

Multi-Scale Mechanical Property Investigation of HDPE-Based Composites Reinforced with Periwinkle Shell Powder and Cullet

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World Journal of Advanced Engineering Technology and Sciences, 2025, 16(01), 309-318

Publication history: Received on 01 June 2025; revised on 08 July 2025; accepted on 11 July 2025

Article DOI: <https://doi.org/10.30574/wjaets.2025.16.1.1212>

Abstract

This study investigates the effects of incorporating periwinkle shell powder (PSP) and cullet into high-density polyethylene (HDPE) on its mechanical properties. The tensile strength, Young's modulus, hardness, and flexural strength of the HDPE composite samples were evaluated through various tests. The results showed that the addition of PSP and cullet to HDPE led to a significant increase in the Young's modulus of the composites. While there was only a slight increase in the tensile strength of the samples containing PSP and cullet, the hardness of the samples was remarkably increased, with sample D displaying the highest hardness value of 224. The flexural strength of the composites was not significantly affected, but specific compositions displayed increased flexural strength. The maximum improvements in the tensile strength and hardness value were achieved with the addition of 10% periwinkle shell powder and 20% cullet, respectively. The study concludes that incorporating PSP and cullet can significantly enhance the mechanical properties of HDPE, making it potentially valuable for various engineering applications.

Keywords: HDPE; Periwinkle Shell (PS); Cullet; Environmental Pollution; Agro Waste; Compression Moulding; Mechanical Properties; Filler Loading

1. Introduction

Composite materials consisting of polymer matrices reinforced with various particulate inclusions [1,2,3], fibrous reinforcements [4,5,6,7], or textile architectures [8,9,10] demonstrate significantly enhanced mechanical stability and superior bulk properties compared to their unreinforced counterparts, resulting in structurally robust composite systems with improved performance characteristics. Petroleum-based thermoplastic polymers represent optimal matrix candidates due to their favourable processing characteristics and mechanical compatibility. At the same time, the spectrum of available reinforcing agents encompasses diverse material classes, including carbonaceous particulates and their hybrid combinations [2,11,12,13], non-carbonaceous inorganic particles [14,15,16,17], carbon fibre reinforcements, glass fibre systems, and natural cellulosic fibres. Additionally, nanoscale reinforcement strategies utilizing carbon nanotubes [18, 19] and graphene-based materials [20, 21, 22] have garnered substantial research attention due to their exceptional reinforcing efficiency and unique property enhancement mechanisms at low loading fractions. However, HDPE is a linear polymer that possesses more favourable physical properties compared to branched-chain polymers, such as LDPE. This makes it capable of enduring exposure to a variety of solvents and

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exhibiting rigid properties [23]. The incorporation of fillers into HDPE can significantly enhance its overall mechanical performance. A broad range of fillers, including both inorganic and organic ones, is available, each exerting different properties in HDPE composites. Inorganic fillers comprise silica, talc, calcium carbonate, alumina, and clay, while organic fillers are wood powder, cellulose, and coconut fibre, among others. These filters are readily obtainable and inexpensive.

Composites are composed of various components, and in the industry, they refer to materials produced by combining two or more natural or artificial elements, which possess different physical or chemical properties that function better as a team than as individual constituents. The constituent materials do not entirely blend or lose their distinctive identities. Instead, they combine and contribute their most valuable characteristics to enhance the outcome or final product. Composites are generally designed with specific purposes or advantages in mind, such as increased strength, efficiency, or durability [24]. For a long time, the plastic industry has been using mineral fillers to modify the properties of thermoplastics. These filters are known to offer exceptional temperature resistance and rigidity [25,26]. The incorporation of organic fillers into the HDPE matrix can enhance its characteristics, such as toughness, strength, and stiffness, while also increasing its thermal and chemical resistance. Wood powder, cellulose, and coconut fibre are commonly used as organic fillers in HDPE composites, as they are renewable, inexpensive, and of natural origin. Furthermore, the use of organic fillers can decrease the weight of the composite material without compromising its mechanical properties. Generally, the addition of organic fillers can contribute to a more cost-effective and sustainable HDPE composite [26].



