



Production and Characterization of Biodegradable Packaging Composites from Recycled Expanded Polystyrene Filled with Groundnut Shell Cassava Peel Powders

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KEYWORDS

Biodegradable composite,
Recycled polystyrene,
Cassava peel powder,
Groundnut shell powder,
Agro-waste filler,
Packaging material.

ABSTRACT

This study reports the production and characterization of biodegradable packaging materials made from recycled expanded polystyrene (REPS) filled with alkali-treated groundnut shell powder (GNSP) and cassava peel powder (CPP) as a green alternative to conventional Styrofoam, which are usually disposed in landfills or burnt in incinerator, resulting in material loss, emission of toxic and carcinogenic substances into the atmosphere, harming the environment and human life. Composite samples (A-E) were formulated with a fixed REPS, GNSP and varying CPP content, compounded on a two-roll mill and formed by compression moulding. The composites were characterized using thermogravimetric analysis, scanning electron microscopy, and Fourier transform infrared spectroscopy, and evaluated for tensile, impact, hardness, flexural, density, water absorption, and soil-burial biodegradability properties. TGA showed a multi-stage thermal decomposition profile for all composite specimens compared with the single-stage profile of the control, denoting reduced thermal stability with increasing filler content. SEM revealed homogeneous filler distribution at intermediate loading (Sample B) and agglomeration with crack formation at higher loading (Sample E). FTIR confirmed hydroxyl and carbonyl groups associated with the natural fillers. Tensile properties improved initially in Sample A (27.66 MPa), with Sample B recording the highest tensile strength (29.22 MPa), while Sample C gave the highest impact strength (0.183 J/mm) and flexural modulus (2686.93 MPa), after which performance declined at higher loadings. Density and hardness decreased, while water absorption increased, with increasing filler content. Biodegradation tests after 63 days showed progressive weight loss from the control to Sample E, with Sample C recording the highest weight loss (0.705%), confirming that natural fillers render the composites more susceptible to microbial attack and thereby enhance biodegradability.

CITATION

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INTRODUCTION

Modernisation and industrialisation have driven a continuous rise in global plastic demand, resulting in severe environmental pollution because conventional plastics are non-biodegradable and accumulate in landfills (Eriksen et al., 2014). Between 1950 and 2017, the world produced about 9.2 billion tonnes of plastic, of which approximately 7 billion tonnes were buried, incinerated, or deposited in landfills (Eriksen et al., 2014). In sub-Saharan Africa, only about 15-25% of packaging waste is recycled, with the rest dumped in open areas or landfills, worsening waste-management challenges as the population and consumption of single-use packaging materials such as expanded polystyrene (Styrofoam) continue to rise (Bakhtiyar et al., 2020).

In Nigeria, agricultural waste is generated in abundance year-round, yet the development of biodegradable plastic packaging technology from such waste remains limited (Babayemi et al., 2018). Cassava peel and groundnut shell, by-products of some Nigerian agro-processing industries are lignocellulosic materials rich in cellulose, hemicellulose, and lignin (Anyanwu et al., 2015; Czaikoski et al., 2020), making them suitable low-cost fillers for eco-friendly composite development. Blending these agro-waste fillers with recycled expanded polystyrene (REPS) offers a dual environmental benefit; valorising agricultural wastes and diverting polystyrene waste from landfills (Ugwu & Obele, 2023), while imparting biodegradability to an otherwise persistent polymer (Shah et al., 2008; Ahmed et al., 2018).

Although several biopolymers such as starch, chitosan, protein and bacterial cellulose have been studied for eco-friendly food packaging (Vroman & Tighzert, 2009; Thakur et al., 2018), many starch sources are edible and could raise food-security concerns if widely adopted for packaging. Non-edible agro-industrial wastes such as cassava peel and groundnut shell therefore offer a more eco-friendly alternative, supporting food security while providing economic benefits through waste valorisation (Rameshkumar et al., 2020). However, information on the combined use of cassava peel and groundnut shell powders as fillers in recycled expanded polystyrene matrices, particularly their thermal, morphological, mechanical, and biodegradation behaviour, remains limited.

This study therefore aims to produce and characterize biodegradable packaging composites from recycled expanded polystyrene filled with groundnut shell and cassava peel powders. The specific objectives are to produce composite samples from REPS, GNSP and varying

proportions of CPP, characterize the composites using TGA, SEM and FTIR, determine the tensile, impact, hardness, flexural, density, water absorption and biodegradability properties of the composites in accordance with ASTM standards and compare the physico-mechanical properties of the composites with an unfilled control sample.

