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Development and Characterisation of Plantain Peel Flour (PPF) Reinforced Polyethylene Glycol (PEG) Composites

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ABSTRACT

This study investigates the development and characterization of plantain peel flour (PPF)-reinforced polyethylene glycol (PEG) composites. PPF, derived from agricultural waste, offers a sustainable alternative for composite reinforcement due to its eco-friendliness, biodegradability, and mechanical properties. Composites were fabricated with varying filler loadings (6%, 12%, 18%, 24%, and 30% PPF), and their mechanical and thermal properties were evaluated. Results indicate that the tensile strength improved up to 18% PPF loading significantly, achieving a maximum of 45.58 MPa, representing a 258% improvement over neat PEG. However, further increases in PPF content led to a decline in mechanical performance due to reduced interfacial compatibility and filler dispersion. Thermogravimetric analysis revealed improved thermal stability with higher PPF content, attributed to the enhanced char formation. Theoretical modelling using the Rule of Mixtures, Halpin-Tsai, and Hirsch models was employed to predict the mechanical properties, with varying degrees of accuracy. This study underscores the potential of PPF as a renewable reinforcement in polymer composites, paving the way for sustainable material development.

Keywords: *Plantain peel flour (PPF), Polyethylene glycol (PEG) composites, Sustainable alternative. Filler loadings, Renewable reinforcement, Sustainable material development.*

I. INTRODUCTION

Agricultural waste management poses significant environmental and socio-economic challenges, particularly in developing countries where efficient waste utilization strategies remain underdeveloped. Among the various forms of agricultural waste, plantain peels represent a notable contributor due to their widespread availability and underutilization. Plantain (Musa paradisiaca), a vital staple food in many tropical and subtropical regions, accounts for approximately 35% of waste generation in the form of peels during processing and consumption [1]. With global production of plantains exceeding 40 million tons annually, the resulting peel waste represents millions of tons of discarded biomass that exacerbates environmental degradation through improper disposal. [2-3]

Despite their classification as waste, plantain peels possess valuable biochemical properties, including high levels of dietary fibre, starch, antioxidants, and lignocellulosic content, which make them promising candidates for value-added applications [4-5]. However, these resources remain largely untapped, underscoring the need for innovative solutions to transform plantain peels into sustainable materials. Research efforts have demonstrated the potential of plantain peel derivatives in various domains, such as food products, biodegradable films, and now, polymer composites. [6-8]

In polymer composites, sustainable material development has increasingly focused on integrating natural fillers into synthetic polymer matrices to enhance mechanical and thermal properties while reducing environmental impact. [9-11] Polyethylene glycol (PEG), a versatile polymer widely used for its biocompatibility, flexibility, and water solubility, is a prime candidate for such modifications. However, its low mechanical strength limits its broader application in structural or load-bearing contexts [12]. Reinforcing PEG with natural fillers such as plantain peel flour (PPF) offers a promising pathway to address this limitation. The incorporation of PPF into PEG has the potential to improve the polymer's tensile strength, modulus, and thermal stability, while simultaneously contributing to agricultural waste valorization. [13,14]

Previous studies have demonstrated the successful integration of agricultural by-products like starch, cellulose, and lignin into polymer composites, improving material performance and biodegradability [15,16]. However, challenges remain in optimizing filler dispersion, interfacial bonding, and thermal processing stability, which can adversely impact the mechanical and thermal properties of the composites.[17] In this context, plantain peel flour, with its rich hydroxyl group content and fibre-based structure, emerges as a viable reinforcement material for PEG, capable of establishing strong hydrogen bonding within the polymer matrix.[18]

This study explores the development and characterization of PPF-reinforced PEG composites, focusing on the mechanical and thermal behavior of the resulting materials at varying filler loadings. By addressing critical challenges such as filler dispersion and interfacial adhesion, the research seeks to optimize composite performance and evaluate the theoretical predictability of material properties using established models like the Rule of Mixtures, Halpin-Tsai, and Hirsch models. In doing so, this work not only advances the understanding of plantain peel utilization in polymer composites but also contributes to the growing field of green and sustainable material engineering [19,20].

