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Sustainable PLA/EFB Biocomposites Advanced with Green Plasticisers Enhancing Mechanical and Structural Integrity

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Polylactic acid (PLA), a widely used biodegradable thermoplastic polymer, offers high stiffness but suffers from inherent limitations such as brittleness, low impact resistance, and poor thermal stability, restricting its deployment for use in structural and dynamic load applications. Empty fruit bunch (EFB) fibres are used as natural reinforcements due to their high cellulose content and stability. However, the hydrophilic nature of EFB and the stiffness of PLA frequently result in poor interfacial bonding and low tensile strength. To address these shortcomings, plasticisers, including xylitol (XY), sorbitol (SORB), and propylene glycol (PG), are incorporated in the PLA/EFB matrix, which can improve the resistance to impact, elongation, and tensile strength. This study aims to investigate the synergistic effects of XY, SORB, and PG plasticisers on the strength and structure of PLA/EFB composites, including tensile (ASTM D638), flexural (ASTM D790), and Shore D hardness (ASTM D2240) testing performance, alongside morphological analysis using scanning electron microscopy (SEM). Fifteen PLA/EFB composite formulations (PLA 55-70 wt%, EFB 30-40 wt%) with various plasticisers (XY, SORB, PG) at 5-15 wt% were tested to evaluate their effect on mechanical properties. Blend 12, PLA/EFB/XY/SORB (60:30:5:5 wt%) composite proved to be the most effective, with tensile strength of 25.852 MPa, flexural strength of 2.871 MPa, and Shore D hardness of 74.8. SEM morphology verified improved fibre dispersion and matrix adherence. These biocomposites exhibit potential for applications in pipeline coatings, agricultural films, car interiors, and eco-friendly packaging.

1. Introduction

Research on polylactic acid (PLA)-based composites for uses such as pipeline coatings, construction, and packaging has intensified due to the growing need for sustainable materials. Although PLA is a biodegradable thermoplastic made from renewable resources, it has poor thermal stability, low impact resistance, and brittleness despite having acceptable processability (Ilyas et al., 2022).

Natural fibre reinforcements, such as empty fruit bunch (EFB) fibres, have been incorporated into PLA to enhance its mechanical properties. EFB is an economical and biodegradable agricultural waste product from the palm oil industry. However, because EFB is hydrophilic and the PLA matrix is hydrophobic, PLA/EFB composites frequently show poor interfacial adhesion, which leads to microvoids and decreased mechanical strength (Yee et al., 2023).

Plasticisers are often used in PLA-based composites to enhance compatibility and flexibility. By reducing the glass transition temperature, PG improves chain mobility and toughness (Coudane et al., 2022). PG enhances surface morphology and increases ductility (De Luca et al., 2023). While SORB lowers internal stresses and increases matrix uniformity (Mastalygina and Aleksanyan, 2024), XY enhances fibre dispersion and surface smoothness (Hou et al., 2021).

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This study investigates the synergistic effects of XY, SORB, and PG in the PLA composites, focusing on the mechanical and morphological properties used in advanced biodegradable engineering applications by evaluating tensile strength, flexural, and hardness. By improving mechanical and morphological qualities, this research is aimed at contributing to accelerating the development of next-generation eco-friendly materials with superior performance. This study is novel in its comparative evaluation of natural (XY and SORB) and synthetic (PG) plasticisers in PLA/EFB matrix, providing new insights into the design of eco-efficient biocomposites for industrial applications demanding mechanical and structural integrity.

2. Materials and Methods

2.1 Materials

The materials used in this research are PLA, EFB, XY, SORB, and PG. The composition ratio of the PLA/EFB/XY/SORB/PG blends was carefully formulated to achieve the optimal formulation between mechanical strength, such as tensile strength, flexural modulus, and hardness. The biocomposites were synthesised via melt blending and compression moulding, and characterised through mechanical testing and surface morphology via Scanning Electron Microscopy (SEM). Melt blending and compression moulding were used for developing the biocomposites, which ensured homogeneous dispersion and matrix-fibre interaction. The finalised composite specimens were allowed to cool down for 20 min before being taken from the mould (Mahendran et al., 2025).