MATERIALS AND METHODS

Materials and Instruments

Waste expanded polystyrene packs, cassava peels and groundnut shells were used as the principal raw materials, along with 5% sodium hydroxide pellets (Sigma-Aldrich), distilled water, cloth and mesh sieves, and standard glassware. Composite processing employed a two-roll mill (Model 5183, North Bergen, USA) and a compression moulding machine (Model 0557, Wenzhou Zhiguang Ltd, China). Mechanical and thermal characterization was carried out using a universal testing machine (D-100KN), a Resil impact tester (Cat. No. 261, Norwood Instruments), a Vickers micro-hardness tester (Model MV1-PC), a TA Q5000 IR thermogravimetric analyser, a Prisma E scanning electron microscope, and a Thermo Nicolet Nexus 870 FTIR spectrometer.

Sample Collection and Preparation

The peels obtained from the processing of cassava root into Garri were purchased at the processing site. Dried groundnut shells were then bought from a local market, and waste Styrofoam were collected from disposal sites within Kaduna metropolis, Nigeria.

In this study, the cassava peels were thoroughly washed before they were subjected to oven drying at 60 °C for 24 h, milled and sieved using a 30 mm mesh into a fine powder (CPP). The groundnut shells were washed, air-dried for 24 h, milled, and sieved using a 30 mm mesh. We treated 500 g of the groundnut powder with 2000 cm³ of 5% (w/v) NaOH solution for 1-2 h, with periodic stirring to enhance interfacial bonding with the polymer matrix. The alkali-treated groundnut shell powder (GNSP) was then filtered, washed with distilled water to remove excess NaOH, and oven-dried at 40 °C for 12 h.

Composite Formulation

Table 1 shows the composite samples, with REPS (100 g) and GNSP (30 g) fixed. Then, varying CPP contents at different levels between 10-50 g gives us samples named sample A-E respectively, and finally, the unfilled sample of REPS acts as our control.

Table 1: Composite Formulation of Recycled Expanded Polystyrene (REPS), Cassava Peel Powder (CPP) and Groundnut Shell Powder (GNSP)

Sample	Control(g)	A(g)	B(g)	C(g)	D(g)	E(g)
REPS	100	100	100	100	100	100
CPP		10	20	30	40	50
GNSP		30	30	30	30	30

Compounding and Compression Moulding

Waste REPS (100 g) was softened on the two-roll mill at 180 °C for 5 min. Upon band and bank formation, the pre-treated GNSP (30 g) was introduced and cross-mixed for 3 min, followed by CPP (10-50 g) mixed for a further 2 min. The resulting compound was sheeted out and labelled A-E accordingly. The compounds were then placed in a 120 mm × 100 mm × 3.2 mm metal mould and hot-pressed at 160 °C and 5.0 MPa for 5 min, cooled under pressure, and demoulded.

Characterization

The fabricated REPS/CPP/GNSP composites (A-E) and the control were characterised for thermal stability (TGA), surface morphology (SEM), functional groups (FTIR), and physico-mechanical performance properties.

Tensile strength: determined per ASTM D-638 using dumbbell-shaped specimens on a universal testing machine, from which tensile strength, elastic modulus and elongation at break were obtained.

Impact strength: determined per ASTM D-256 on notched specimens (64 × 12.7 × 3.2 mm) using an Izod (Resil) impact tester; impact strength (J/mm) was calculated as the average impact energy divided by specimen thickness (3.2 mm).

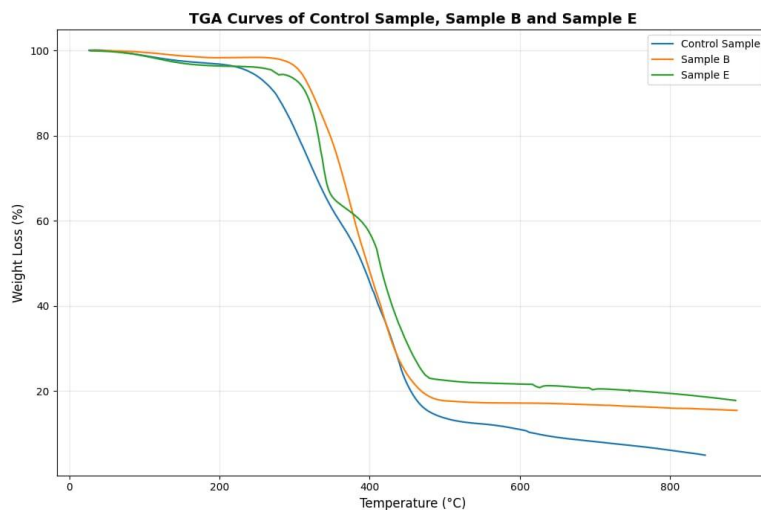
Hardness: determined per ASTM D2240 using a Vickers microhardness tester, with readings taken in triplicate at different positions on each sample and averaged.