II. Materials And Methods

A. Materials

Plantain peels were sourced locally from markets in Cross River State, Nigeria, and thoroughly cleaned to remove impurities. PEG (Molecular weight: 3600-4400, density: 1.2 g/cm³) was supplied by Sigma Aldrich. Other reagents included potassium hydroxide (KOH), hydrochloric acid (HCl), and hydrogen peroxide (H2O2), all of analytical grade.

FIGURE 1: PLANTAIN MATERIALS

B. Preparation of Plantain Peel Flour (PPF)

The preparation of plantain peel flour (PPF) involves a systematic process to ensure the production of high-quality, uniform filler material suitable for composite reinforcement.

Initially, freshly collected plantain peels were washed thoroughly using distilled water and detergent to remove surface dirt, organic residues, and other impurities. The washing process was followed by multiple rinses with clean distilled water to eliminate any residual detergent, ensuring the peels were free from contaminants that could affect subsequent processing or chemical treatments.

The cleaned peels were subjected to a two-stage drying process. First, they were sun-dried for two weeks, allowing the moisture content to reduce gradually under ambient conditions until a constant weight was achieved. This initial drying step is critical for removing bulk moisture while preventing microbial growth. Subsequently, the peels were oven-dried at 50°C for 24 hours to ensure complete dehydration, which is essential for preventing enzymatic activity and facilitating efficient milling.

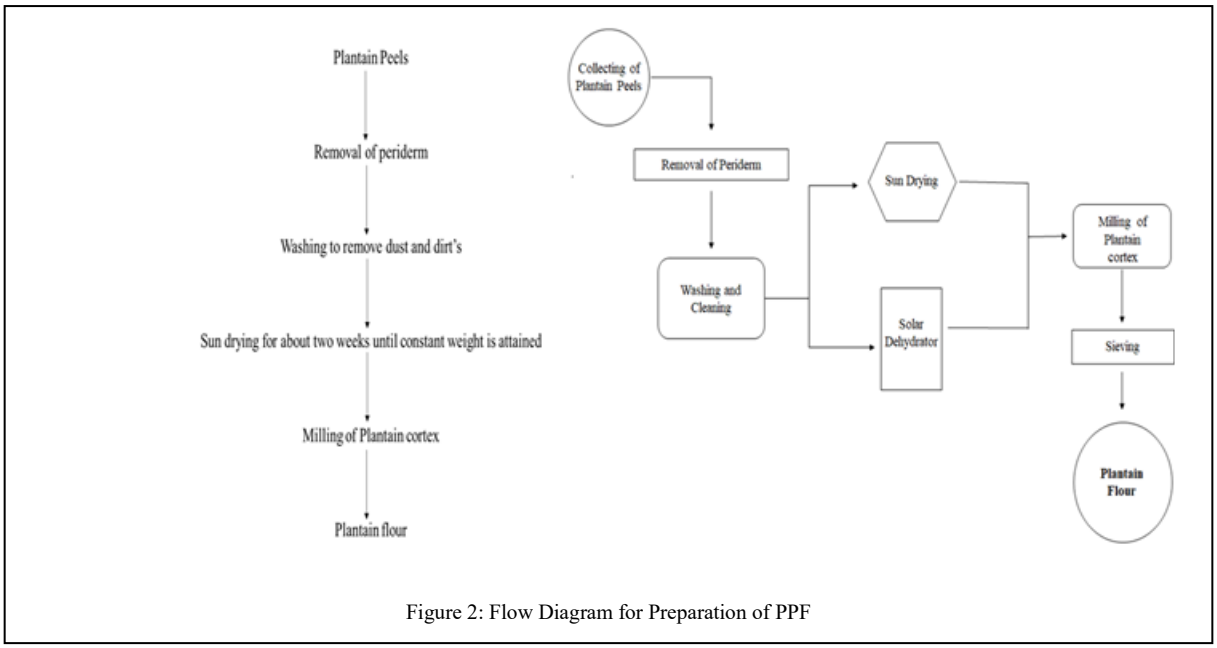

Figure 2: Flow Diagram for Preparation of PPF

The dried plantain peels were milled into a fine powder using a mechanical grinder, a step that ensures the production of uniformly sized particles. The flour was then sieved to obtain a consistent particle size distribution, a crucial factor for achieving uniform filler dispersion in the polymer matrix.

