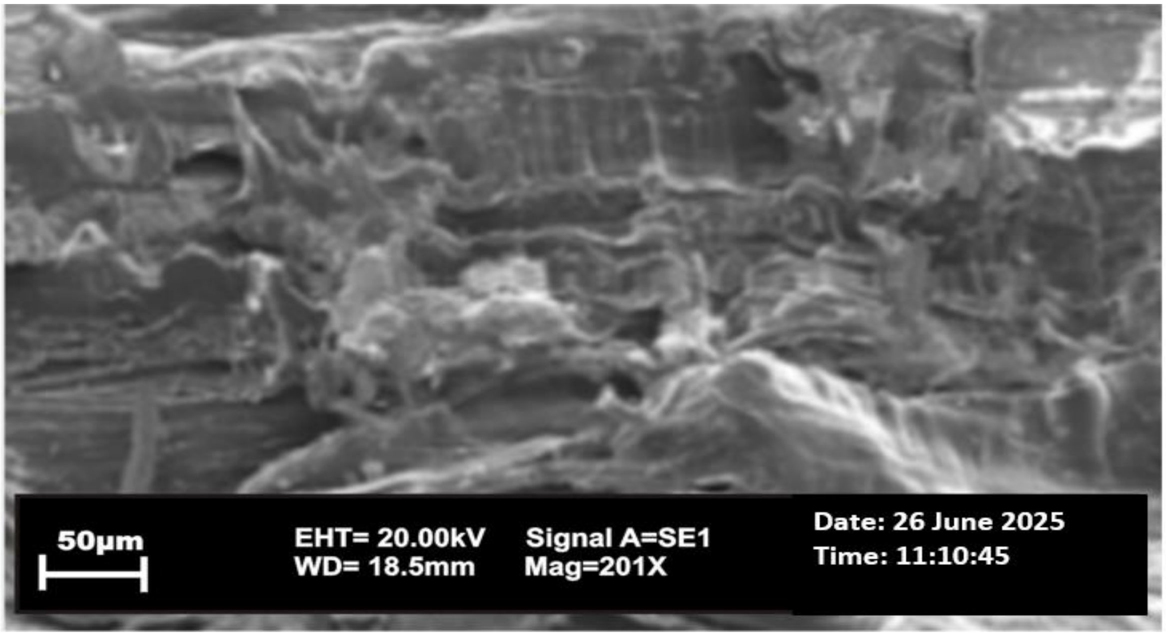

Figure 11a: Micrograph (SEM) of RPE matrix reinforced with 5wt%BGSA of 100µm particle size

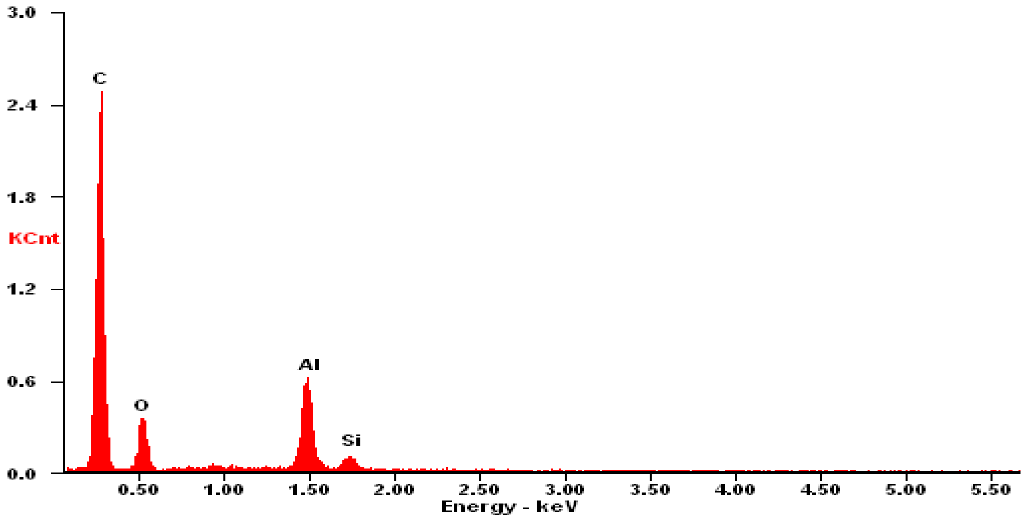

Figure 11b: EDS spectrum of RPE matrix reinforced with 5wt%BGSA of 100µm particle size