Flexural strength and modulus: determined per ASTM D-790 by three-point bending of specimens (100 × 25 × 3.2 mm) over an 80 mm support span, using Flexural strength = $3FL/2bd^2$ and Flexural modulus = $FL^3/4bd^3D$, where F is the maximum load, L the support span, b the specimen width, and d the specimen thickness.

Water absorption: determined per ASTM D570 by total immersion of specimens with dimensions (50 × 50 × 3.2 mm) in distilled water for 7 days, with weight recorded after every 24 hours. Percentage water absorption = $(W_2 - W_1)/W_1 \times 100$.

Density: The samples' densities were determined in accordance with ASTM D792-08 by the water-displacement method, using density = mass/volume.

Soil-burial biodegradation: carried out per ASTM D6003. Dried specimens (60 × 60 × 3 mm) of known initial weight were buried horizontally at 5 cm depth in natural topsoil maintained at ~60% water-holding capacity and 25 ± 2 °C for 63 days, after which percentage weight loss was determined.

RESULTS AND DISCUSSION**Thermogravimetric Analysis (TGA) Result****Figure 1: TGA Curves for Control Sample, Sample B and Sample E**

The TGA graph showed that the mass of the control (REPS) sample changed with increasing temperature. At approximately 50-150 °C, there is a small, gradual decrease in mass. Between 200 and 350 °C, the sample undergoes a major and rapid loss of mass, which is the main degradation region of the material. After around 350-400 °C, the rate of weight loss becomes much slower, and the curve begins to level off, representing the thermally stable residue mass, or char. It exhibited a single major degradation stage typical of polystyrene, indicating the presence of its aromatic hydrocarbon backbone and its resistance to thermal decomposition. In contrast, composite Sample B showed good thermal stability up to 300 °C, as only a small amount of weight was lost up to this temperature. Major thermal degradation occurs between 300-400 °C. After about 500 °C, the sample becomes relatively stable again, leaving 15-17% residue. Sample E showed relative stability at lower

temperatures, with only a small mass reduction below 250 °C. A major decrease in mass occurs from 300-450 °C. Above 450-500 °C sample E becomes almost stable, suggesting that decomposable material has already been lost, with only 10-15% of the original mass remaining as residue. This multi-stage degradation corresponds to sequential dehydration, hemicellulose and cellulose decomposition, and finally lignin breakdown. Sample E, with the highest CPP content, showed the lowest thermal stability and the earliest onset of degradation, indicating that increasing agro-waste filler content reduces thermal resistance while improving biodegradability. Despite this reduction, the composites retained sufficient thermal stability for packaging applications, where extreme processing or service temperatures are uncommon. Residual char observed at the end of the TGA runs was attributed to lignin-derived carbonaceous residues (Figure 1).