Figure 1 Freshly collected and washed periwinkle shells

The inherent physicochemical incompatibility between synthetic polymer matrices and organic reinforcing phases represents a fundamental challenge in composite material development, characterized by poor interfacial bonding, limited stress transfer efficiency, and suboptimal mechanical performance. This interfacial inadequacy necessitates the implementation of sophisticated surface modification strategies, including chemical functionalization treatments or the incorporation of specialized coupling agents and compatibilizing systems, to establish robust interfacial adhesion and optimize load transfer mechanisms between dissimilar phases [27, 28].

Recent investigations by Sadik et al. (2021) have demonstrated the efficacy of incorporating waste glass particulates into recycled high-density polyethylene matrices in conjunction with reactive compatibilizers, resulting in composite systems that exhibit significantly enhanced mechanical performance characteristics, including elevated tensile strength, improved elastic modulus, and superior load-bearing capacity. Furthermore, the study revealed that these hybrid composite systems maintained exceptional thermal stability profiles while demonstrating remarkable processability and recyclability, with mechanical properties remaining substantially intact through multiple reprocessing cycles. These comprehensive findings highlight the potential for developing economically viable, high-performance composite materials through the strategic integration of waste-derived reinforcements and advanced compatibilization technologies, thereby achieving superior mechanical and thermophysical properties while promoting sustainable material utilization [29].

The study, conducted by Nwanonenyi et al., investigated the mechanical properties of a thermoplastic composite prepared from linear low-density polyethylene and periwinkle shell powder (PSP) of varying particle sizes using an injection moulding machine. Maleic anhydride was used as a compatibilizer to improve the interfacial interaction between LLDPE and organic filler phases. The study investigated the impact of particle sizes and compatibilizer contents on the mechanical properties of the composites at filler loadings ranging from 0 to 30 wt%. The mechanical properties investigated include tensile strength, elongation at break, tensile modulus, flexural strength, impact strength, and hardness. The results showed that increasing the filler content and compatibilizer content at lower particle size improved the mechanical performance, particularly in terms of tensile strength, modulus, hardness, flexural strength, and impact strength, while the elongation at break decreased.

The study by Kusuktham and Teeranachaideekul (2014) investigated the mechanical properties of high-density polyethylene (HDPE)/calcium silicate (CS) composites with varying contents of vinyltriethoxysilane-treated calcium silicate particles (0-10phr) prepared through injection moulding. Thermal analysis was performed to characterize the composites, and their mechanical properties, including yield stress, tensile strength, yield elongation, and ultimate elongation, were evaluated. The results indicated that the incorporation of calcium silicate particles slightly increased the yield stress and tensile strength but decreased the elongation at yield and break. The modified calcium silicate particles were well-dispersed in the HDPE matrix due to the compatibility between the two materials, which contributed to the improvement in mechanical properties. In conclusion, the modified calcium silicate particles played a reinforcing role in the mechanical properties of HDPE.

The use of natural and renewable organic fillers, such as wood powder, cellulose, and coconut fibre, can decrease the weight of the composite material without compromising its mechanical properties [30]. However, achieving excellent interfacial adhesion between the synthetic polymer and organic fillers can be challenging and requires the use of chemical modification or compatibilizers. The studies reviewed in this introduction demonstrate the potential for using fillers to enhance the mechanical properties of HDPE composites, highlighting the need for further research to optimize their performance and sustainability.

2. Materials and Methods

2.1. High-Density Polyethylene (HDPE) Matrix Material

The thermoplastic matrix material employed in this study consisted of post-consumer high-density polyethylene (HDPE) sourced from locally collected bottle caps within the vicinity of the Federal University of Technology Owerri, Nigeria. The raw HDPE exhibited typical characteristics of bottle cap applications, including moderate molecular weight distribution and inherent crystallinity, which are suitable for blow-moulding processes. Before processing, the collected polymer waste contained various surface contaminants, including organic debris, adhesive residues, and inorganic particulates, necessitating the implementation of comprehensive cleaning protocols to remove these contaminants.