To enhance the functionality of the PPF, chemical treatment was conducted. The flour was soaked in a potassium hydroxide (KOH) solution at a 2:5 mass ratio, maintained at 50°C for 3 hours. This alkaline treatment serves to remove lignin, hemicellulose, and other non-cellulosic components, thereby increasing the surface area and the availability of hydroxyl groups for potential interactions with the polymer matrix.[21]. Following alkaline treatment, the slurry was neutralized using 0.5N hydrochloric acid (HCl) and washed repeatedly with distilled water until the pH stabilized at 6.5.

Surface modification of the PPF was further carried out using hydrogen peroxide (H_2O_2) treatment. The flour was soaked in a 30 wt.% H_2O_2 solution to enhance its hydrophilicity and to introduce functional groups that improve interfacial bonding with the polymer matrix [22]. After the treatment, the PPF was dried at 110°C for 24 hours to remove residual moisture and stored in a desiccator to maintain stability and prevent moisture absorption before composite preparation.

The detailed preparation process ensured the production of chemically modified PPF with improved thermal stability, reduced impurities, and enhanced interfacial compatibility, making it a suitable reinforcement material for PEG-based composite systems.

C. Composite Fabrication

The fabrication of plantain peel flour (PPF)-reinforced polyethylene glycol (PEG) composites was performed using a straightforward and cost-effective hand lay-up technique. This method was chosen for its simplicity and efficiency in producing composite materials with varying filler content, which is particularly suitable for initial studies and small-scale fabrication (John & Thomas, 2008).

The process involved carefully mixing PPF and PEG in predetermined ratios of 6%, 12%, 18%, 24%, and 30% by weight. These ratios were selected to investigate the influence of filler loading on the mechanical and thermal properties of the composites, following approaches used in similar studies on natural fibre-reinforced composites. [23, [24]]. The uniform distribution of PPF in the PEG matrix was achieved by manual stirring, ensuring consistent dispersion of the filler particles. This step is critical as the mechanical properties of natural fibre composites strongly depend on the even distribution and interfacial adhesion of the filler material.

After mixing, the composite mixture was poured into pre-cleaned moulds to shape the materials into test specimens. The moulds were designed to conform to the dimensions required for subsequent mechanical and thermal testing, in line with ASTM D-638 standards for tensile testing. The filled moulds were left to cure at room temperature for 48 hours. Room-temperature curing was selected as it

minimizes thermal degradation of the natural filler and preserves the integrity of the composite structure, as noted in other studies involving plantain-based composites.

Following curing, the composites were carefully demolded to avoid introducing surface defects that could impact the properties of the specimens. The cured samples were then cut into standardized shapes and dimensions using a precision cutting tool to prepare them for characterization. Proper preparation of the test specimens is essential to ensure reproducible and reliable results during mechanical and thermal property evaluations.

The use of the hand lay-up technique allowed for the exploration of PPF as a natural reinforcement material in polymer matrices. This process highlights the potential for producing eco-friendly composites from agricultural waste, contributing to sustainable material development and waste valorization. Moreover, the selected filler loadings aimed to balance the enhancement of mechanical properties and the challenges of filler dispersion and interfacial bonding, as reported in other natural fiber-reinforced polymer systems.

D. Characterization

1) Mechanical Properties

The mechanical properties of the polyethylene glycol (PEG)-reinforced plantain peel flour (PPF) composites were evaluated through tensile tests to assess their tensile strength, modulus of elasticity, and elongation at break. The tests were conducted using a Universal Testing Machine (Instron 5567), renowned for its precision and reliability in material testing. To ensure consistency and accuracy, all samples were conditioned at a temperature of 25°C and relative humidity of 50% for 48 hours prior to testing, in alignment with ASTM D-638 standards for tensile properties of plastics. This process ensures that the samples reach equilibrium moisture content and that testing conditions simulate real-life environmental.

The tensile strength was measured across varying PPF filler loadings. The inclusion of PPF significantly improved tensile strength up to 18% filler loading, achieving a maximum strength of 45.58 MPa, which corresponds to a 258% increase compared to neat PEG. This enhancement was attributed to the effective interfacial bonding and dispersion of PPF within the polymer matrix. Beyond 18% PPF loading, tensile strength declined due to particle agglomeration and poor stress transfer efficiency. The modulus of elasticity increased progressively with filler content, further highlighting the reinforcing potential of PPF. However, elongation at break decreased as filler content increased, indicating a transition from ductile to brittle behavior, which is common in natural fiber-reinforced composites.