Scanning Electron Microscopy (SEM) Results

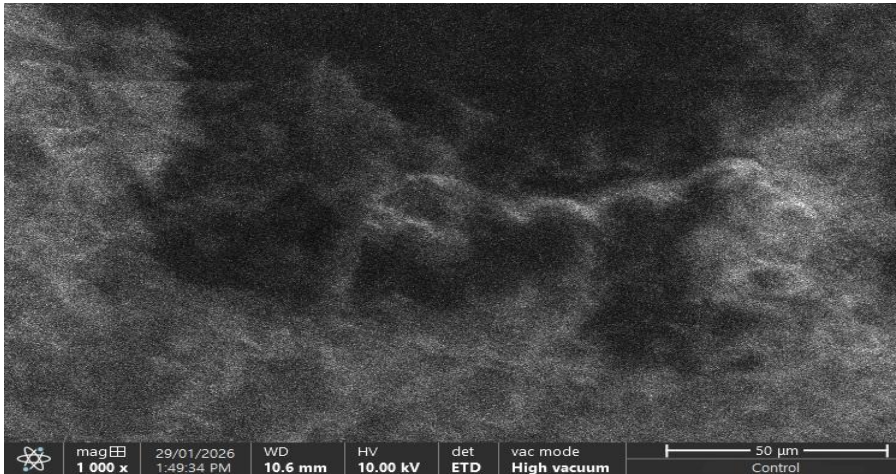


Figure 2: SEM Micrograph of Control Sample (100% REPS) at 1000x Magnification

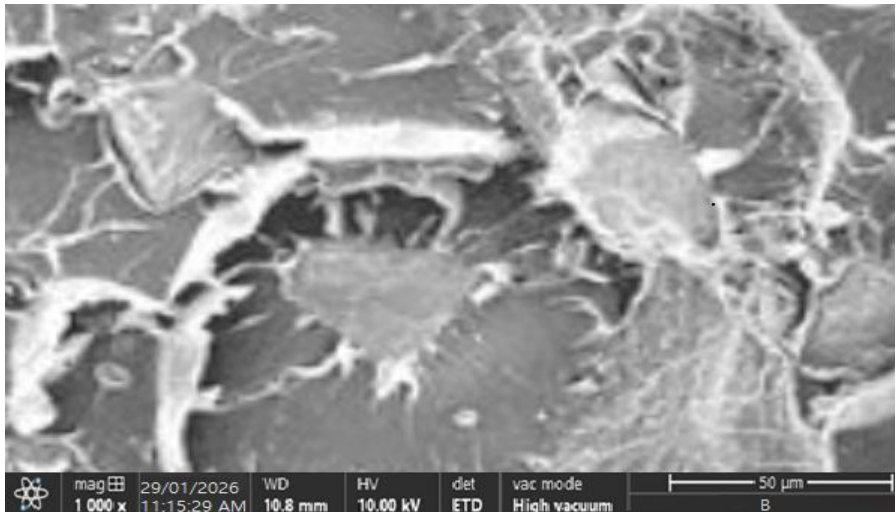


Figure 3: SEM Micrograph of Sample B (REPS/CPP/GNSP: 100-20-30) at 1000x Magnification

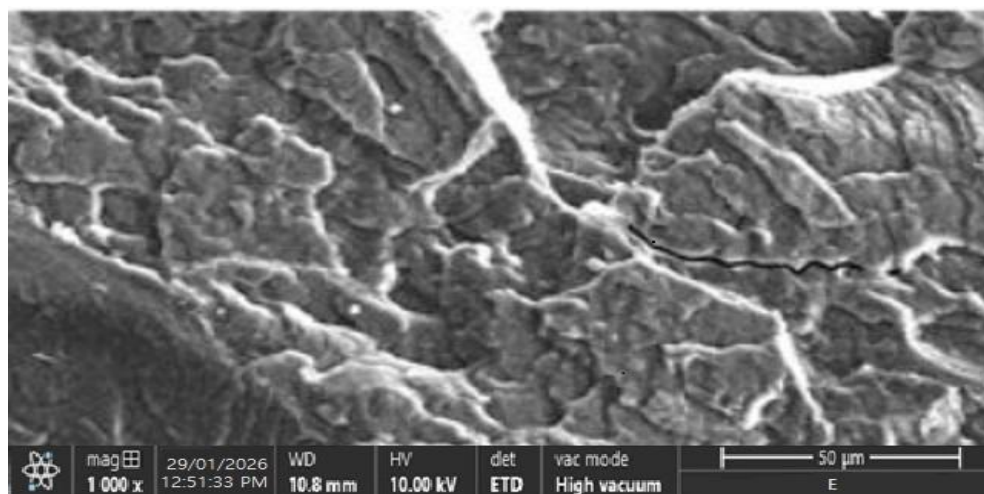


Figure 4: SEM Micrograph of Sample E (REPS/CPP/GNSP: 100-50-30) at 1000x Magnification

The SEM micrographs of the control sample (Figure 2) showed a smooth, homogeneous surface typical of a dense, nonporous structure characteristic of a pure polystyrene matrix. This accounts for its good resistance to moisture ingress, thus rendering it resistant to microbial attack.

Sample B (100:20:30 REPS/CPP/GNSP) (Figure 3) presented relative uniformity in distribution of fillers and bettered interface adhesion due to alkali treatment of the

groundnut shell, which accounts for enhanced tensile performance.

Sample E (100:50:30) (Figure 4) presented pronounced agglomeration, voids, and micro-cracks which can be attributed to bad wetting of the fillers by the polymeric matrix, indicating poor wetting of the filler by the polymer matrix at such high loadings. These voids act as stress concentrators that lower mechanical strength but also increase surface area for microbial colonisation, thereby enhancing biodegradability.