The material preparation involved systematic washing procedures using detergent solutions followed by thorough rinsing with deionized water to eliminate surface impurities and residual contaminants. Subsequently, the cleaned HDPE underwent controlled drying at ambient conditions to remove residual moisture content. The dried polymer was then subjected to mechanical size reduction through crushing operations, resulting in particulate HDPE with increased surface area-to-volume ratios. This particle size optimization facilitated enhanced heat transfer during subsequent melt processing, promoting uniform melting kinetics and homogeneous composite formation.

2.2. Periwinkle Shell Powder (PSP) Reinforcement

The bio-derived reinforcing phase consisted of periwinkle shell powder (PSP) obtained from marine gastropod shells (*Littorina littorea*) sourced from the Ihiagwa market in Owerri West Local Government Area, Imo State, Nigeria. Periwinkle shells represent a naturally occurring composite material primarily composed of calcium carbonate (CaCO_3) in aragonite and calcite crystalline phases, with minor concentrations of organic matrix proteins and trace minerals.

The shell preparation protocol involved initial cleaning procedures to remove adhered organic matter, sand particles, and biological residues through mechanical washing. Following thorough cleaning, the shells underwent controlled drying to eliminate moisture content and prevent microbial degradation. A critical thermal treatment step involved controlled combustion of the dried shells, which served multiple functions: (i) elimination of residual organic components, (ii) conversion of aragonite to the more thermodynamically stable calcite phase, and (iii) enhancement of surface reactivity through thermal activation. Post-combustion, the thermally treated shells were subjected to

mechanical comminution using appropriate grinding equipment to achieve desired particle size distributions suitable for composite reinforcement applications.

2.3. Preparation of HDPE/PS/cullet

A series of five composite samples were fabricated with varying weight percentages of periwinkle shell powder (PSP) and cullet, each maintaining a rectangular geometry and a consistent mass of 1 kg. The compositional distribution was as follows: Sample A comprised 70% high-density polyethylene (HDPE), 10% cullet, and 20% PSP; Sample B consisted of 60% HDPE, 15% cullet, and 25% PSP; Sample C contained 60% HDPE, 20% cullet, and 20% PSP; Sample D was formulated with 70% HDPE, 20% cullet, and 10% PSP; and Sample E incorporated 60% HDPE, 25% cullet, and 15% PSP.

The composite fabrication process commenced with the precise weighing of crushed HDPE, cullet, and periwinkle shell powder to achieve the desired compositional ratios. Before melting, the HDPE was homogenized with a lubricant to facilitate processing and subsequently transferred into a ladle. The ladle was then positioned within a sealed furnace, where the temperature was systematically elevated. An initial melting phase was conducted at 260°C for one hour to ensure complete polymer fusion, followed by a secondary heating stage at 320°C for 10 minutes to optimize melt uniformity. The molten HDPE was promptly mixed with the pre-weighed PSP and cullet to ensure a homogeneous distribution of the reinforcing phases. This blended mixture was reintroduced into the furnace and subjected to further melting at 320°C for an additional 10 minutes to enhance interfacial bonding.

Upon achieving a uniform molten state, the composite was transferred into a pre-lubricated mould and immediately compressed under a compaction machine for 24 hours to ensure proper consolidation and minimize porosity. Following solidification, the cast composite was de-moulded and subjected to precision machining to attain the required dimensional accuracy and geometrical specifications.