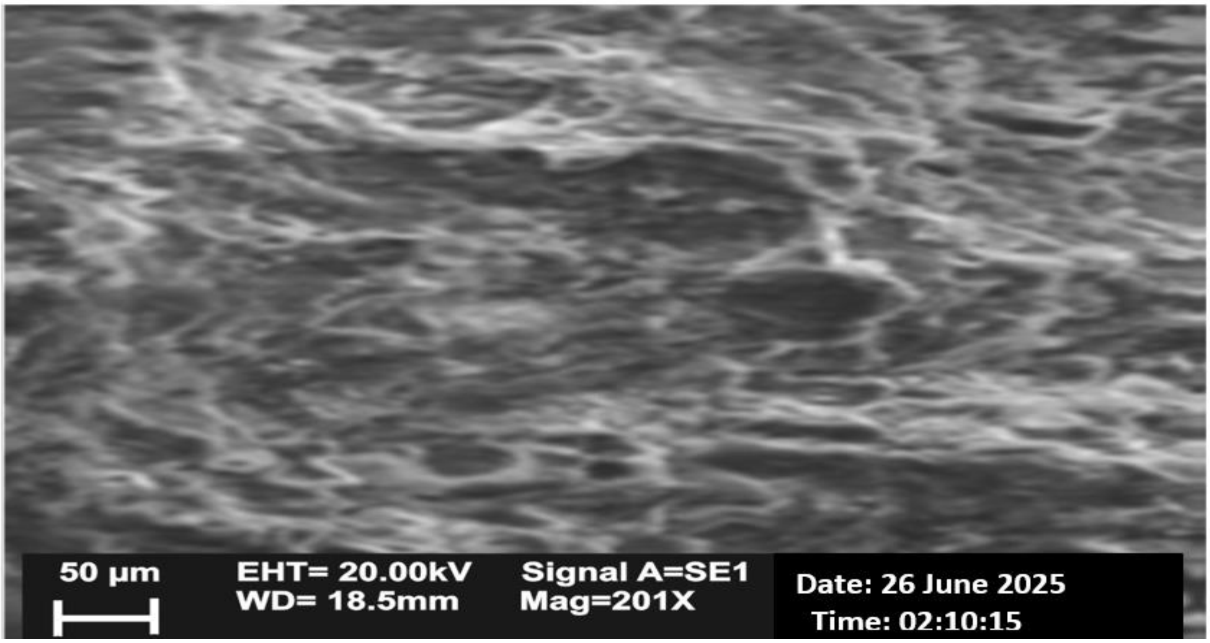

Figure 12a: Micrograph (SEM) of RPE matrix reinforced with 20wt%BGSA of 100µm particle size