Fourier Transform Infrared (FTIR) Analysis Results

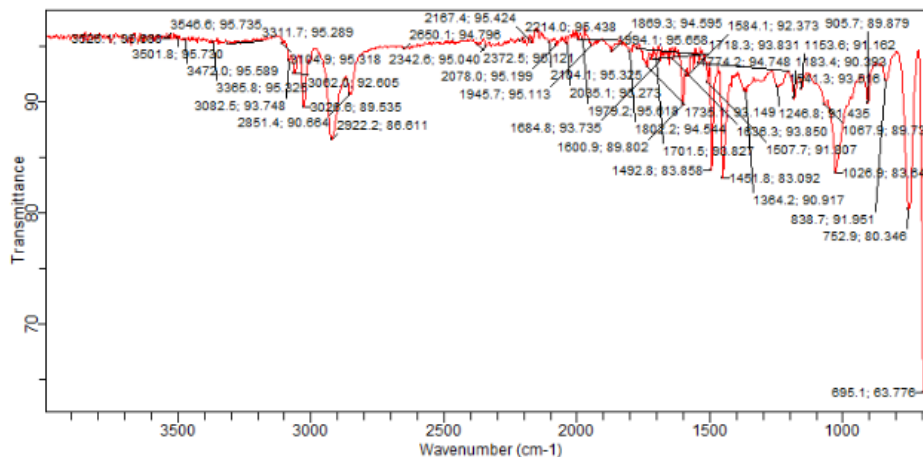
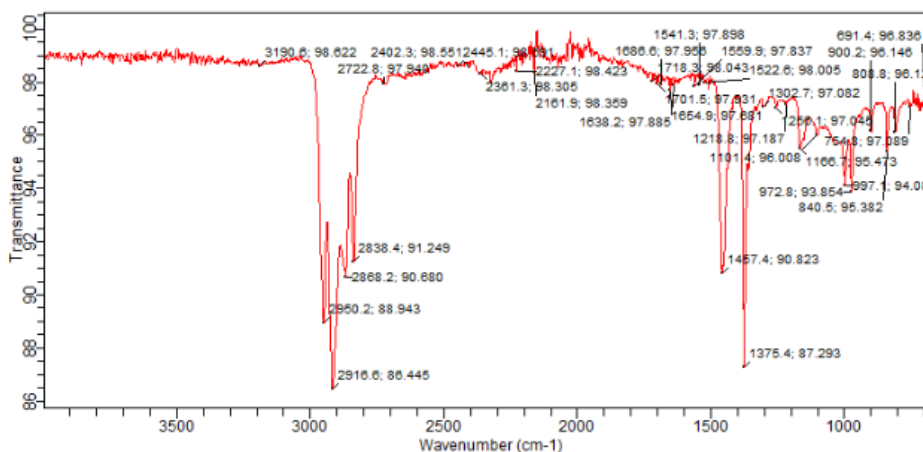
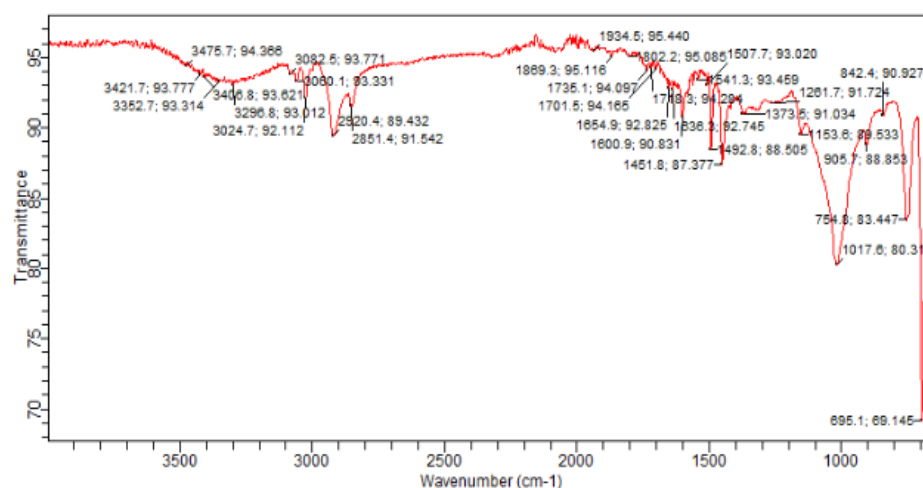
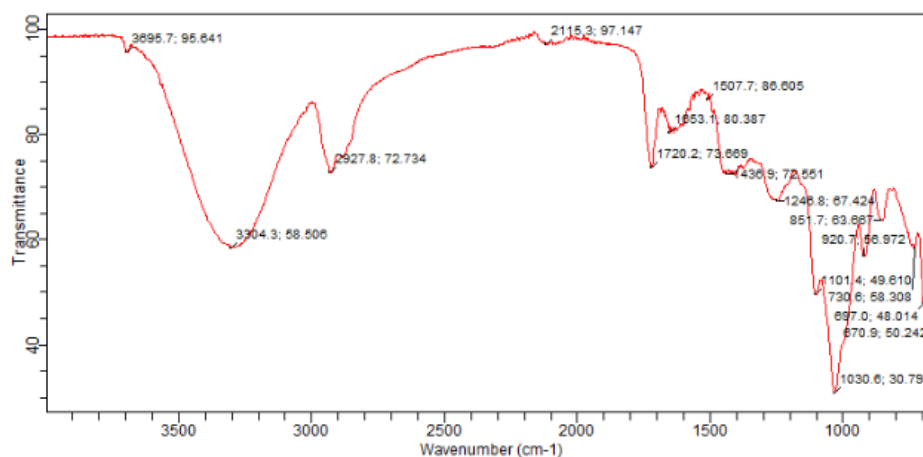