Polylactic acid (PLA) pellets (commercial grade, supplied by Poly Scientific Enterprise Sdn Bhd, Malim, Melaka, Malaysia) were the matrix polymer. Empty fruit bunch (EFB) fibres were obtained from a local palm oil plant in Melaka, Malaysia. EFB were treated with sodium hydroxide (NaOH) at a concentration of 0.333 g/L, prepared by dissolving 30 g of NaOH in 90 L of water and immersed in the solution for 3 h, to enhance fibre-matrix bonding in the composite (Smith & Brown, 2020), then oven-dried at 80°C for 24 h, and finally ground to a particle size of around 250 µm, which adheres to ISO 3310-1. Plasticisers (Sigma-Aldrich) used included Xylitol (XY), Sorbitol (SORB) and Propylene glycol (PG). All chemicals were used as received without further purification.

2.2 Composite Preparation

A total of 15 composite formulations were prepared, varying PLA between 55-70 wt% and EFB fibres between 30-40 wt% (Table 1). To assess their impact on composite performance, plasticisers (XY, SORB, and PG) were added separately or in specific combinations at concentrations of 5-15 wt%. Three mechanical strengths data tabulated for all fifteen blends, consisting of tensile (MPa), flexural (MPa), and hardness (Shore D). Blends 1 and 2 are unplasticized, whereas blends 3-15 contain varying percentages of XY, SORB, and PG. Three plasticisers were added to the composite formulations to evaluate the impact of plasticiser incorporation on the mechanical performance of PLA/EFB composites.

Table 1: Mechanical Properties of PLA/EFB Composites With and Without Plasticisers

Blends	PLA wt%	EFB wt%	XY wt%	SORB wt%	PG wt%	Tensile (MPa)	Flexural (MPa)	Hardness (Shore D)
1	60	40	-	-	-	14.2004	4.023	68.1
2	70	30	-	-	-	16.902	2.207	66.7
3	55	30	-	-	15	1.349	7.530	44.7
4	60	30	-	-	10	3.629	3.297	56.9
5	65	30	-	-	5	2.265	5.694	58.1
6	55	30	-	15	-	19.097	2.568	68.4
7	60	30	-	10	-	14.837	1.883	67.2
8	65	30	-	5	-	14.645	2.458	59.3
9	55	30	15	-	-	17.728	2.423	65.5
10	60	30	10	-	-	23.3	3.240	50.4
11	60	30	5	5	-	25.491	2.347	56.3
12	60	30	5	5	-	25.852	2.871	74.8
13	60	30	5	-	5	5.382	2.110	67.9
14	60	30	-	5	5	9.838	6.101	70.1
15	55	30	5	5	5	2.85	5.790	66.46

PLA pellets were dried for 6 h at 60°C to remove moisture and prevent hydrolytic degradation during melt processing. PLA (55-70 wt%), plasticiser (5-15 wt%), and EFB fibres (30-40 wt%) are mixed. The process of blending included: (1) PLA, plasticisers, and EFB fibres are mixed dry, (2) Melt blending using an internal mixer at and heating them at 155°C for 25 min, and (3) compression moulding at 30 kPa for 15 min. (4) Test specimens are compressed using a hydraulic hot press at 170°C under 5 MPa for 5 min. The 40 g total weight composite mixture is placed in the 140 mm x 60 mm mould.

2.3 Characterisation

Mechanical properties were assessed using an Instron 3365 and Shore D durometer for tensile (Instron Model 8872) (ASTM D638), flexural (Instron Model 5585) (ASTM D790), and hardness (Durometer Analogue Shore 'D') (ASTM D2240) testing, as well as SEM (JEOL JSM-6010PLUS/LV) to examine fracture surfaces and fibre dispersion.

3. Results and Discussion

3.1 Mechanical Performance for PLA/EFB composites with and without plasticisers

Plasticisers like Xylitol (XY), Sorbitol (SORB), and Propylene Glycol (PG) have a major effect on the mechanical properties of PLA/EFB composites. These plasticisers modify the internal structure of the polymer matrix, affecting stress distribution, chain mobility, and interfacial bonding. They are therefore essential in adjusting the tensile, flexural, and hardness characteristics of the composite for certain engineering uses.

3.1.1 Plasticised Blends

Tensile Strength: Blend 12, PLA/EFB/XY/SORB (60:30:5:5 wt%) demonstrated the maximum tensile strength (25.85 MPa), suggesting the synergistic effect of dual plasticisers. Blends 11 (25.49 MPa) and 10 (23.30 MPa), which also contain XY and/or SORB, produced comparable high results.

In contrast, Blend 3 (PG-based, 15 wt%) had a significantly lower tensile strength (1.35 MPa), most likely due to poor interfacial bonding and reduced matrix cohesion produced by excessive PG. Balaji Ayyanar et al. (2023) discovered a similar pattern, with ideal filler ratios increasing tensile strength (39.23 MPa) and Shore D hardness (79.8), emphasising the necessity of strong filler-matrix interactions and minimum plasticiser interference.

