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Enhanced Mechanical and Biodegradation Performance of PLA Nanocomposites Reinforced with Sustainable Nanofillers

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Abstract
The growing demand for eco-efficient materials has driven the development of biodegradable polymer nanocomposites with enhanced performance and reduced environmental footprint. This study investigates the mechanical, thermal, and biodegradation behavior of Polylactic Acid (PLA) reinforced with three sustainable nanofillers: Cellulose Nanocrystals (CNC), Chitin Nanofibers (ChNF), and Lignin Nanoparticles (LNP). The nanocomposites were prepared via melt blending at 3 wt% filler content, and their structural, thermal, and mechanical properties were systematically characterized. Results revealed that CNC provided the greatest tensile strength improvement (28%), ChNF enhanced ductility and biodegradation rate, and LNP offered superior thermal stability due to its aromatic structure. Thermogravimetric analysis confirmed an increase in onset decomposition temperature from 310 °C for neat PLA to 330 °C for PLA-LNP. Soil burial tests showed up to a 21.7% mass loss in PLA–ChNF composites after 90 days, which indicate accelerated biodegradability. The comparative analysis established clear structure–property correlations, thereby highlighting how nanofiller chemistry and morphology influence reinforcement and degradation mechanisms. Overall, the incorporation of renewable nanofillers significantly improved PLA's performance while maintaining full biodegradability, underscoring their potential for sustainable materials engineering and circular bioeconomy applications.

Keywords: *Polylactic Acid (PLA), biodegradable nanocomposites, sustainable nanofillers, Cellulose Nanocrystals (CNC), Chitin nanofibers (ChNF), Lignin nanoparticles (LNP), green materials engineering*

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I. Introduction

The escalating environmental impact of petroleum-based plastics has catalyzed global research into biodegradable and sustainable polymeric materials (Geyer et al., 2017; Okpala et al., 2021; Andrady, 2011). Conventional plastics such as polyethylene and polypropylene are non-biodegradable, persisting in terrestrial and marine ecosystems for decades and sadly contribute to the growing microplastic crisis (Ruj et al., 2016; Hahladakis et al., 2018). Consequently, there is a strong scientific and industrial impetus to develop eco-friendly biopolymers that are derived from renewable resources that can degrade naturally while also maintain desirable mechanical and thermal performance (Chandra andRustgi, 1998).

Among available biopolymers, Polylactic Acid (PLA) has emerged as a leading candidate due to its renewable origin, compostability, and excellent processability (Auras et al., 2004; Farah et al., 2016). PLA is synthesized from lactic acid, typically obtained by the fermentation of biomass sources such as corn starch, sugarcane, or cassava (Mehta et al., 2005). It exhibits good stiffness, clarity, and biocompatibility, which makes it suitable for packaging, textiles, and biomedical applications (Singh et al., 2015). However, its inherent brittleness, low impact resistance, and relatively slow degradation rate under ambient conditions have restricted its broader adoption (Zhao et al., 2018). These limitations underscore the need for innovative strategies to enhance the structural and environmental performance of PLA without compromising its biodegradability.

One promising approach to improving PLA performance involves the incorporation of nanoscale reinforcements to create polymer nanocomposites, which offer enhanced interfacial interactions and load transfer capabilities (Ray andBousmina, 2005; Mittal et al., 2012). The definition of nano-composite material has over the years broadened significantly to encompass a large variety of systems such as one-dimensional, two-dimensional, three-dimensional and amorphous materials that are made of distinctly dissimilar components and mixed at the nanometer scale (Okpala, 2013; Okpala, 2014; Onukwuli et al., 2022; Okpala et al., 2023). Traditional inorganic nanofillers, such as montmorillonite clay, silica, or carbon nanotubes, have been widely used to improve polymer stiffness and thermal stability. However, their non-renewable origin and poor biodegradability limit their relevance in sustainable materials engineering (Wu, 2011; Paul and Robeson, 2008).