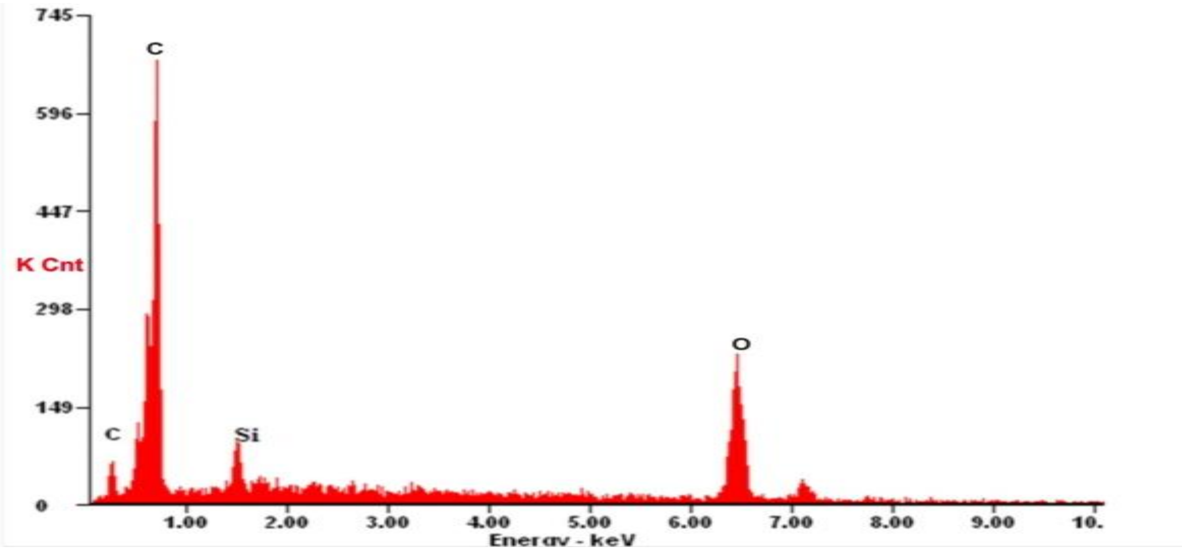

Figure 12b: EDS spectrum of RPE matrix reinforced with 20wt%BGSA of 100µm particle size

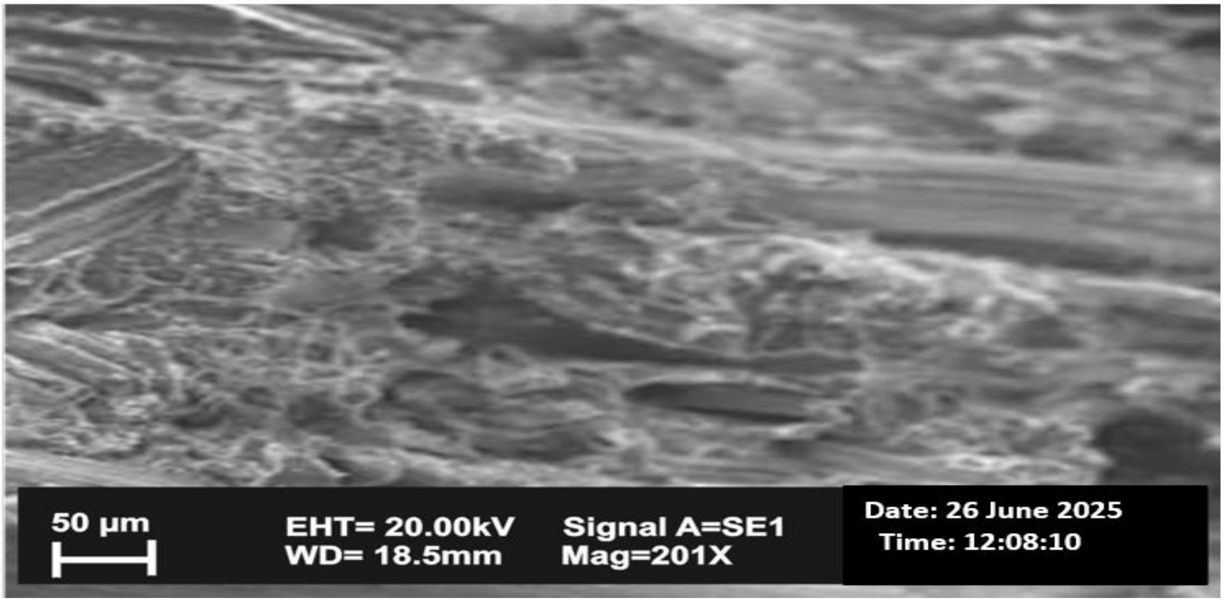

Figure 13a: Micrograph (SEM) of RPE matrix reinforced with 25 wt% BGSA of 100µm particle size