Figure 5: FTIR Spectra for Control Sample (REPS) before Biodegradation



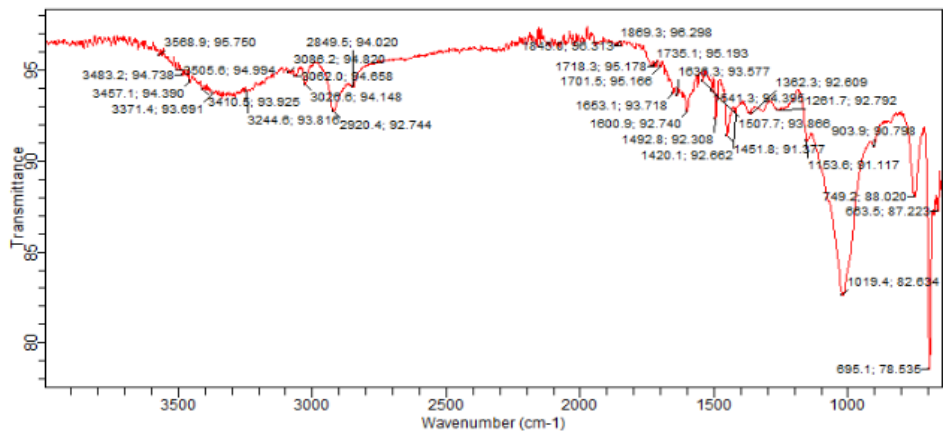


Figure 9: FTIR Spectra for Sample E (REPS/GNSP/CPP) before Biodegradation

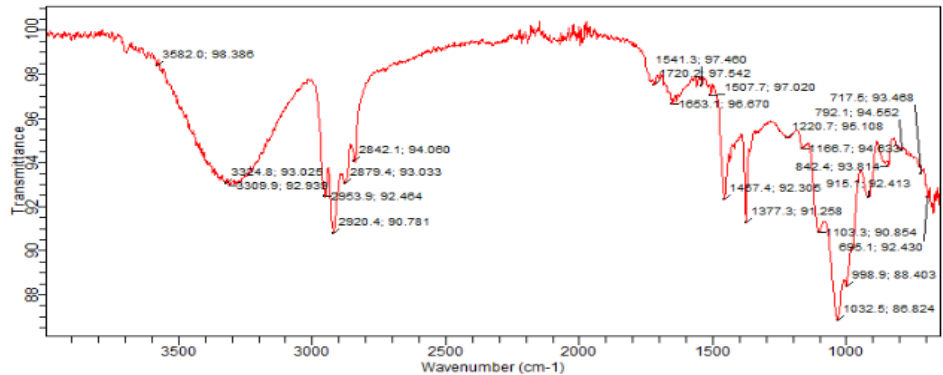


Figure 10: FTIR Spectra for Sample E (REPS/GNSP/CPP) after Biodegradation

The FTIR spectra of the control sample showed typical absorbance peaks of polystyrene at 3020-3080 cm⁻¹ (aromatic C-H), 2920-2850 cm⁻¹ (aliphatic C-H), 1600 and 1490 cm⁻¹ (aromatic C=C) and 760/698 cm⁻¹ (mono-substituted benzene ring). After soil burial, a new broad O-H band appeared near 3304 cm⁻¹ with a C=O band at 1720 cm⁻¹ (Figures 5 and 6). These results suggested the occurrence of new functional groups due to surface oxidation but without any changes to polystyrene fingerprints. Composite Samples C and E displayed strong intensity of O-H, C=O, and C-O bands before degradation (Figures 7

and 9). Presence of such bands in the infrared region suggests the existence of natural fillers in the composite sample. After soil burial (Figures 8 and 10), these bands and the aromatic C-H/ring bands were substantially reduced or lost, most markedly in Sample E, confirming extensive breakdown of both the filler and, to a lesser extent, the polystyrene backbone. The absence of new peaks or major peak shifts indicates that filler-matrix interaction is predominantly physical (physical interlocking and hydrogen bonding) rather than chemical, and that alkali treatment of the groundnut shell improves this physical compatibility.