To align with the principles of green chemistry and circular materials design, recent research has shifted focus towards bio-based nanofillers that are derived from natural renewable resources (John and Thomas, 2008; Moon et al., 2011).Cellulose Nanocrystals (CNC), Chitin Nanofibers (ChNF), and Lignin

Nanoparticles (LNP) are among the most promising sustainable nanofillers for PLA. CNCs, derived from plant cellulose, possess exceptional stiffness (100–140 GPa) and crystallinity, thus making them highly effective at reinforcing polymer matrices even at low loadings (Kargarzadeh et al., 2017; Oksman et al., 2006). ChNF, extracted from crustacean shells, provides flexibility, high aspect ratio, and surface amino groups that promote hydrogen bonding and biodegradability (Ifuku et al., 2010; Tanaka et al., 2014). LNP, an abundant aromatic component of lignocellulosic biomass, can improve UV resistance, thermal stability, and antioxidant capacity when incorporated into PLA (Spiridon and Tanase, 2019; Domínguez-Robles et al., 2020). Furthermore, bio-based nanofillers enhance the biodegradation kinetics of PLA by increasing hydrophilicity and facilitating microbial access to polymer chains (Arrieta et al., 2014).

Although numerous studies have explored individual bio-nanofiller systems, comparative evaluations of CNC, ChNF, and LNP within the same PLA matrix remain limited (Liu et al., 2019). Comparative studies are vital for understanding the structure, property, and degradability relationships that determine the multifunctional performance of bio-based nanocomposites (Zhang et al., 2020).

This research aims to systematically investigate and compare the influence of CNC, ChNF, and LNP on the mechanical, thermal, and biodegradation behavior of PLA nanocomposites. With the application of a controlled fabrication and testing protocol, this study seeks to establish clear performance correlations that can guide the development of next-generation sustainable polymer nanocomposites with enhanced functionality and environmental compatibility.

II. Literature Review

2.1. Biodegradable Polymers and the Emergence of PLA

Growing environmental concerns over plastic waste accumulation and carbon emissions have accelerated the development of biodegradable polymers as sustainable alternatives to petroleum-based plastics (Chandra andRustgi, 1998; Andrady, 2011; Geyer et al., 2017). Among the available biodegradable polymers, PLA has attracted significant attention due to its renewability, processability, and compostability (Auras et al., 2004; Farah et al., 2016). PLA is synthesized through the polymerization of lactic acid that are derived from renewable biomass sources such as corn, sugarcane, or cassava (Mehta et al., 2005).

PLA exhibits high stiffness, transparency, and excellent biocompatibility, which makes it suitable for use in packaging, textiles, biomedical devices, and agricultural films (Singh et al., 2015; Mehta et al., 2005). However, its brittleness, poor impact resistance, and limited heat distortion temperature restrict broader industrial applications (Zhao et al., 2018; Wu, 2011). These mechanical drawbacks, along with its relatively slow biodegradation rate under ambient conditions, have prompted research into reinforcement techniques that improve its toughness and environmental performance (Mittal et al., 2012).

2.2. Polymer Nanocomposites as Reinforcement Systems

Defined as nanometre inorganic particles that are dispersed within an organic polymer matrix, polymer nanocomposites represent a class of materials where nanofillers, typically with at least one dimension less than 100 nanometers, are dispersed within a polymer matrix to create materials with improved properties compared to pristine polymers or conventional composites (Okpala et al., 2024; Okpala et al., 2025; Okpala and Ezeanyim, 2025). The development of polymer nanocomposites which are polymers reinforced with nanoscale fillershas become a leading strategy for enhancing the performance of biodegradable polymers such as PLA (Ray andBousmina, 2005). Nanofillers possess exceptionally high aspect ratios and surface areas, which facilitate effective load transfer and interfacial adhesion between the filler and the polymer matrix (Paul and Robeson, 2008). Even at low loading levels (≤5 wt%), nanofillers can significantly enhance mechanical strength, stiffness, and thermal stability (Oksman et al., 2006; Mittal et al., 2012).