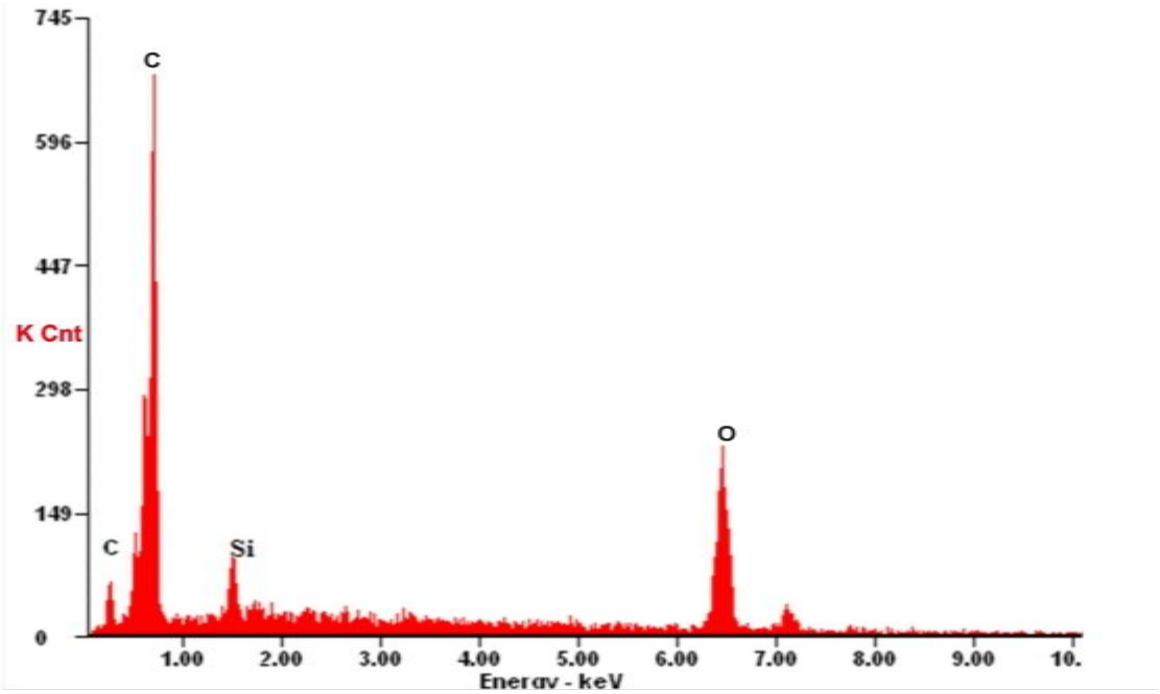

Figure 13b: EDS spectrum of RPE matrix reinforced with 25wt%BGSA of 100µm particle size

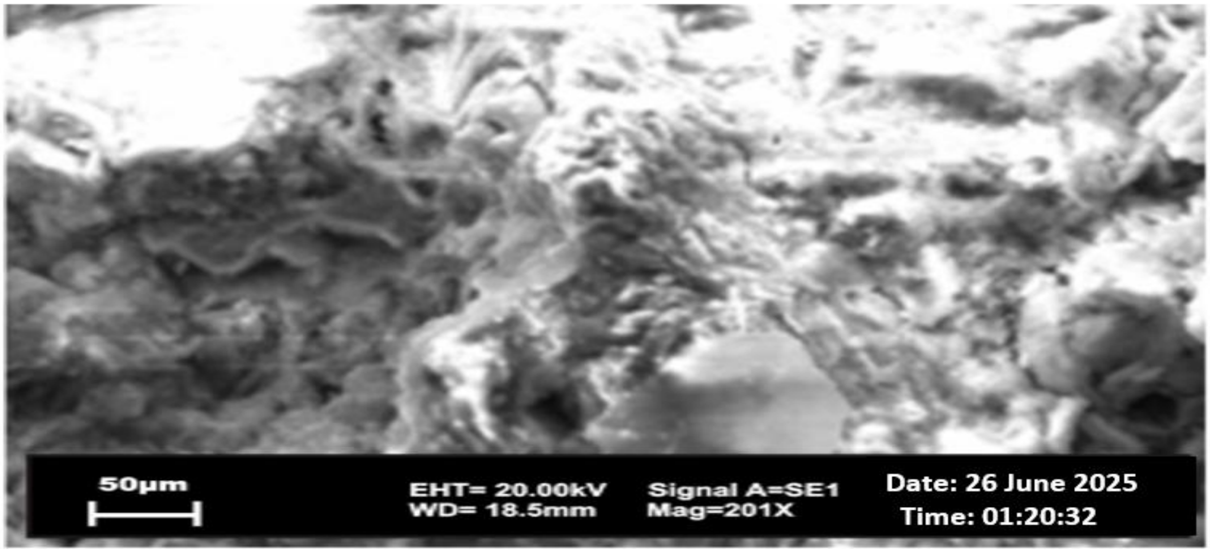

Figure 14a: Micrograph (SEM) of RPE matrix reinforced with 25wt%BGSA of 150µm particle size