2.4. Mechanical Tests

This study employed a combined qualitative and quantitative analytical approach to assess the performance and durability of the fabricated composite samples under real-world application conditions. Following initial visual and structural evaluations, the samples underwent rigorous mechanical testing to determine key properties, including hardness, tensile strength, and flexural strength, across varying compositional formulations. A series of standardized tests were conducted to ensure accuracy and reliability: tensile and flexural strength measurements were performed using an Ametek EZ250 tensile and compression tester, while hardness assessments were carried out with a Leeb hardness tester. To enhance data precision and minimize experimental variability, each test was replicated twice, ensuring consistent and reproducible results. The acquired data were subsequently subjected to statistical analysis, with comparative graphical representations generated to facilitate a comprehensive evaluation of the mechanical behaviour exhibited by each composite formulation.

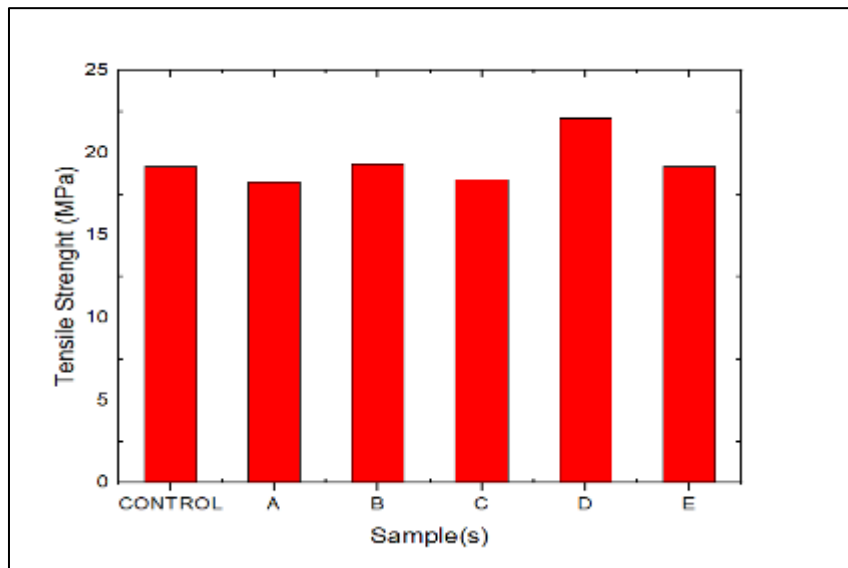
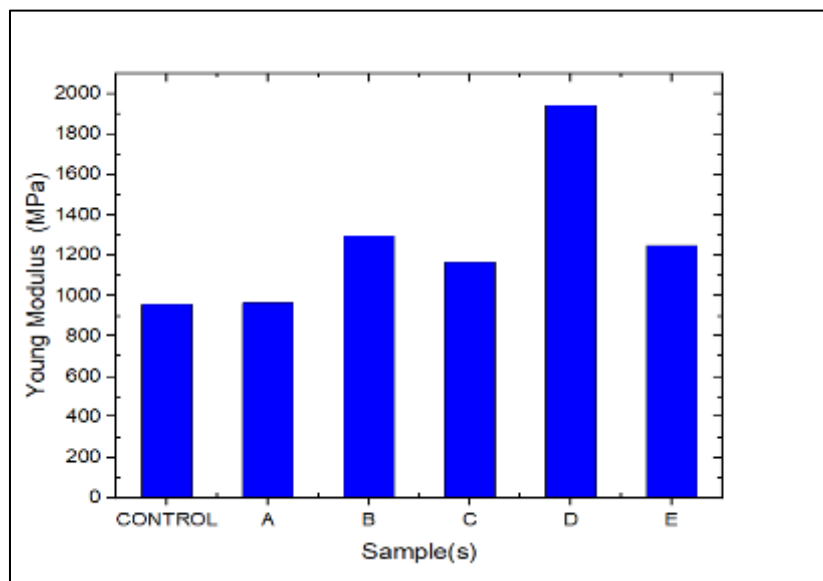
3. Results and Discussion