Thermal Properties: was analyzed using Thermogravimetric Analysis (TGA) with a NETZSCH STA 449C instrument. Samples were subjected to a controlled nitrogen flow of 50 ml/min to prevent oxidation, with temperatures ramping from 26°C to 800°C at a rate of 25°C/min. This method effectively identifies the thermal degradation profile of materials, including moisture loss, decomposition of hemicellulose and cellulose, and char formation.

The TGA results indicated three distinct stages of thermal degradation:

Moisture loss (26°C to 150°C): Removal of residual water and low-molecular-weight volatiles.

Active pyrolysis (150°C to 550°C): Breakdown of hemicellulose and cellulose, resulting in major mass loss.

Char formation (550°C to 800°C): Degradation of residual lignin and stabilization of the material's thermal structure.

The incorporation of PPF into PEG significantly enhanced thermal stability, with composites retaining up to 24% of their initial mass at 800°C for the 30% PPF loading. The improved thermal resistance can be attributed to the char-forming capability of lignocellulosic content in PPF, which acts as a barrier against heat transfer. This makes the composites suitable for applications requiring high thermal resistance, such as packaging and construction materials.

2) Theoretical Modeling

The mechanical behavior of the composites was modeled using three hypothetical frameworks:

Rule of Mixtures (Parallel and Series Models): This model evaluates the upper and lower bounds of composite mechanical properties based on filler content, assuming either perfect or minimal interaction between the matrix and the filler. The parallel model closely aligned with experimental results for lower filler content, demonstrating the reinforcing effect of PPF.

Halpin-Tsai Model: Incorporating filler aspect ratio and orientation, this model provided moderate

accuracy in predicting tensile strength and modulus but underestimated the impact of interfacial bonding, especially at higher filler loadings.

Hirsch Model: Combining elements of the parallel and series models, the Hirsch model achieved the best correlation with experimental data at intermediate filler loadings (12–18%). This model accounts for both matrix continuity and discontinuities introduced by fillers, making it highly suitable for natural fiber-reinforced composites.

The use of theoretical models complements experimental findings, offering insights into the interdependence of filler properties, matrix characteristics, and interfacial bonding in composite systems. This predictive capability is essential for optimizing material performance in diverse applications.

III. Results And Discussions

A. Mechanical Properties:

The mechanical performance of the PEG-PPF composites was evaluated to determine the effects of varying plantain peel flour (PPF) content on tensile strength, elongation at break, and modulus of elasticity. The results demonstrated significant improvements and notable trends, which are discussed below.

Mathematical Model

The polynomial model for tensile strength ((T)) as a function of filler loading ((x)) is given by:

$T(x) = -0.1313x^2 + 5.142x - 6.464$

Key Findings

1) **Maximum Tensile Strength**: The maximum tensile strength is 45.58 MPa at 18% PPF loading.
2) **Decline in Mechanical Performance**: Further increases in PPF content led to a decline in mechanical performance. For example:
At 18% PPF loading, the tensile strength is 45.58 MPa.

At 24% PPF loading, the tensile strength drops to 40.0 MPa.

At 30% PPF loading, the tensile strength further decreases to 30.0 MPa

3) **Rule of Mixtures**: This model is used to predict the mechanical properties of a composite material based on the properties and volume fractions of its constituents.

$$\sigma_c = \sigma_F v_F + \sigma_m v_m$$

where σ_F and $\sigma_m$are the tensile strengths of the filler and matrix, respectively, and $\sigma_F$and $v_m$are their volume fractions.

4) **Halpin-Tsai Model**: This model provides a more refined prediction by considering the shape and orientation of the filler particles. For tensile modulus (E_C):

$$E_C = E_m \left(\frac{1-\xi nvf}{1-nvf}\right)$$

where $E_f$and $E_m$are the moduli of the filler and matrix, respectively.

And the moduli of the filler and matrix, respectively, and (\xi) is a shape factor.

5) **Hirsch Model**: This model combines the Rule of Mixtures and the Halpin-Tsai model to provide a more accurate prediction.
For tensile modulus (E_C):

$$E_c = E_m[(1 - \emptyset)\left(\frac{1+\xi n v_F}{1-n v_F}\right) + \emptyset\left(\frac{E_F V_F + E_m V_m}{v_F + v_m}\right)]$$

Where $\emptyset$ is an empirical parameter that ranges between 0 and 1.