Flexural Strength: Blend 14 (6.10 MPa) had the maximum flexural strength, as it comprised both SORB and PG. This shows that particular combinations of plasticisers can improve the composite's bending strength. PG-only blend, Blend 3, PLA/EFB/PG (55:30:15 wt%) depicts the highest flexural stress at 7.530 MPa; it demonstrated superior flexural performance, although with lower tensile strength.

Inversely, Blend 7, PLA/EFB/SORB (60:30:10 wt%) had the lowest flexural stress value, at 1.883 MPa. At a higher SORB (10 %), overplasticization softens the matrix and weakens fibre-matrix bonding, whereas decreased interfacial adhesion is probably the cause of the mechanical performance decline.

According to Coudane et al. (2022), incompatibility between PLA and plasticisers frequently results in suboptimal mechanical performance, particularly when plasticisers exceed optimal thresholds. Similarly, Bikiaris et al. (2023) pointed out that whereas plasticisers increase flexibility, excessive usage of them might disrupt matrix cohesiveness and lower load transfer efficiency.

Blend 12 with PLA/EFB/XY/SORB had the highest average hardness, with ratios of (60:30:5:5 wt%) at 74.8 Shore D. The synergistic effect of XY and SORB at low concentrations (5 %), which may have improved the matrix's densification and polymer chain interaction without significantly weakening the PLA structure, is responsible for this higher hardness. The matrix was probably reinforced by the modest 39% EFB concentration, which further enhanced stiffness.

Hardness: Hardness values varied between plasticised blends, with Blend 12 again having the greatest value (74.8 Shore D), indicating that dual plasticization can increase surface resistance. However, PG-rich blends, such as Blend 3, had the lowest hardness (44.7 Shore D), implying a softer, more compliant surface.

The observed improvement in hardness at low XY and SORB concentrations in PLA/EFB composites is consistent with findings from recent biopolymer research. Hou et al. (2021) revealed that the regulated addition of xylitol-based plasticisers increased toughness and surface integrity in PLA/PBS blends, with excessive plasticiser leading to decreased mechanical resistance.

Conclusively, the outcomes substantiate that the low concentrations of XY and SORB improve matrix cohesion and fibre bonding in PLA/EFB composites, resulting in higher surface hardness. In contrast, overplasticization softens the matrix and weakens its resistance to indentation, reducing mechanical integrity.

3.1.2 Unplasticized Blends

Blends 1 and 2, unplasticized, demonstrated average mechanical performance.

Tensile Strength: Blend 2, PLA/EFB (70:30 wt%) obtained 16.90 MPa, a higher value than Blend 1 (14.20 MPa), most likely because of the increased PLA content, which strengthened matrix continuity and load transfer.

Flexural Strength: Blend 1 has a higher flexural strength (4.02 MPa) than Blend 2 (2.21 MPa), potentially due to the increased EFB content, which contributes to stiffness.

Hardness: Both blends had reasonably high hardness values (68.1 and 66.7 Shore D, respectively), indicating strong surface rigidity in the absence of plasticisers.

The structural features of both plasticised and unplasticized PLA/EFB composites are illustrated in Figure 1. Plasticised composites perform better with certain mechanical properties, especially when the optimal XY and SORB compositions are employed. The concentration of plasticiser has a major impact on how effectively plasticization performs; excessive amounts of PG can have a detrimental effect on tensile strength and hardness. However, unplasticized composites are more mechanically stable but have limited flexibility and adaptability by nature.