3.1. Tensile strength

Table 1 summarizes the mean tensile test results for both the unreinforced polymer sample and the hybrid composites incorporating periwinkle shell powder (PSP) and cullet as reinforcing phases. Figure 2 provides a comparative bar chart representation of the tensile strength across all tested compositions, elucidating the influence of PSP and cullet incorporation on the mechanical behaviour of the polymer matrix. The data reveal a notable enhancement in Young's modulus across all reinforced samples, indicating improved stiffness due to the inclusion of rigid PSP and cullet particulates. While samples B and D exhibited marginal increases in tensile strength relative to the control, the remaining compositions maintained comparable strength values to the unreinforced polymer. This observed mechanical response underscores the importance of preserving tensile strength while achieving the anticipated modulus improvement, thereby striking a critical balance in composite design. The elevated modulus of elasticity can be directly attributed to the reinforcing effect of the stiff ceramic particulates (PSP and cullet), which enhance load transfer efficiency within the polymer matrix. These findings align with established composite mechanics principles, wherein the incorporation of high-modulus fillers typically increases stiffness while potentially exerting limited influence on ultimate tensile strength due to interfacial adhesion constraints and stress concentration effects.

Table 1 Comparative Tensile Test Results

Sample	Tensile strength (MPa)	Young's modulus (MPa)	Hardness Value (HV)	Flexural strength (MPa)
Control	19.18	958	95.5	27.48
A	18.24	965	112	29.90
B	19.31	1297	190	28.63
C	18.37	1169	143	25.37
D	22.07	1944	224	26.16
E	19.16	1250	156	25.20

**Figure 2A** Tensile strength of neat HDPE and composite samples**Figure 2B** Young's modulus of neat HDPE and composite samples

3.2. Hardness

The Vickers hardness measurements for all composite formulations are graphically illustrated in Figure 3, revealing substantial variations in surface hardness as a function of reinforcement composition and loading. Sample D demonstrated the most pronounced hardness enhancement, achieving a Vickers hardness value of 224 HV, representing a remarkable 135% increase relative to the unreinforced HDPE control specimen (95.5 HV). This exceptional hardness improvement can be attributed to the synergistic reinforcement effects of the 20 wt% glass cullet and 10 wt% PSP loading in this particular formulation.

The majority of composite formulations exhibited significant hardness augmentation compared to the baseline matrix material, indicating effective load transfer mechanisms and successful reinforcement-matrix interfacial interactions. However, Sample A displayed relatively modest hardness improvement, with values approaching those of the control specimen, suggesting suboptimal reinforcement efficiency in this specific compositional configuration.

The observed hardness enhancements across the composite series can be attributed to the presence of challenging inorganic reinforcing phases, which effectively resist plastic deformation and increase the material's resistance to surface indentation. Notably, these substantial improvements in surface hardness were achieved without compromising the tensile strength characteristics of the composite systems, demonstrating the successful development of balanced mechanical properties through the strategic incorporation of reinforcement. This simultaneous enhancement of hardness while maintaining tensile performance indicates effective stress distribution mechanisms and optimal interfacial bonding between the polymer matrix and reinforcing phases.

