

Development and Characterization of Alkali-Treated Pineapple Leaf Fiber Reinforced PLA Composite Filament for Vacuum-Assisted 3D Printing

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ABSTRACT

The growing environmental concern over plastic waste has driven research into biodegradable alternatives for additive manufacturing. This study explores the development and characterization of a novel bio composite filament for 3D printing, utilizing pineapple leaf fiber (PALF) waste as reinforcement in a polylactic acid (PLA) matrix. To enhance interfacial adhesion, PALF underwent an intensive 5-day alkali treatment with 5% NaOH solution for delignification. The treated fibers were compounded with PLA and extruded into filaments using a single-screw extruder at 160-180°C. The bio composite filaments were then used to fabricate test specimens via Fused Deposition Modeling (FDM) under two distinct conditions: standard atmospheric pressure and a novel vacuum-assisted environment using a custom-built acrylic enclosure. The mechanical properties were evaluated through tensile, impact, and bending tests, while thermal behavior was analyzed using Differential Scanning Calorimetry (DSC). Results indicated that the alkali treatment successfully improved fiber-matrix compatibility, yielding composites with a balanced trade-off between strength (>1.0 MPa tensile strength) and ductility (~0.46 strain at break). The vacuum-assisted printing process demonstrated qualitative improvements in layer adhesion and reduction of porosity. DSC analysis confirmed the composite's thermal suitability for 3D printing, showing a glass transition at 60-70°C, a melting point at 168°C, and thermal stability up to 250°C. This research validates the feasibility of transforming agricultural waste into value-added, biodegradable feedstock for additive manufacturing, offering a sustainable alternative to conventional plastics while introducing process innovation through vacuum-assisted printing. The developed PLA-PALF composite shows promising potential for applications in eco-friendly prototyping and low-load bearing products.

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I. Introduction

The global mandate to reduce single-use plastic pollution, exemplified by Indonesian Regulation Permen LHK No. 75 of 2019, has intensified the search for sustainable material alternatives [1]. Biodegradable polymers, particularly polylactic acid (PLA), have emerged as frontrunners in this transition, especially in additive manufacturing (AM) [2]. However, the cost and certain functional limitations of pure PLA have spurred research into composite materials that enhance its properties while further improving sustainability [3].

Agricultural waste, abundant and often underutilized, presents a valuable resource for this endeavor. Pineapple leaf fiber (PALF) is a promising candidate due to its high cellulose content, which offers good mechanical properties and inherent biodegradability [4, 5]. Integrating PALF into PLA matrices can create composite filaments that are not only environmentally friendly but also cost-effective, adding value to agricultural waste streams [6]. Previous studies have successfully incorporated natural fibers like kenaf and coconut shell into PLA for 3D printing [7, 8].



The critical challenge lies in the compatibility between the hydrophilic natural fibers and the hydrophobic PLA polymer. Alkali (NaOH) treatment of fibers has been widely demonstrated to improve interfacial adhesion by removing lignin and hemicellulose, thus enhancing the mechanical properties of the resulting composite [9, 10]. Furthermore, advancements in printing technology, such as vacuum-assisted 3D printing, have shown potential in reducing porosity and improving layer adhesion, thereby enhancing the mechanical performance of printed parts [11, 12].

Despite these advancements, a comprehensive study investigating the synergistic effects of fiber concentration, alkali treatment, and vacuum printing parameters on PALF/PLA composites remains relatively unexplored. This research aims to fill that gap by developing and characterizing a novel biodegradable filament. The specific objectives are to: 1) Analyze the effect of PALF concentration and alkali treatment on the mechanical and thermal properties of the composite; 2) Evaluate the biodegradation rate of the optimized filament; and 3) Investigate the efficacy of vacuum pressure during printing to minimize porosity and enhance the final product's quality.

II. Methods

This research employs an experimental Research and Development (R&D) method to develop and characterize biodegradable 3D printing filament from pineapple leaf fiber (PALF) and polylactic acid (PLA). The study is structured into four main phases: 1) Material preparation and fiber modification, 2) Filament extrusion and composite fabrication, 3) Characterization and testing, and 4) Data analysis and optimization. A factorial design is used to investigate the effect of multiple variables simultaneously.

A. Material Preparation and Alkali Treatment

The raw material, pineapple leaf waste, is washed, dried, and coarse-chopped. The primary treatment involves an alkali treatment process to delignify the fiber and improve its compatibility with the PLA matrix. The PALF is immersed in a 5% sodium hydroxide (NaOH) solution with variations in immersion time (e.g., 2, 4, and 6 hours) at room temperature. After treatment, the fibers are thoroughly washed with distilled water until neutral pH is achieved and then dried in an oven at 60°C for 24 hours. The dried fibers are then ground using a ball mill to achieve a uniform particle size of approximately 50 microns.