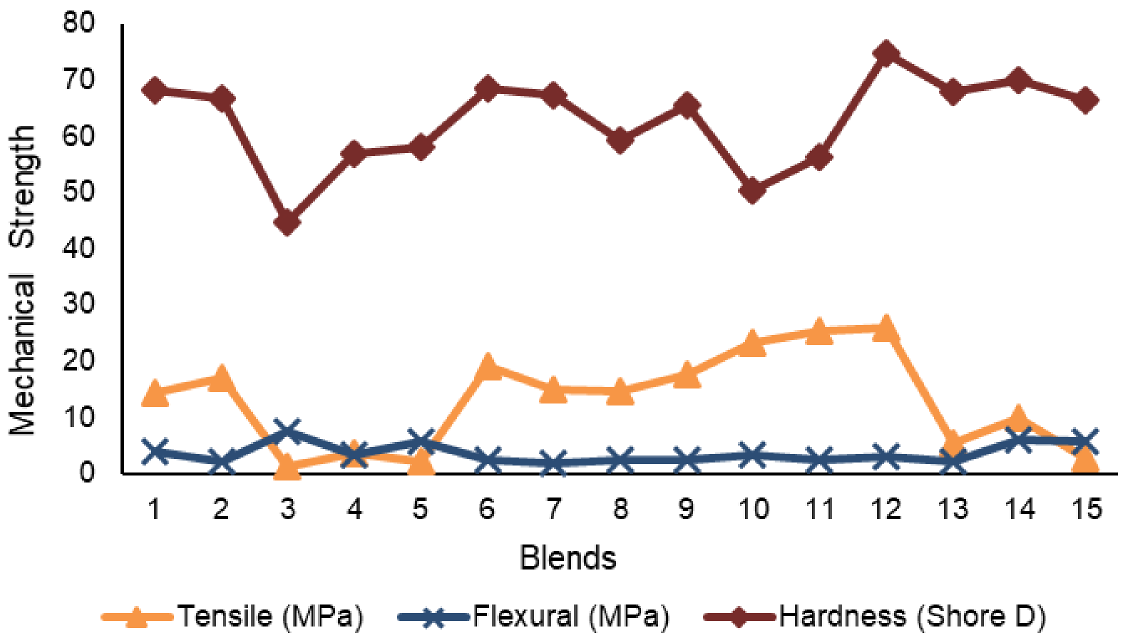

Figure 1: Comparative Mechanical Performance of PLA/EFB Composites with Varying Fibre Ratios and Plasticiser Content (Plasticised or Unplasticized)

3.2 Microstructural Analysis of Plasticised and Unplasticized Interfaces

The microstructural investigation of plasticised and unplasticized interfaces provides an essential understanding of the morphological evolution caused by plasticiser incorporation. The presence of plasticiser considerably affects phase dispersion, interfacial adhesion, and matrix continuity, affecting the overall structural integrity and performance of the PLA/EFB composite matrix.

Figure 2 shows the SEM micrographs of fractured surfaces at ×20 magnification for three PLA/EFB-based composite blends, illustrating the influence of composition and plasticiser addition on morphology. SEM examination reveals major morphological changes between the three PLA/EFB blends. Blend 1, PLA/EFB (60:40 wt%), has noticeable voids, fibre breakage, and poor fibre-matrix adhesion, indicating brittle fracture due to the high EFB amount and insufficient plasticiser. Blend 2, PLA/EFB (70:30 wt%), has fewer voids and slightly better matrix continuity, but still has weak interfacial bonding. Blend 12, PLA/EFB/XY/SORB (60:30:5:5 wt%) has a more uniform surface with fewer voids and increased flexibility, which is attributed to the plasticising effects of XY and SORB.

Although microcracks can be apparent in Blend 12, their presence does not significantly damage the composite's integrity. Instead, they exhibit the dynamic interaction of phases during plasticiser diffusion. The overall improvement in matrix continuity, decreased void formation, and increased ductility strongly indicate Blend 12 as the optimal formulation for balancing mechanical flexibility and structural performance in PLA/EFB composites.