Tensile Strength, Elastic Modulus and Elongation at Break

Table 2: Tensile Test Results for the REPS/CPP/GNSP Composites

Sample	Tensile Strength (MPa)	Elastic Modulus (MPa)	Elongation at Break (%)
Control	23.82	319.77	11.04
A	27.66	257.31	15.85
B	29.22	285.21	13.65
C	23.26	266.09	11.71
D	25.99	286.02	9.73
E	20.93	220.46	9.74

Tensile strength initially increases from 23.82 MPa (control) to a peak value of 29.22 MPa in Sample B, but then decreases to 20.93 MPa in Sample E. This trend shows

effective stress transfer and good filler dispersion at moderate loading, whereas agglomerated particles at high CPP levels, along with poor interfacial bonding and voids

observed via SEM, reduce tensile strength at higher CPP contents.

Elastic modulus followed a similar variation pattern with the incorporation of filler, increasing with filler addition at lower loadings due to the restriction of polymer chain mobility by the rigid lignocellulosic fillers in the matrix system. Elongation at break decreased progressively from

11.04% (control), with elongation increasing slightly for samples A and B before decreasing for samples D and E, showing a shift toward more brittle behaviour as filler content increased. These results show an optimum filler composition (Sample B) that maximises reinforcement without excessive ductility loss.

Impact Strength

Table 3: Impact Strength Results for the REPS/CPP/GNSP Composites

Sample	Impact Energy (J)			Average Impact Energy (J)	Impact Strength (J/mm)
	1 st	2 nd	3 rd		
Control	0.362	0.368	0.421	0.384	0.12
A	0.554	0.558	0.557	0.556	0.174
B	0.564	0.563	0.563	0.563	0.176
C	0.571	0.588	0.595	0.585	0.183
D	0.546	0.549	0.561	0.552	0.173
E	0.418	0.452	0.412	0.427	0.134

The results showed an increase in impact strength values when compared with the control sample's value of 0.120 J/mm to the maximum of 0.183 J/mm in Sample C, indicating that natural fillers act as crack arresters that increase energy absorption and toughness up to an

optimum loading. Beyond Sample C, impact strength declined (Sample E: 0.134 J/mm) owing to filler agglomeration, poor interfacial bonding, and increased brittleness at higher filler content.

Hardness

Table 4: Hardness Test Results for the REPS/CPP/GNSP Composites

Sample	Hardness (Hv)			Average Hardness (Hv)
	1 st	2 nd	3 rd	
Control	57.8	58.6	59	58.47
A	57.4	57.2	57.5	57.37
B	55.9	55.6	54.6	55.37
C	51.2	51.5	51.4	51.37
D	53	53.3	53.9	53.4
E	47.4	47.2	47.5	47.37

There was a progressive decrease in hardness values (from 58.47 HV (control) to 47.37 HV (Sample E)) with increasing filler content, indicating the softer nature of the lignocellulosic fillers relative to polystyrene, together with increased porosity and reduced intermolecular cohesion

at higher loadings. Although reduced hardness may compromise durability at very high filler content, it benefits flexible packaging applications and correlates with the improved biodegradability observed in the higher-filled samples.

Flexural Strength and Modulus

Table 5: Flexural Test Results for the REPS/CPP/GNSP Composites

Sample	Average Load(N)	Deflection (mm)			Flexural Strength (MPa)	Flexural Modulus (MPa)
		Initial	Final	"D" (Initial - Final)		
Control	73.83	24.64	16.82	7.82	34.61	1475.184
A	86.32	24.29	15.13	9.16	40.46	1404.948
B	84.45	24.16	14.76	9.4	39.59	1403.757
C	80.74	24	13.04	10.96	37.85	2686.93
D	77.24	24.13	12.91	11.22	36.21	1075.646
E	64.02	24.12	15.24	8.88	30.01	1126.478

The flexural strength of samples with a low amount of filler reached its maximum value for sample A (40.46 MPa) before declining at higher filler content, indicating that low filler loading improves bending resistance through effective stress distribution, whereas excessive loading introduces structural defects that weaken the composite.

Flexural modulus, however, was highest in Sample C (2686.93 MPa), suggesting more favourable filler alignment and stress distribution at that composition; this indicates that peak strength and peak stiffness do not necessarily occur at the same filler loading.