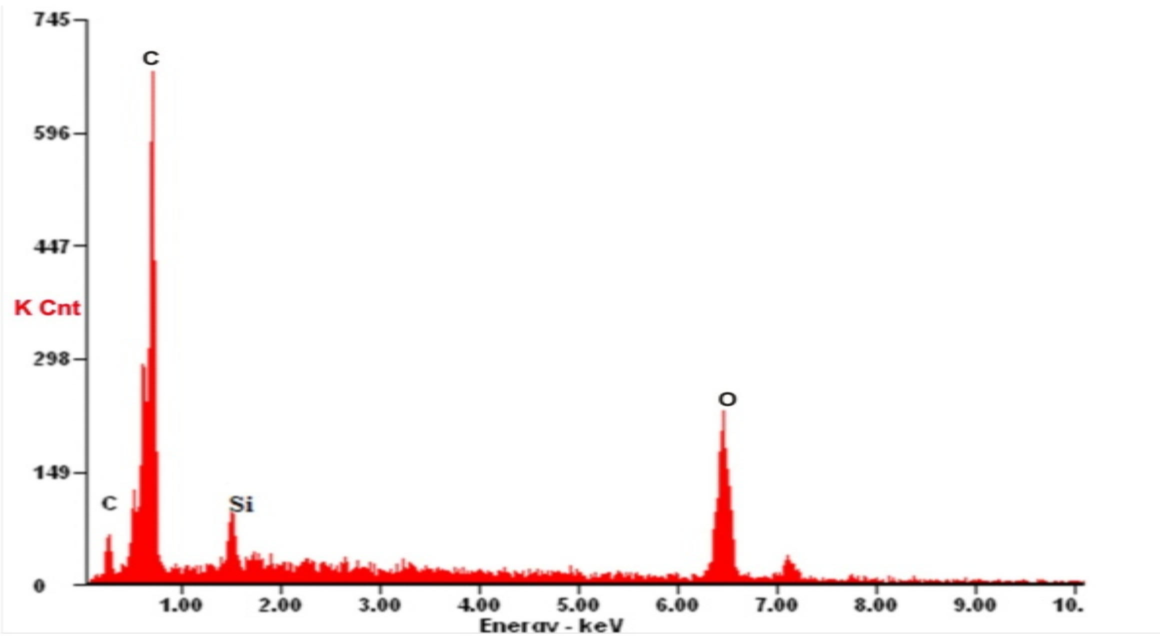

Figure 14b: EDS spectrum of RPE matrix reinforced with 25wt%BGSA of 150µm particle size