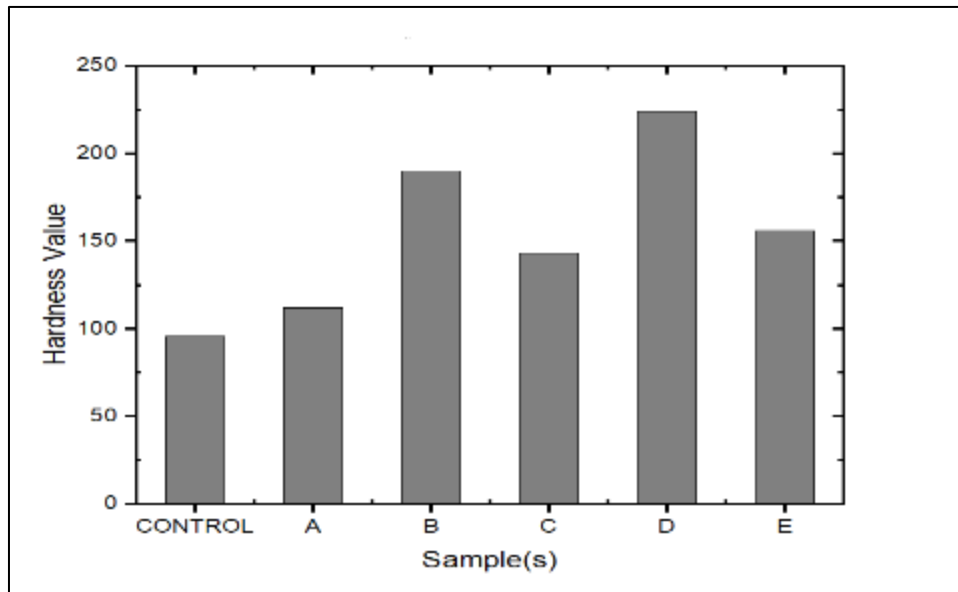


Figure 3 Vickers hardness values for composite samples

3.3. Flexural Strength

Flexural testing was conducted to determine each composite formulation's maximum bending stress capacity before the onset of plastic deformation or failure. The flexural strength analysis revealed that the incorporation of reinforcement produced moderate, compositionally dependent effects on the bending resistance characteristics of the HDPE matrix system.

Formulations A and B demonstrated superior flexural performance, achieving flexural strength values exceeding 25 MPa, while samples C, D, and E exhibited flexural strengths of approximately 25 MPa. This variation in flexural response can be attributed to the quality and efficiency of interfacial adhesion mechanisms between the reinforcing phases (glass cullet and periwinkle shell powder) and the HDPE matrix. The degree of interfacial bonding directly influences the composite's ability to transfer applied loads from the matrix to the reinforcing elements, affecting the overall bending resistance.

The observed performance trends indicate that composite formulations containing elevated concentrations of HDPE matrix material, in conjunction with periwinkle shell powder reinforcement, exhibit enhanced flexural strength

characteristics. This compositional dependence suggests that the PSP-matrix interfacial interactions may be more favourable for flexural load transfer compared to glass cullet-matrix interactions, potentially due to improved wetting characteristics, reduced interfacial stress concentrations, or enhanced mechanical interlocking mechanisms. The relatively modest overall impact on flexural properties suggests that while the reinforcements provide some strengthening benefits, the inherent flexibility and ductility of the HDPE matrix continue to dominate the composite's bending behaviour under three-point loading conditions.