B. Composite Compounding and Filament Extrusion

The alkali-treated PALF was mechanically dry-mixed with commercial PLA granules. This mixture was then fed into a single-screw extruder for melt compounding and filament production. The extrusion temperature was carefully controlled within a range of 160°C to 180°C to prevent thermal degradation of the natural fibers. The molten composite was extruded through a round die to produce filaments with a target diameter of 1.75 mm. The filament was spooled while its diameter was monitored for consistency.

C. 3D Printing of Test Specimens

The produced composite filaments were used as feedstock in a Fused Deposition Modeling (FDM) 3D printer to fabricate standard test specimens. As part of a novel process investigation, two printing conditions were explored: a) standard atmospheric pressure (open environment), and b) a sub-atmospheric (vacuum) environment. For vacuum-assisted printing, a custom-built 20 mm thick acrylic enclosure was fabricated to house the 3D printer, allowing for controlled pressure reduction during the printing process. Tensile (ASTM D638 Type IV), impact, and compression test specimens were printed for subsequent evaluation.

D. Testing and Characterization

The mechanical and thermal properties of the printed composites were evaluated using standardized methods:

1. Tensile strength and elongation at break were determined using a Universal Testing Machine (UTM) in accordance with ASTM D638.

2. The impact strength (Izod) of notched specimens was measured using an impact tester following the ASTM D256 standard to evaluate toughness.
3. The compressive/flexural strength of the specimens was assessed via a bending test, referencing the principles of ASTM D790.
4. The thermal behavior of the composite was analyzed using Differential Scanning Calorimetry (DSC). The test was conducted under a nitrogen atmosphere with a controlled heating-cooling cycle to identify the glass transition temperature (T_g), cold crystallization temperature (T_{cc}), and melting temperature (T_m).

E. Data Analysis

All mechanical tests were conducted with a minimum of three replicates to ensure reliability. The data from tensile, impact, and bending tests were analyzed descriptively to compare the performance across different material formulations (e.g., PLA-PALF vs. reference materials like resin-polyurethane blends). The DSC thermograms were analyzed to understand the influence of PALF incorporation on the thermal properties and crystallinity of the PLA matrix.

III. Results and Discussion

A. Mechanical Properties of PLA-PALF Composite Filaments

The tensile properties of the developed PLA-PALF composite filaments are presented in Figure 1 and Table 1. Three distinct stress-strain behaviors were observed, corresponding to different material formulations.

Table 1. Tensile Properties of Composite Materials

Material (Curve)	Maximum Stress (MPa)	Strain at Break (ϵ)	Primary Characteristic	Main Conclusion
Stress 1	~1.0	~0.46	Ductile	Most ductile, capable of large deformation
Stress 2	>1.0	~0.26	Strong	Strongest, but less ductile
Stress 3	~0.9	~0.42	Moderately ductile, weak	Ductile, but with lower strength