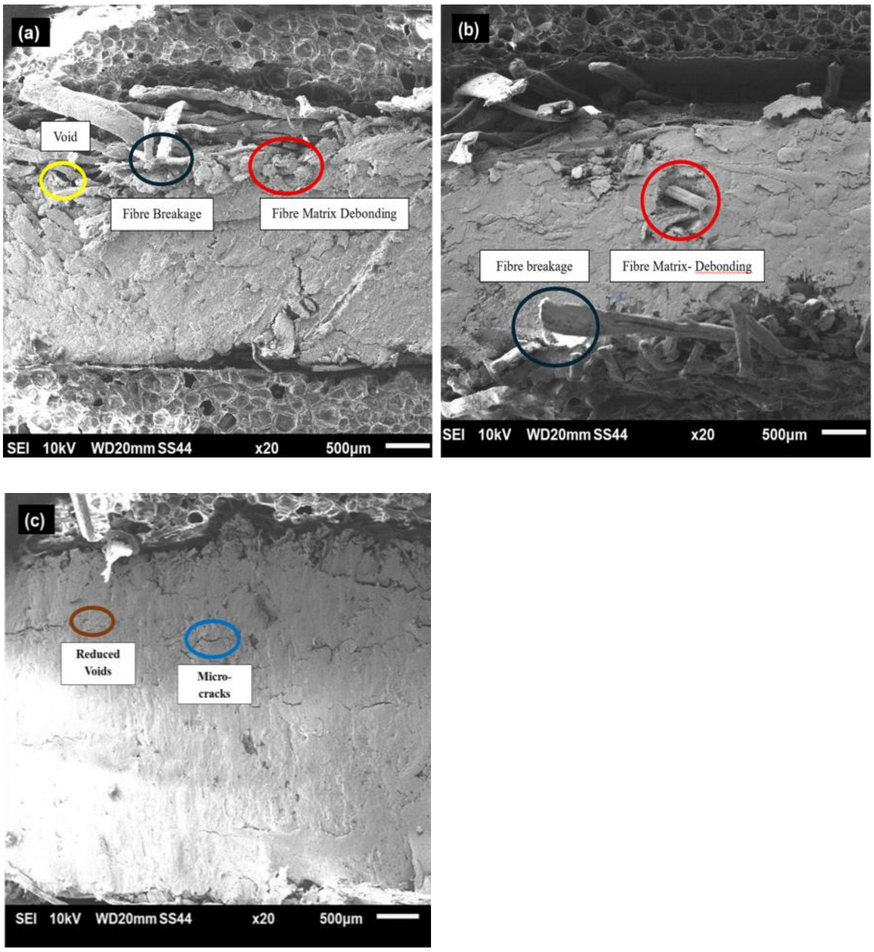

Figure 2: Illustration of (a) Blend 1, PLA/EFB (60:40 wt%), (b) Blend 2, PLA/EFB (70:30 wt%), and Blend 12, PLA/EFB/XY/SORB (60:30:5:5 wt%) of PLA/EFB Composites with Varying Fibre Ratios and Plasticiser Content (Plasticised or Unplasticized*)*

Table 2: SEM Morphology Analysis of PLA/EFB Composites With and Without Plasticisers

Crititcal Justification	Blend 1 PLA/EFB (60:40 wt%)	Blend 2 PLA/EFB (70:30 wt%)	Blend 3 PLA/EFB/XY/SORB (60:30:5:5 wt%)
SEM Morphology	Visible voids, fibre breakage (brittle fracture), and fibre-matrix debonding (poor adhesion) (Ilyas et al., 2022).	Similar to Blend 1, but with fewer voids, it still demonstrates fibre breakage and debonding (Faheem and Khan, 2025).	Surface appears more continuous, with micro-cracks and fewer voids than Blend 1 and 2 (Mastalygina and Aleksanyan, 2024).
Effect of Plasticisers on PLA/EFB composites	Insufficient plasticiser leads to poor interfacial bonding and increased brittleness (Omar et al., 2021).	Higher PLA ratio reduces fibre content, resulting in slightly better matrix continuity but still fragile (Pathariya et al., 2025).	The addition of XY/SORB plasticisers enhances matrix flexibility and lowers fibre pull-out while introducing microcracks due to plasticiser migration (Chen et al., 2024).
Conclusion	High EFB content causes increased stress concentration, leading to voids and brittle fractures (Ilyas et al., 2022).	Lower EFB improves dispersion, but adhesion remains weak without a plasticiser (Faheem and Khan, 2025).	Plasticisers improve ductility and minimise voids, while microcracks signal phase separation or over-plasticization (De Luca et al., 2023).

4. Conclusion and Research Implications

This research aimed to optimise PLA/EFB composites by integrating xylitol (XY), sorbitol (SORB) and propylene glycol (PG) plasticisers to improve mechanical and morphological performance for advanced biodegradable engineering applications. Plasticised blends exhibited significant improvements in tensile strength, flexural resistance, and Shore D hardness as compared to unplasticized composites, indicating the effectiveness of plasticiser incorporation in reducing brittleness and increasing toughness. SEM analysis supports these mechanical findings, demonstrating that plasticised blends, particularly Blend 12 (PLA/EFB/XY/SORB at 65:30:5:5 wt%), had a homogenous matrix with fewer voids and stronger fibre-matrix adhesion, resulting in greater stress transfer and ductility. Unplasticized blends, however, exhibited brittle fracture characteristics such as fibre pull-out, debonding, and large voids, which were associated with decreased toughness and strength. Although tiny microcracks were found in plasticised samples, they did not affect overall flexibility and toughness. Incorporating EFB fibres offered natural reinforcement, whereas XY, SORB and PG increased interfacial bonding and reduced brittleness. These findings highlight the potential of PLA/EFB/XY/SORB systems as environmentally friendly materials that promote green innovation and circular economy activities.

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