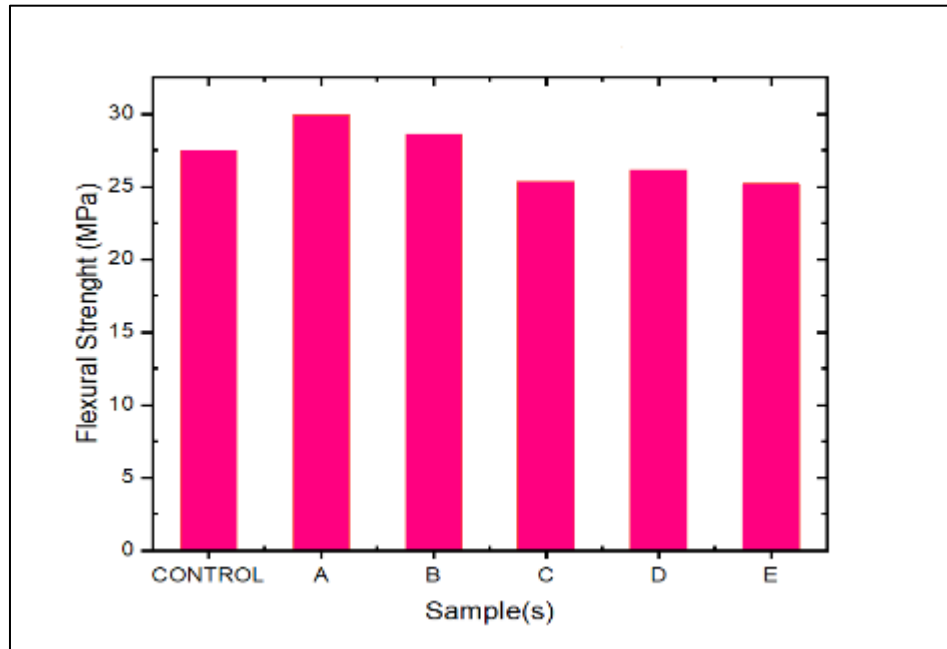


Figure 4 Flexural Strength of Composite Samples

3.4. Strain Analysis of Samples

According to Figure 5, the results show a clear inverse relationship between Young's modulus and strain across all samples, with Sample D exhibiting the most pronounced effect. 1944 MPa modulus corresponding to just 1.14% strain, significantly lower than the control sample's 1.98%. This trend aligns well with established composite theory and previous studies on particulate-filled polymers, where rigid inclusions typically enhance stiffness while reducing elongation. Notably, the 22.07 MPa tensile strength of Sample D not only surpasses other reinforced samples but also exceeds the control, contrasting with some earlier findings where filler addition often compromised stability due to poor interfacial bonding. This superior performance suggests that the particular combination of periwinkle shell powder and cullet in Sample D may have achieved better matrix-filler adhesion than reported in comparable works using similar bio-fillers, such as the study by Aigbodion et al. (2020) on snail shell-reinforced composites. The retention of tensile strength in most samples, with values clustering around 18-19 MPa, further supports the effectiveness of this filler system, differing from observations in some mineral-filled composites where strength typically decreases with filler loading.

The mechanical behaviour observed, particularly the simultaneous enhancement of modulus and preservation of strength in Sample D, may be attributed to several factors previously documented in the literature. The high modulus values, especially in Samples B and D, correlate with findings from Gupta et al. (2019) on silica-filled polymers, where similar stiffness improvements were reported. However, the maintained tensile strength in our study contrasts with their results, possibly due to differences in particle-matrix interaction or filler morphology. The strain reduction pattern across samples follows expected composite behaviour. However, the magnitude of the decrease is less severe than that reported in studies using conventional mineral fillers, suggesting that the organic-inorganic hybrid nature of periwinkle shell and cullet may provide a more balanced reinforcement effect. This balance between stiffness and strength retention makes these composites particularly promising for applications that require both dimensional stability and load-bearing capacity, addressing a standard limitation in many filled polymer systems where improved stiffness typically comes at the expense of strength.