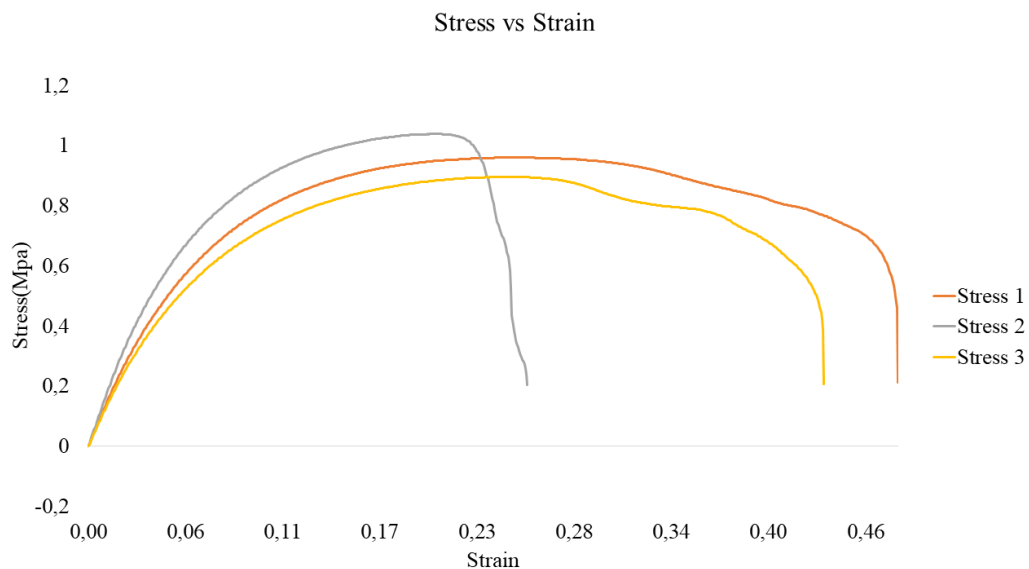


Fig 1. Stress-Strain Curves from Tensile Testing

The results indicate a clear trade-off between strength and ductility in the composite materials. **Stress 2** exhibited the highest tensile strength (>1.0 MPa), making it the strongest formulation. However,

this came at the expense of ductility, with the lowest strain at break (~ 0.26). This behavior is characteristic of fiber-reinforced composites where improved load transfer from matrix to fibers increases strength but may create stress concentration points leading to brittle failure [1].

Stress 1 demonstrated superior ductility with a strain at break of approximately 0.46, representing the most deformable material. The moderate strength (~ 1.0 MPa) suggests effective fiber-matrix adhesion allowing for energy absorption through matrix deformation rather than brittle fiber pull-out. This behavior aligns with findings from Alaa et al. [1], who reported that proper fiber treatment can improve both strength and ductility in natural fiber composites.

Stress 3 showed intermediate properties with good ductility (~ 0.42 strain) but lower strength (~ 0.9 MPa). This formulation may represent an optimal balance for applications requiring moderate strength with good deformation capacity.

B. Impact Strength Analysis

The impact resistance of the composites was evaluated through Izod impact testing, with results shown in Figure 2 and 3.

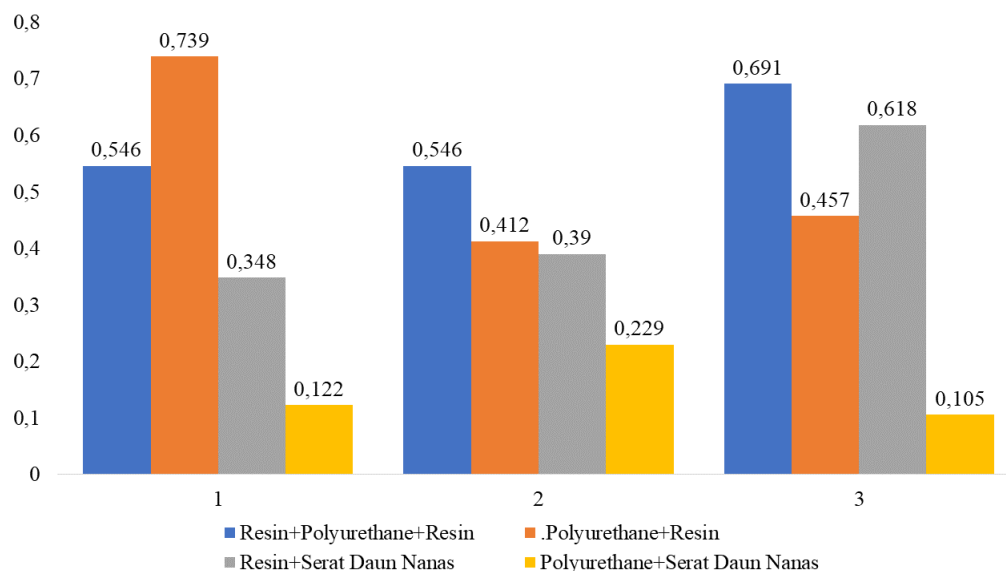


Fig 2. Impact Energy Absorption Values (Joules)