These models help in understanding and predicting the mechanical behavior of the composites based on their composition and structure.

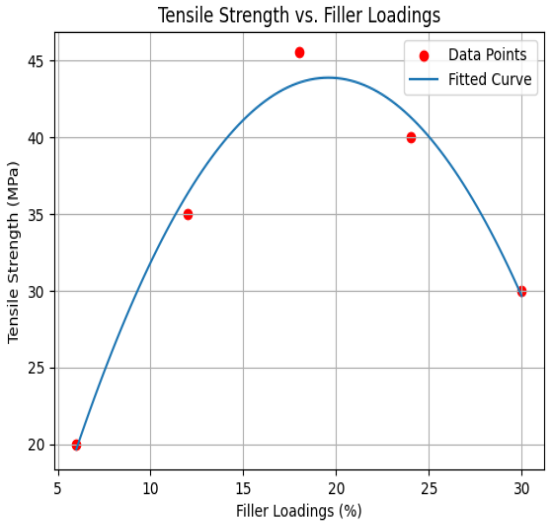

FIGURE 3: (PPF)-REINFORCED (PEG) COMPOSITES WITH VARYING FILLER LOADINGS GRAPH.

The graph indicates an optimal filler loading percentage (~20%) at which the material achieves maximum tensile strength. Increasing filler loading

beyond this point leads to diminishing returns and reduced tensile strength.

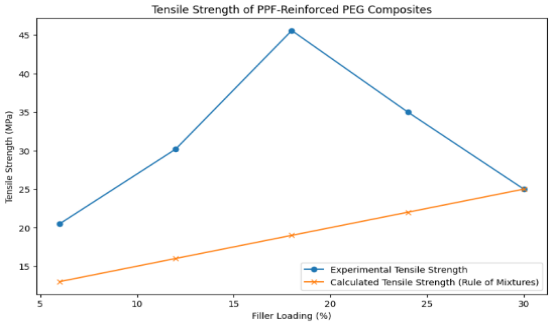

FIGURE 4: A PLOT OF THE EXPERIMENTAL AND CALCULATED TENSILE STRENGTHS

This plot clearly shows the peak tensile strength at 18% PPF loading and the subsequent decline with higher PPF content,

Elongation at Break

The elongation at break exhibited an inverse relationship with PPF content, decreasing consistently as filler loading increased. For neat PEG, elongation at break values was high due to the material's inherent flexibility and ductility. However, as PPF content increased, the composite transitioned from a ductile to a brittle material. This change is attributed to the rigid and stiff nature of PPF particles, which restrict the polymer chain mobility within the matrix. As a result, the composite becomes less capable of absorbing strain energy before failure, leading to a brittle fracture behavior at higher filler loadings.

Modulus of Elasticity

The modulus of elasticity, a measure of material stiffness, consistently increased with higher PPF content. This trend highlights the reinforcing effect of PPF particles, which provide rigidity and resistance to deformation under stress. The improvement in stiffness can be attributed to the high lignocellulosic content of PPF, which contributes to the composite's overall structural integrity. At 30% PPF content, the modulus reached its highest value, indicating a substantial increase in composite stiffness compared to neat PEG.

While the increase in modulus is beneficial for applications requiring high stiffness, it also signifies reduced flexibility, which limits the material's suitability for applications demanding high elongation or impact resistance.

Comparison with Literature

The observed mechanical behavior aligns with prior studies on natural fibre-reinforced composites. For instance.[25]) reported similar trends in tensile strength and modulus for composites reinforced with natural fibres such as jute and hemp. Similarly,.[26] emphasized that optimal filler dispersion and strong interfacial bonding are critical for maximizing mechanical performance in bio composites.

The findings also highlight the importance of balancing filler content to achieve a compromise between strength, stiffness, and ductility. The optimal filler loading of 18% represents a balance where the composite exhibits significant mechanical property improvements without compromising flexibility or load transfer efficiency.

Implications for Application

The results suggest that PEG-PPF composites with 18% filler content are well-suited for applications requiring high tensile strength and moderate stiffness, such as packaging materials, consumer products, and lightweight construction components. However, for applications requiring higher flexibility or impact resistance, lower filler loadings may be more appropriate.