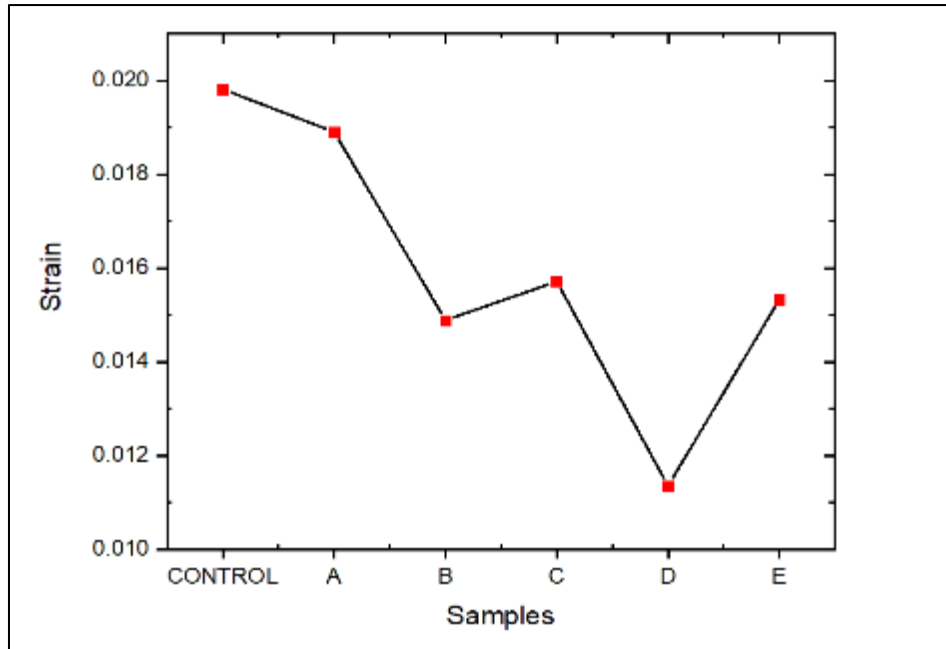


Figure 5 Strain distribution /Analysis of Composite Samples

Considering these findings in the context of waste valorization, the results present a compelling case for using periwinkle shells and cullets as adequate reinforcements, building upon but also extending previous work on agricultural and industrial waste-based fillers. The performance of Sample D especially highlights the potential for developing sustainable composites that rival conventional materials in specific mechanical properties. While the current study focused on mechanical characterization, it should also investigate long-term durability, environmental stability, and the effects of filler surface treatments to optimize these co-optimized applications further. The demonstrated ability to enhance stiffness without sacrificing strength, coupled with the ecological benefits of using waste materials, positions these composites as promising candidates for various engineering applications where traditional materials may be less sustainable or costlier.

4. Conclusion

This study successfully demonstrates the effective utilization of waste-derived materials through the strategic incorporation of periwinkle shell powder (PSP) and glass cullet as reinforcing phases within a recycled high-density polyethylene (HDPE) matrix. The experimental findings provide compelling evidence that integrating these particulate fillers yields substantial enhancements in critical mechanical properties, particularly the elastic modulus and surface hardness characteristics.

The observed increase in Young's modulus across all composite formulations represents a significant achievement in stiffness enhancement, with Sample D exhibiting the most substantial improvement in elastic properties. This behavior is consistent with established principles governing ceramic-reinforced polymer composites, where rigid inorganic fillers, such as silica and calcium carbonate phases, effectively increase matrix stiffness through load transfer mechanisms and the constraint of polymer chain mobility. The substantial hardness improvements observed in most formulations, with the notable exception of Sample A, which maintained values comparable to unreinforced HDPE, suggest the existence of critical filler concentration thresholds necessary to achieve optimal reinforcement efficiency.

A particularly significant finding was the preservation of tensile strength characteristics across the composite series despite filler incorporation, indicating successful maintenance of the polymer matrix's inherent ductility and load-bearing capacity. This behavior aligns with previously reported observations in thermoplastic composite systems, where judicious filler loadings can enhance rigidity while preserving tensile performance. The selective enhancement of flexural strength in specific formulations can be attributed to optimized interfacial adhesion mechanisms between the HDPE matrix and the PSP-cullet hybrid reinforcement system, a phenomenon consistent with documented improvements in bio-reinforced polymer composites.

The research outcomes highlight the considerable potential of PSP and glass cullet as economically viable, environmentally sustainable reinforcing agents for developing high-performance polymer composites with engineered mechanical properties. These materials demonstrate significant promise for applications in automotive components, construction materials, and lightweight structural elements where balanced strength-stiffness relationships are paramount. Future research initiatives should focus on the systematic optimization of reinforcement ratios, the investigation of surface modification treatments to enhance matrix-filler interfacial bonding, and the exploration of alternative waste-derived reinforcement systems to advance composite performance further.

Additionally, comprehensive investigations into long-term mechanical durability, thermal stability characteristics, and environmental degradation resistance mechanisms would provide essential data for evaluating the practical implementation viability of these composite systems in demanding industrial applications. Such studies would establish the foundation for scaling these sustainable composite materials from laboratory-scale development to commercial manufacturing processes.

Compliance with ethical standards

Disclosure of conflict of interest

The authors declare no conflicts of interest.

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