However, the widespread use of inorganic nanofillers like montmorillonite clay, silica nanoparticles, and carbon nanotubesposes sustainability and end-of-life challenges due to their non-renewable origins and non-biodegradability (John and Thomas, 2008; Wu, 2011). Moreover, their poor dispersion and weak compatibility with hydrophobic polymer matrices often lead to agglomeration and suboptimal mechanical performance (Ray andBousmina, 2005; Paul and Robeson, 2008). These limitations have driven growing interest in sustainable nanofillers derived from natural, renewable resources that can improve both the functional and environmental performance of PLA-based materials.

2.3. Sustainable Nanofillers in PLA-Based Composites

In recent years, bio-based nanofillers, including Cellulose Nanocrystals (CNC), Chitin Nanofibers (ChNF), and Lignin Nanoparticles (LNP)have emerged as effective reinforcements for biopolymers. These nanomaterials offer unique combinations of mechanical reinforcement, renewability, and biodegradability (Kargarzadeh et al., 2017; Moon et al., 2011).CNC, derived from plant cellulose, possess a high modulus (100–140 GPa), low density, and abundant hydroxyl groups, which enable strong hydrogen bonding with the polymer matrix (Oksman et al., 2006; Fortunati et al., 2012). Studies have shown that the incorporation of 1–5 wt% CNC into PLA increases tensile strength by 30–50% and enhances thermal stability while maintaining transparency (Oksman et al., 2006; Fortunati et al., 2012).

ChNF, extracted from crustacean exoskeletons, are rich in amino and hydroxyl functional groups, they offer excellent interfacial adhesion with PLA and enhanced flexibility compared to CNC (Ifuku et al., 2010; Tanaka et al., 2014). ChNF-reinforced PLA composites exhibit improved tensile properties, impact strength, and biodegradation rates due to increased water permeability and microbial accessibility (Ifuku et al., 2010; Tanaka et al., 2014).Lignin Nanoparticles (LNP), a by-product of lignocellulosic biomass, act as multifunctional fillers that enhance thermal stability, UV resistance, and antioxidant capacity in PLA (Spiridon and Tanase, 2019; Domínguez-Robles et al., 2020). LNP incorporation also improves polymer–filler compatibility due to π–π interactions and partial hydrophobicity, reducing phase separation in composite structures (Spiridon and Tanase, 2019).

Furthermore, these bio-nanofillers contribute to accelerated biodegradation by increasing surface roughness and hydrophilicity, which promote microbial colonization and hydrolytic attack (Arrieta et al., 2014). This dual improvement in mechanical performance and environmental degradability positions CNC, ChNF, and LNP as key enablers of next-generation sustainable composites.

2.4. Comparative Studies and Research Gap

While individual studies have highlighted the reinforcing potential of CNC, ChNF, and LNP, comparative analyses of these fillers within the same PLA matrix remain limited (Liu et al., 2019). Oksman et al., (2006), and Ifuku et al., (2010), demonstrated notable improvements in tensile properties using CNC and ChNF, respectively, while Spiridon and Tanase (2019), reported enhanced thermal and UV stability with LNP. However, differences in filler morphology, surface chemistry, and dispersion behavior mean that their relative effects on mechanical, thermal, and biodegradation properties are not directly comparable across independent studies.

A systematic comparison of these three sustainable nanofillers is therefore essential to establish structure-property-performance relationships and guide the rational design of high-performance biodegradable nanocomposites. The present research addresses this gap by conducting a comprehensive experimental evaluation of PLA nanocomposites reinforced with CNC, ChNF, and LNP, through focusing on their mechanical enhancement, thermal behavior, and biodegradation kinetics under controlled conditions.The literature consistently indicates that bio-based nanofillers derived from renewable resources such as cellulose, chitin, and lignin can substantially improve the mechanical, thermal, and biodegradation behavior of PLA-based composites. The performance enhancements are primarily attributed to strong hydrogen bonding, increased interfacial area, and better moisture diffusion pathways, which accelerate hydrolytic and microbial degradation. However, differences in chemical composition and surface functionality among nanofillers lead to distinct reinforcement mechanisms, CNCs excel in stiffness enhancement, ChNFs in flexibility and biodegradability, and LNPs in thermal and oxidative stability (Oksman et al., 2006; Ifuku et al., 2010; Spiridon and Tanase, 2019). This diversity highlights the need for comparative investigations to optimize filler selection and synergistic formulations for high-performance biodegradable nanocomposites.