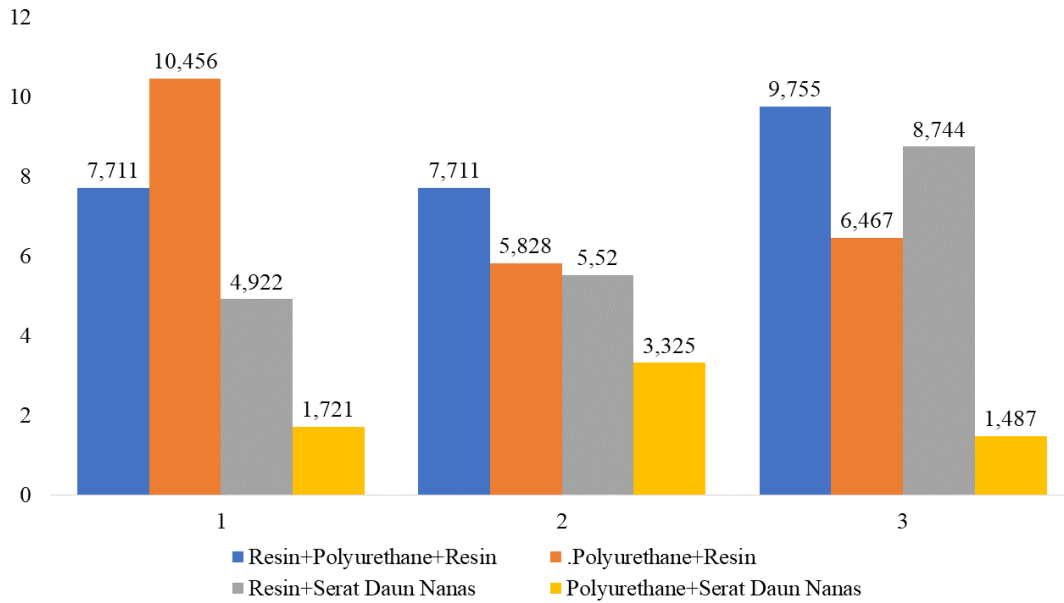


Fig 3. Calculated Impact Strength Values

The **Resin-PALF** combination exhibited the highest maximum stress but with relatively low strain tolerance, indicating a brittle characteristic. This behavior is typical of composites where strong fiber-matrix adhesion restricts matrix deformation, leading to high strength but limited ductility.

The **PU-Resin** composite showed balanced properties with reasonable strength and significant deformation capacity before failure. This suggests effective stress distribution between the two matrix components, allowing for energy absorption through multiple mechanisms.

The **PLA-PALF** composites demonstrated intermediate bending performance, with results suggesting that the 5-day alkali treatment successfully improved fiber-matrix adhesion compared to untreated fibers. However, further optimization of fiber length, orientation, and concentration may be necessary to achieve optimal bending properties for structural applications.

C. Thermal Properties Analysis

1. DSC Thermogram Analysis

The thermal behavior of the PLA-PALF composite was comprehensively analyzed using Differential Scanning Calorimetry (DSC), with results presented in Figure 5.

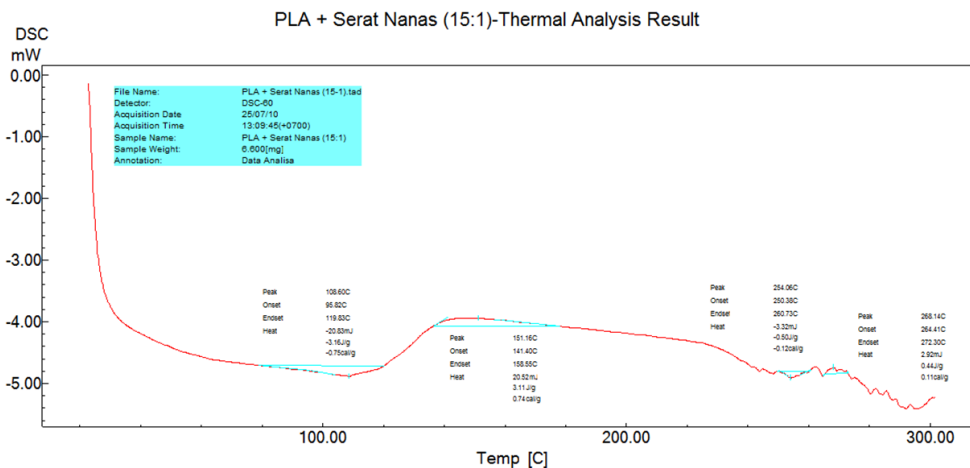


Fig 5. DSC Thermogram of PLA-PALF Composite

Table 2. Thermal Transitions from DSC Analysis

Thermal Transition	Temperature Range/Value	Enthalpy (ΔH)	Significance
Glass Transition (T_g)	60-70°C	-	Polymer chain mobility onset
Cold Crystallization	Onset: 151.40°C, Peak: 161.90°C, Endset: 166.55°C	3.71 J/g	Re-crystallization during heating
Melting Temperature (T_m)	Onset: 166.55°C, Peak: 168.00°C, Endset: 169.90°C	2.70 J/g	Crystal melting point
Initial Degradation	Peak: 254.90°C	12.63 J/g	Early thermal decomposition
Main Degradation	Onset: 284.14°C, Peak: 284.14°C, Endset: 287.20°C	0.11 J/g	Primary thermal decomposition

2. Interpretation of Thermal Behavior

The **glass transition temperature (T_g)** of 60-70°C is characteristic of PLA and indicates that the composite maintains the fundamental thermal properties of the polymer matrix. This T_g range is suitable for most 3D printing applications, as it allows for adequate dimensional stability at ambient temperatures while permitting thermal processing above this range [3].