Density

Table 6: Density Test Results for the REPS/CPP/GNSP Composites

Sample	Mass (g)	Volume (cm ³)	Obtained Value for Density (g/cm ³)	Density (g/cm ³)
Control	13.421	11.52	1.165885	1.17
A	13.414	11.52	1.081944	1.08
B	13.277	11.52	1.065711	1.07
C	13.146	11.52	1.05434	1.05
D	13.121	11.52	1.043489	1.04
E	13.112	11.52	1.013194	1.01

The density gradually declined from 1.17 g/cm³ (control) to 1.01 g/cm³ (Sample E). This can be ascribed to the introduction of voids from incorporating low-density agro-

waste fillers into the composite system, resulting in lightweight materials suitable for packaging applications.

Water Absorption

Table 7: Water Absorption Results for the REPS/CPP/GNSP Composites (7-Day Immersion)

Sample	Day								Amount of Water Absorbed (g)	Water Absorption (%)
	0	1	2	3	4	5	6	7		
Control	6.132	6.132	6.133	6.133	6.134	6.135	6.135	6.135	0.003	0.05
A	6.096	6.096	6.098	6.101	6.102	6.102	6.102	6.102	0.006	0.1
B	6.12	6.12	6.124	6.127	6.133	6.14	6.142	6.142	0.022	0.36
C	6.186	6.188	6.19	6.192	6.203	6.209	6.21	6.21	0.024	0.39
D	6.162	6.17	6.175	6.176	6.2	6.204	6.204	6.204	0.042	0.68
E	6.186	6.189	6.192	6.198	6.236	6.247	6.25	6.25	0.064	1.03

Water absorption was found to increase progressively as both the immersion period and the filler content increased (from 0.05% in the control to 1.03% in Sample E). This can be attributed to the hydrophilic nature of the lignocellulosic fillers, which introduces more hydroxyl groups and hence porosity in the composite matrix.

Although an increased moisture uptake could hinder their usage in high-moisture applications without appropriate surface treatment, its positive correlation to enhanced biodegradability of the highly filled sample has been highlighted.

Soil-Burial Biodegradability

Table 8: Biodegradability Test Results for the REPS/CPP/GNSP Composites after 63 Days

Sample	Initial Weight 'W ₁ '(g)	Final Weight 'W ₂ '(g)	Change in weight 'W ₁ -W ₂ ' (g)	% weight loss
Control	5.12	5.11	0.01	0.195
A	5.086	5.065	0.021	0.413
B	5.012	4.99	0.022	0.439
C	4.963	4.928	0.035	0.705
D	4.895	4.862	0.033	0.674
E	4.864	4.836	0.028	0.576

A comparison between the two shows an increase in the degree of weight reduction experienced by all samples following 63 days of soil burial from 0.195% for the control to a maximum value of 0.705% in Sample C.

The observed trend indicates improved biodegradation properties among filled composites compared to unfilled ones. This could be due to microorganisms' utilisation of cellulose and hemicellulose constituents found in the agro-waste fillers as a source of carbon. A little bit reduced

percentage weight loss was also observed for sample E than sample C; even though the latter has more filler content, this could be due to high compaction associated with the filling process, thereby limiting microbial access to the composite's matrix.

From the above analyses on biodegradation coupled with studies carried out on both water absorption and scanning electron microscopy (SEM), it can be deduced that addition of cassava peel and groundnut shells powders enhances environmental degradable potentials of recycled polystyrene.

CONCLUSION

The current study has demonstrated successful production and characterization of biodegradable packaging composites from recycled expanded polystyrene filled with groundnut shell and cassava peel powders. Incorporating these natural fillers reduced the thermal stability, density, and hardness of the composites relative to the unfilled control sample, but improved their impact strength, water absorption, and soil-burial biodegradability.

Mechanical properties such as tensile strength and flexural strength were optimized at moderate filler loadings (Samples B and A, respectively), while impact strength, flexural modulus and biodegradation rate peaked in Sample C, beyond which excessive filler content led to agglomeration, void formation, and reduced mechanical soundness, as confirmed by SEM. FTIR analysis.

Alkali treatment of groundnut shell improved interfacial compatibility, amongst others. The resultant lightweight, biodegradable materials could be used as eco-friendly packaging options, paving the way for valorizing agriculture/plastic waste towards achieving circular economy objectives. Subsequent work should explore compatibilizers or surface treatments to further improve interfacial bonding and moisture resistance, alongside pilot-scale production trials and life-cycle assessment to establish commercial viability.

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