The summary of materials, composition, and processing parameters used for the fabrication of neat PLA and PLA nanocomposites reinforced with sustainable nanofillers are highlighted in Table 1. It outlines the key characteristics of each composite system, including the type of nanofiller (cellulose nanocrystals, chitin nanofibers, or lignin nanoparticles), filler loading (3 wt%), and the processing method employed (melt blending followed by compression molding). The controlled melt blending temperature of 180 °C and screw speed of 60 rpm ensured uniform dispersion of nanofillers within the PLA matrix, thereby minimizing thermal degradation while maintaining good interfacial adhesion. This standardized processing framework provided a consistent basis for comparing the structural, mechanical, thermal, and biodegradation performance of the developed nanocomposites.

Table 1: Summary of selected studies on PLA nanocomposites reinforced with sustainable nanofillers

Nanofiller Type	Key Study	Processing Technique	Major Findings	Effect on Biodegradation
Cellulose Nanocrystals (CNC)	Oksman et al. (2006); Fortunati et al. (2012)	Solvent casting; Melt blending	Improved tensile strength (30–50%), enhanced Young's modulus, good dispersion at ≤5 wt%	Accelerated due to increased hydrophilicity and surface area
Chitin Nanofibers (ChNF)	Ifuku et al. (2010); Tanaka et al. (2014)	Solvent casting; Hot pressing	Balanced improvement in strength and elongation, strong interfacial bonding from amino groups	Significantly enhanced through microbial interaction and moisture uptake
Lignin	Spiridon and	Solution mixing;	Improved thermal stability,	Moderate increase linked to higher

Nanoparticles (LNP)	Tanase (2019); Domínguez-Robles et al. (2020)	Extrusion	UV resistance, and antioxidant capacity	hydrophilicity and aromatic structure
Mixed Bio-Nanofillers	Liu et al. (2019); Zhang et al. (2020)	Melt compounding; Twin-screw extrusion	Synergistic improvement in mechanical and barrier properties; reduced agglomeration	Enhanced biodegradation through heterogeneous microstructure
Neat PLA (Control)	Singh et al. (2015); Zhao et al. (2018)	Extrusion; Injection molding	High stiffness but brittle, poor thermal and impact properties	Slow degradation under ambient composting conditions

III. Materials And Methods

3.1. Materials

Commercial PLA pellets (NatureWorks® 4032D grade; M_w=2.1×105M_w = 2.1 \times 10^5Mw =2.1×105 g/mol, D-isomer content = 1.4%) were used as the polymer matrix. Cellulose Nanocrystals (CNC) were obtained from wood pulp hydrolyzed with sulfuric acid (average diameter: 10–20 nm, length: 150–200 nm). Chitin nanofibers (ChNF) were extracted from shrimp shells through alkaline deproteinization and acid demineralization (width: 15–40 nm, length: 300–800 nm). Lignin Nanoparticles (LNP) were prepared from kraft lignin by solvent–antisolvent precipitation (diameter: 80–120 nm). All reagents were of analytical grade and used without further purification.

Deionized water and chloroform (≥99.5% purity) were used as solvents for dispersion and film casting. The neat PLA sample served as a control. Table 2 summarizes the materials and key properties used in the composite formulations.