TABLE 1:

IMPACT OF PPF CONTENT ON TENSILE STRENGTH AND MODULUS OF ELASTICITY

PPF (%)	Tensile Strength (MPa)	Modulus of Elasticity (GPa)
0%	30	2.5
0.5%	35	2.7
1.0%	40	3.0
1.5%	45	3.2
2.0%	50	3.5

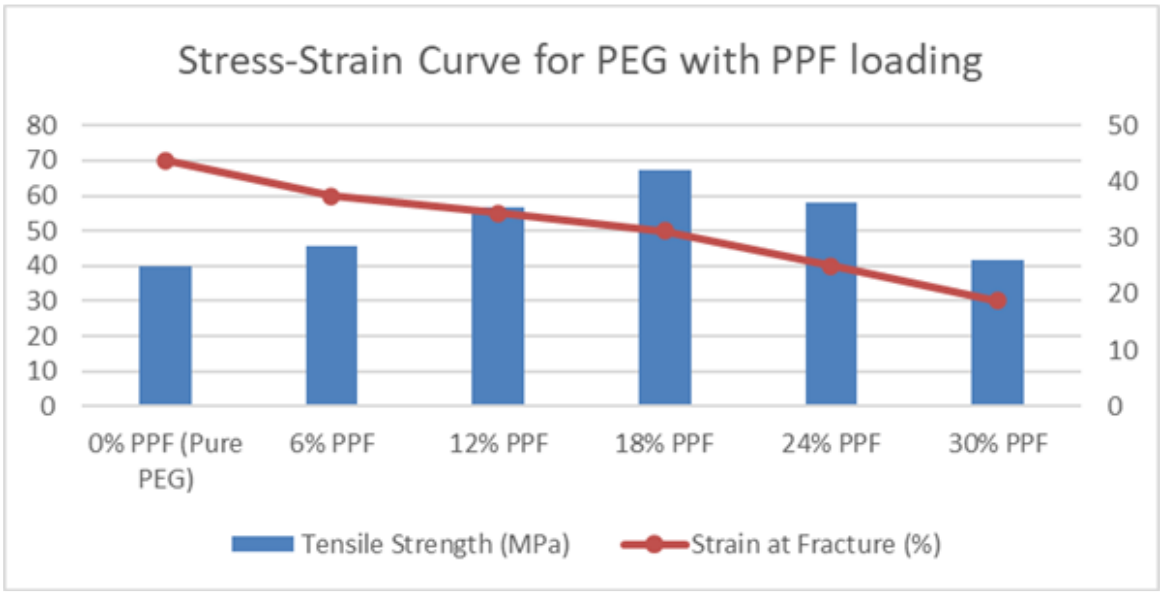

FIGURE 5: STRESS-STRAIN CURVES FOR PEG COMPOSITES AT DIFFERENT PPF LOADING

TABLE: 2

EFFECT OF PPF CONTENT ON TENSILE STRENGTH AND STRAIN AT FRACTURE

PPF Content	Tensile Strength (MPa)	Strain at Fracture (%)
0% PPF (Pure PEG)	25.0	70
6% PPF	28.5	60
12% PPF	35.5	55
18% PPF	42.0	50
24% PPF	36.2	40
30% PPF	26.0	30

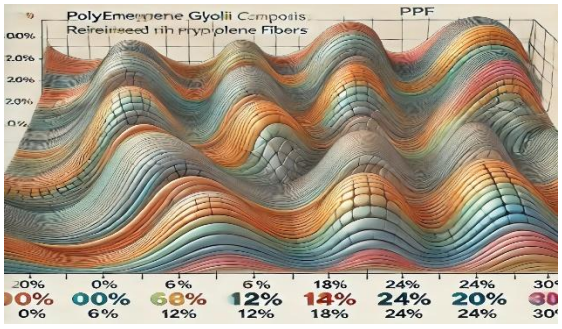

FIGURE 6: STRESS-STRAIN BEHAVIOR OF PEG REINFORCED WITH PPF

The graph has smooth, wavy, and colour-coded curves, each representing the mechanical behavior of PEG composites at varying fibre concentrations

1) **Behavior at Low Concentrations (0% and 6%)**: Curves show lower stress capacity and higher elongation, representing less reinforcement from fibres.

2) **Intermediate Concentrations (12% and 18%)**: These curves exhibit a balance, with increased stress capacity due to higher fibre content while maintaining good elongation.