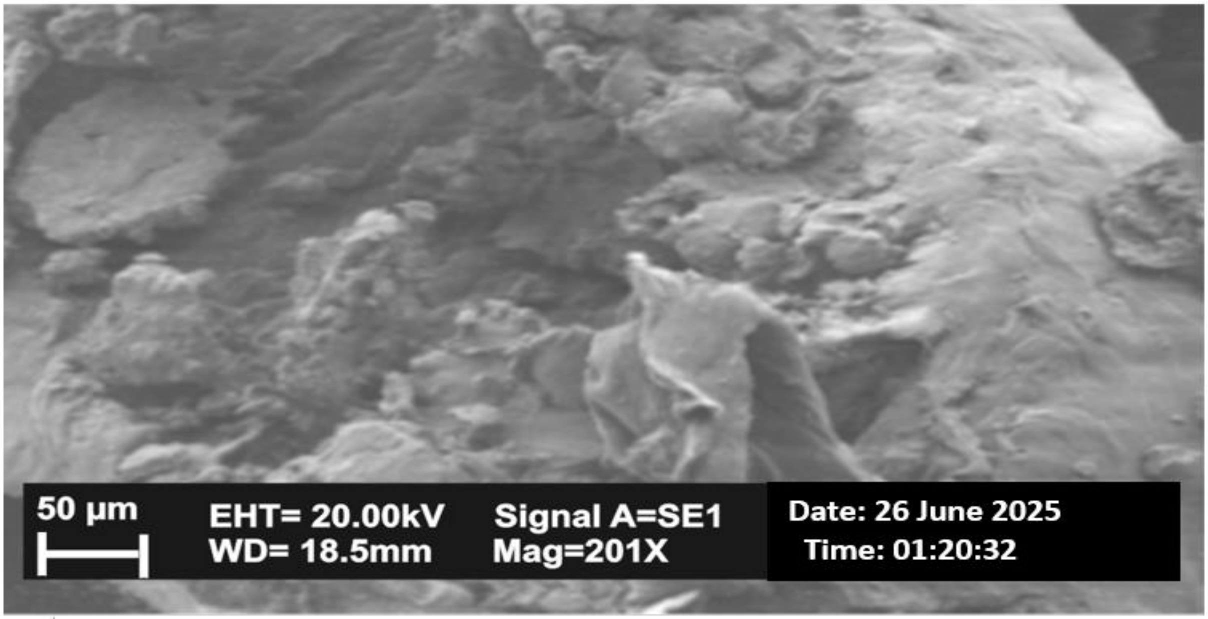

Figure 15a: Micrograph (SEM) of RPE matrix reinforced with 25wt%BGSA of 200µm particle size

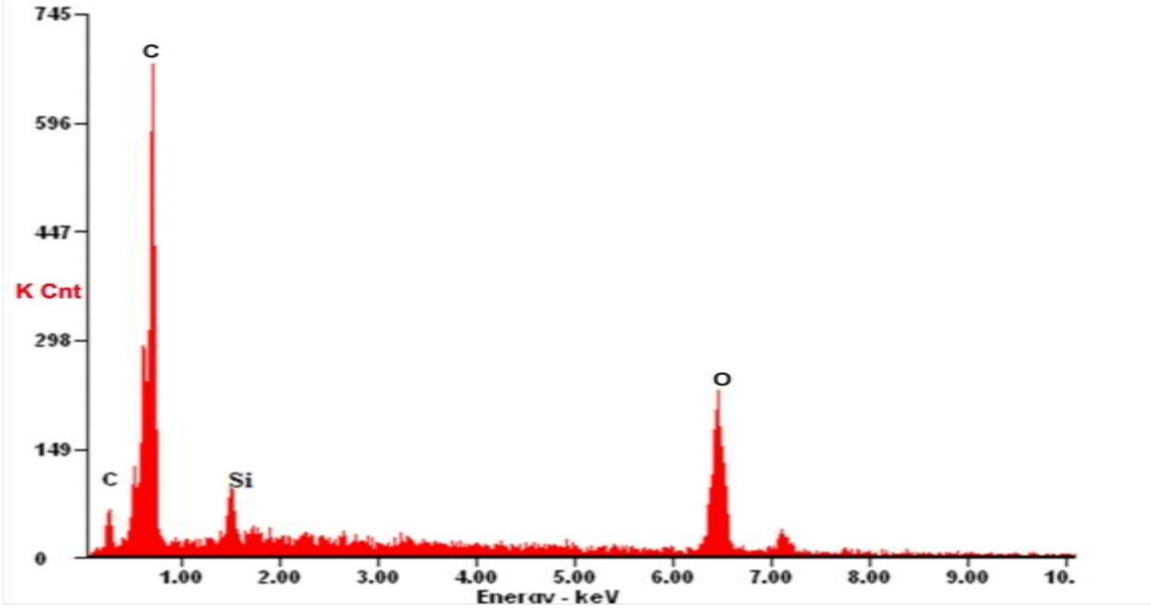

Figure 15b: EDS spectrum of RPE matrix reinforced with 25wt%BGSA of 200µm particle size

CONCLUSION

This study investigated the effect of Bambara groundnut shell ash (BGSA) as a sustainable reinforcement in recycled high-density polyethylene (HDPE) bio-composites. BGSA contents of 5–25 wt% and particle sizes of 100–250 µm were evaluated through mechanical testing and microstructural analysis. The results showed that optimal performance occurred at 15–20 wt% BGSA loading with 200µm particle size, yielding a balance of strength, hardness, and wear resistance. Maximum tensile strength of 131 MPa was obtained at 10 wt% loading, while superior wear resistance was achieved at higher loadings up to 25 wt%. Microstructural examination confirmed uniform particle distribution and strong interfacial bonding, validating predictions of composite theory. Overall, BGSA reinforcement provided both fundamental insights into bio-composite behavior and practical guidelines for sustainable materials development.

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