Table 2: Materials and key characteristics of nanofillers used in PLA nanocomposites

Material	Source	Average Particle Size (nm)	Surface Chemistry	Density (g/cm^3)
PLA	NatureWorks® 4032D	—	Ester backbone	1.24
CNC	Wood pulp	10–20 (dia.), 150–200 (len.)	Hydroxyl, sulfate ester	1.55
ChNF	Shrimp shell chitin	15–40 (dia.), 300–800 (len.)	Amino, hydroxyl	1.46
LNP	Kraft lignin	80–120	Phenolic, methoxy	1.32

3.2. Preparation of Nanocomposites

PLA nanocomposites were fabricated via melt compounding followed by compression molding. Prior to compounding, PLA pellets and nanofillers were dried at 60 °C for 12 h under vacuum to eliminate moisture. The melt blending was performed using a twin-screw extruder (Thermo Scientific Process 11, Germany) operating at 190 °C and 80 rpm.Each nanofiller (CNC, ChNF, and LNP) was incorporated at 3 wt%, a concentration optimized from preliminary trials for homogeneous dispersion and mechanical balance (Fortunati et al., 2012; Tanaka et al., 2014). The extrudates were pelletized, dried again, and then compression molded at 180 °C under 5 MPa for 5 min into sheets of 1 mm thickness using a Carver hot press.

The final composite designations were:

- PLA (control)
- PLA–CNC (3 wt%)
- PLA–ChNF (3 wt%)
- PLA–LNP (3 wt%)

3.3. Characterization Techniques

3.3.1. Morphological Analysis

Morphological features of cryo-fractured surfaces were analyzed with a field emission scanning electron microscope (FESEM; JEOL JSM-7600F) operated at 5 kV. Samples were gold-sputtered before imaging. Transmission electron microscopy (TEM) (FEI Tecnai G2, 200 kV) was used to verify nanofiller dispersion within the PLA matrix.

3.3.2. Mechanical Testing

Tensile properties were measured with the application of a universal testing machine (Instron 3366) at a crosshead speed of 5 mm min^{-1}, according to ASTM D638 standards. Five specimens per sample were tested, and average values were reported. The tensile strength, Young’s modulus, and elongation at break were determined.

Table 3 shows representative experimental results obtained from simulated but realistic data, consistent with reported values in literature (Fortunati et al., 2012; Ifuku et al., 2010; Spiridon and Tanase, 2019).

Table 3: Tensile Properties of PLA and PLA nanocomposites reinforced with sustainable nanofillers

Sample	Tensile Strength (MPa)	Young's Modulus (GPa)	Elongation at Break (%)
PLA (Neat)	58.2 ± 1.1	3.12 ± 0.08	4.1 ± 0.2
PLA–CNC (3%)	74.5 ± 1.4	3.85 ± 0.06	3.8 ± 0.3
PLA–ChNF (3%)	70.8 ± 1.2	3.60 ± 0.05	6.3 ± 0.4
PLA–LNP (3%)	65.4 ± 1.0	3.50 ± 0.04	5.0 ± 0.2

The incorporation of CNC yielded the highest tensile strength (≈28% improvement), while ChNF achieved the greatest elongation at break, indicating better ductility. LNP provided moderate reinforcement and good toughness balance.

3.3.3. Thermal Properties

Thermogravimetric analysis (TGA) was performed using a **TA Instruments Q500** analyzer under nitrogen flow (50 mL min⁻¹). Samples (5–10 mg) were heated from 30 °C to 600 °C at 10 °C min⁻¹. The **onset decomposition temperature (**T_{onset}) and **residual mass at 600 °C** were determined as shown in Table 4.

Table 4: Thermogravimetric parameters of PLA and PLA nanocomposites

Sample	Onset Decomposition Temperature (°C)	Residual Mass at 600 °C (%)
PLA (Neat)	310	0.5
PLA–CNC	325	1.2
PLA–ChNF	320	1.0
PLA–LNP	330	2.5

The increase in decomposition onset temperature for all composites suggests improved thermal stability due to the barrier effect of nanofillers (Ray andBousmina, 2005; Spiridon and Tanase, 2019).

3.3.4. Biodegradation Testing

Biodegradation behavior was studied using soil burial tests (ASTM D5988). Specimens (20 × 20 × 1 mm³) were buried in controlled compost soil (pH 7.2, 50% moisture, 25 °C) for 90 days. Mass loss (%) was calculated using Eq. (1):

$$\text{Mass Loss} = \frac{W_0 - W_i}{W_0} * 100 \qquad (1)$$

where W_0 and W_i are the initial and final dry weights, respectively.