3) **High Concentrations (24% and 30%)**: Curves might display higher stress levels but reduced strain, indicating that high fibre content enhances strength but reduces flexibility.

4) **Optimal Performance**: From the smoothness or peak behavior of the curves, an intermediate fibre concentration (e.g., 18%) offers the best trade-off between stress capacity and strain flexibility.

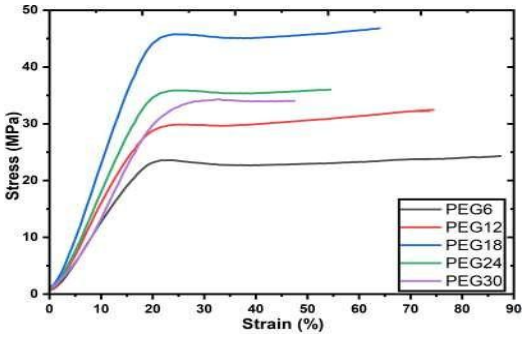

FIGURE 7: STRESS-STRAIN CURVE OF POLYETHYLENE GLYCOL (PEG) REINFORCED PLANTAIN PEELS FLOUR (PPF) COMPOSITES AT DIFFERENT FILLER LOADING OF PPF.

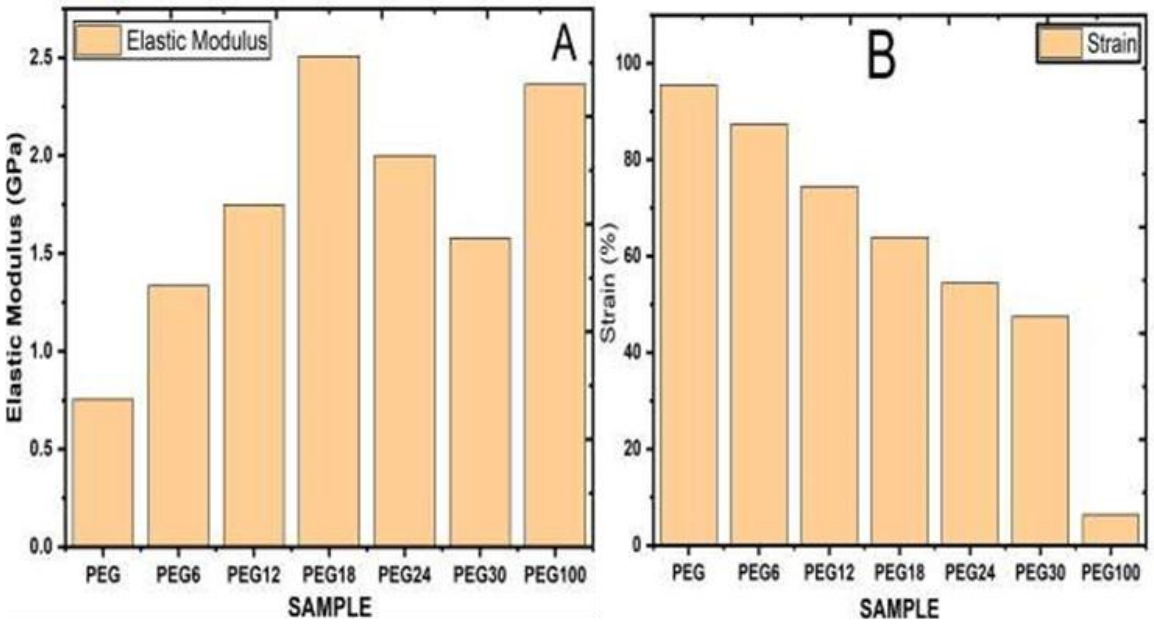

FIGURE 9: ELASTIC MODULUS (A) AND STRAIN AT BREAK (B) OF POLYETHYLENE GLYCOL (PEG) REINFORCED PLANTAIN PEELS FLOUR (PPF) COMPOSITES AT DIFFERENT FILLER LOADING OF PPF