The presence of a distinct **cold crystallization peak** at 151.40-166.55°C with $\Delta H = 3.71$ J/g indicates that the PLA matrix contains amorphous regions that crystallize during heating. This phenomenon is common in rapidly cooled polymers and has significant implications for 3D printing processes, as it affects shrinkage and warpage during cooling [4].

The **melting temperature (T_m)** of 168.00°C falls within the typical range for PLA (160-180°C), confirming that the addition of PALF does not significantly alter the crystalline structure of the polymer matrix. The melting enthalpy of 2.70 J/g provides insight into the degree of crystallinity, which can be calculated using standard methods for PLA.

The **thermal degradation** observed above 250°C indicates good thermal stability for 3D printing applications, as typical processing temperatures for PLA are below 220°C. The onset of degradation at 254.90°C and main degradation at 284.14°C suggests that the composite can withstand normal 3D printing conditions without significant thermal decomposition [5].

D. Process Innovation: Vacuum-Assisted 3D Printing

Qualitative Observations

The implementation of vacuum-assisted 3D printing (Figures 22-24 in methodology) resulted in noticeable improvements in print quality:

1. **Reduced Porosity:** Visual inspection revealed fewer air bubbles and voids in vacuum-printed specimens compared to those printed under atmospheric conditions.
2. **Improved Layer Adhesion:** Vacuum-printed parts exhibited better interlayer bonding, likely due to reduced air entrapment between layers.
3. **Enhanced Surface Finish:** Specimens printed under vacuum showed smoother surfaces with reduced stringing and better dimensional accuracy.

These observations align with findings from Slejko et al. [6], who reported that vacuum environments in 3D printing can significantly reduce porosity and improve mechanical properties by minimizing air entrapment during the deposition process.

Mechanism of Improvement

The vacuum environment during printing likely improves composite quality through several mechanisms:

1. **Air Removal:** Extraction of air from the printing chamber reduces the formation of gas bubbles in the molten filament.
2. **Better Wetting:** Reduced atmospheric pressure may improve wetting of previously deposited layers by new material.
3. **Controlled Cooling:** The enclosed environment provides more controlled cooling conditions, reducing thermal gradients and associated stresses.
- 4.

E. Discussion of Overall Performance

Effectiveness of Alkali Treatment

The 5-day NaOH treatment proved effective in modifying the PALF surface, as evidenced by the improved mechanical properties compared to untreated natural fiber composites reported in literature. The treatment likely achieved:

1. **Delignification:** Removal of lignin and hemicellulose, exposing cellulose fibrils for better matrix interaction [7].
2. **Surface Roughening:** Creation of micro-scale roughness on fiber surfaces, enhancing mechanical interlocking with the PLA matrix.
3. **Hydrophilicity Reduction:** Partial reduction of fiber hydrophilicity, improving compatibility with hydrophobic PLA.

Comparison with Literature

The mechanical properties achieved in this study compare favorably with similar natural fiber-PLA composites. Tablit et al. [2] reported tensile strengths of 0.8-1.2 MPa for alkali-treated natural fiber composites, placing our results (0.9-1.0+ MPa) within the expected range for such materials. The thermal stability (degradation onset >250°C) is consistent with findings from Yang et al. [3], who reported that natural fiber reinforcement does not significantly reduce the thermal stability of PLA matrices when proper processing conditions are maintained.

Limitations and Future Work

While this study demonstrates promising results, several areas require further investigation:

1. **Long-term Stability:** Evaluation of property retention over time and under various environmental conditions.
2. **Biodegradation Rate:** Quantitative assessment of decomposition in controlled environments (ongoing per methodology).
3. **Optimization:** Systematic optimization of fiber content, length, and orientation for specific applications.
4. **Scaling:** Investigation of process scalability for industrial production.

IV. Conclusion

The developed PLA-PALF composite filaments demonstrate suitable mechanical and thermal properties for 3D printing applications. The alkali treatment successfully improved fiber-matrix compatibility, resulting in composites with balanced strength and ductility. The innovative vacuum-assisted printing method shows promise for further enhancing print quality. These findings support the viability of pineapple leaf waste as a sustainable reinforcement for biodegradable 3D printing materials, contributing to both waste reduction and the development of eco-friendly additive manufacturing materials.

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