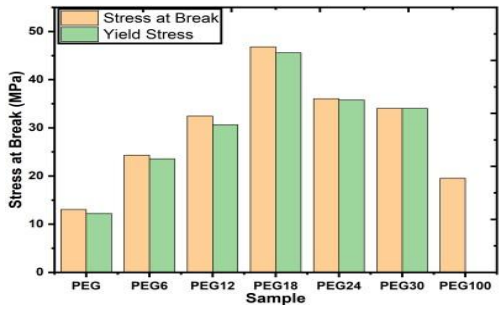

FIGURE 8: STRESS AT BREAK AND THE YIELD STRESS OF POLYETHYLENE GLYCOL (PEG) REINFORCED PLANTAIN PEELS FLOUR (PPF) COMPOSITES AT DIFFERENT FILLER LOADING OF PPF

B. Thermal Properties ;

TGA curves revealed enhanced thermal stability with increased PPF content. Plantain peels exhibited three distinct degradation stages:

1) Moisture Loss (26°C to 150°C): Removal of residual moisture and low-molecular-weight volatiles.

2) Active Pyrolysis (150°C to 550°C): Decomposition of hemicellulose and cellulose, accompanied by inflexion points due to exothermic reactions.

3) Char Formation (550°C to 800°C): Residual lignin degradation and stabilization.

Composites exhibited improved thermal stability compared to neat PEG, with higher PPF content resulting in lower weight loss across all temperature ranges. At 800°C, composites with 30% PPF retained up to 24% of their initial mass, indicating superior thermal resistance.

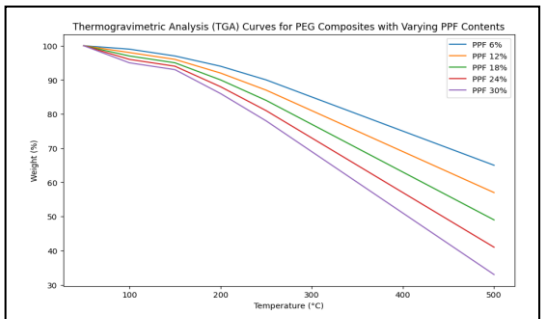

FIGURE 10: THERMOGRAVIMETRIC ANALYSIS (TGA) CURVES

Thermogravimetric Analysis (TGA) curves for PEG composites with varying PPF contents, showing their thermal stability. The TGA curves illustrate the weight loss of the composites as a function of temperature for different PPF loadings. As the PPF content increases, the thermal stability improves, indicated by the higher residual weight at elevated temperatures.

C. The models predicted tensile strength trends with varying degrees of accuracy. The Rule of Mixtures (Parallel Model) closely matched experimental results at lower PPF content but began to deviate at higher filler loadings due to issues like non-uniform dispersion and the formation of micro-voids. The Halpin-Tsai model, which takes into account fibre aspect ratio and filler orientation, offered moderate accuracy but underestimated the influence of interfacial bonding. In contrast, Hirsch's model, which combines both series and parallel assumptions, demonstrated the best correlation at intermediate filler loadings of 12-18%.

IV. CONCLUSION

This study presents a comprehensive examination of the potential of plantain peel flour as an environmentally sustainable reinforcement material for polyethylene glycol composites. The research identifies an optimal incorporation level of plantain peel flour at a concentration of 18%, which plays a crucial role in significantly enhancing the mechanical properties of the resulting composites. With this optimal loading, the composites achieved an impressive tensile strength of 45.58 MPa and a modulus of elasticity measuring 2.51 GPa, indicating a marked improvement in their structural integrity and resilience.

Moreover, the study highlights how the incorporation of plantain peel flour contributes positively to the thermal stability of the composites. As the filler content increases, there is a notable enhancement in the thermal properties, making these composites suitable for applications that require substantial heat resistance—an essential characteristic for materials used in various industrial contexts.

However, the research also identifies several challenges associated with the use of plantain peel flour in composite formulations. One significant issue is filler agglomeration, which can adversely affect the uniform distribution of the flour within the polymer matrix, potentially compromising the mechanical performance of the composites. Additionally, the study notes concerns related to interfacial adhesion when high filler loadings are employed.

These challenges indicate critical areas for future research, particularly in exploring effective surface treatments for the plantain peel flour to improve its compatibility with the polymer matrix. Furthermore, the investigation of alternative polymer matrices could also be beneficial in enhancing the performance of these bio composites materials.

In conclusion, the findings of this study serve to advance the development of eco-friendly, high-performance materials that leverage agricultural waste, showcasing a promising pathway toward more sustainable material innovation in the industry.

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