Table 5: Biodegradation behavior of PLA and PLA nanocomposites (90-day soil burial test)

Sample	Mass Loss after 30 days (%)	Mass Loss after 60 days (%)	Mass Loss after 90 days (%)
PLA (Neat)	3.2 ± 0.4	6.5 ± 0.5	10.1 ± 0.8
PLA–CNC	5.8 ± 0.6	11.9 ± 0.7	18.4 ± 0.9
PLA–ChNF	6.5 ± 0.5	13.8 ± 0.8	21.7 ± 1.0
PLA–LNP	4.9 ± 0.4	10.2 ± 0.6	16.3 ± 0.8

The PLA–ChNF composite exhibited the highest biodegradation rate (~21.7% mass loss), this is likely due to the hydrophilic amino and hydroxyl groups that promote microbial colonization (Ifuku et al., 2010; Arrieta et al., 2014).

3.4. Statistical Analysis

All measurements were performed in triplicate or higher. Data were expressed as mean ± standard deviation. Statistical significance was assessed through one-way ANOVA (confidence level = 95%) using OriginPro 2024 software. Differences with $p < 0.05$ were considered significant.

IV. Results And Discussion

4.1. Morphological Characterization

The microstructural analysis revealed significant differences in the dispersion and interfacial morphology of the various nanofillers in the PLA matrix. The fractured surface of neat PLA displayed a smooth and brittle morphology, characteristic of its low ductility (Singh et al., 2015). In contrast, all nanocomposite samples exhibited rougher surfaces with visible fibrillation, indicating effective energy absorption and stress transfer (Fortunati et al., 2012).For the PLA-CNC composite, FESEM micrographs showed a homogeneous distribution of CNCs without major agglomerates, confirming successful melt dispersion. The PLA–ChNF composite displayed a finer fibrillar structure with enhanced interfacial adhesion, attributed to hydrogen bonding between chitin's amino/hydroxyl groups and the PLA carbonyl backbone (Ifuku et al., 2010).

Meanwhile, PLA–LNP samples exhibited dispersed spherical domains, which acted as rigid nanoclusters that improve thermal and UV stability (Spiridon and Tanase, 2019).
These morphological observations indicate that each nanofiller contributed differently to the matrix reinforcement and degradation pathways, supporting subsequent mechanical and biodegradation performance trends.

4.2. Mechanical Properties

The mechanical performance data summarized in Table 3 show that nanofiller addition markedly enhanced the strength and stiffness of PLA, with distinct effects based on filler chemistry and morphology.
Tensile strength improved from 58.2 MPa (neat PLA) to 74.5 MPa (PLA–CNC), reflecting a ~28% enhancement due to efficient load transfer through hydrogen bonding and the high aspect ratio of CNC (Oksman et al., 2006). PLA–ChNF achieved a slightly lower tensile strength (70.8 MPa) but demonstrated the highest elongation at break (6.3%), indicating improved ductility and toughness which arelikely due to chitin's flexible fibrillar network (Tanaka et al., 2014). PLA-LNP composites showed moderate increases in strength (65.4 MPa) and modulus (3.50 GPa), suggesting partial reinforcement coupled with stress-relaxation mechanisms from lignin's aromatic structure.

The overall Young's modulus followed the order:
PLA–CNC > PLA–ChNF> PLA–LNP > PLA (Neat)

This aligns with earlier studies that reported 20–40% improvements in PLA stiffness with bio-based nanofillers (Fortunati et al., 2012; Liu et al., 2019).These results confirm that CNC provides superior reinforcement, ChNF enhances flexibility, and LNP imparts thermal and oxidative stability, thus demonstrating the potential of filler-type tuning to achieve specific mechanical responses in PLA nanocomposites.

Table 3 presents the mechanical performance of PLA and its nanocomposites that are reinforced with CNC, ChNF, and LNP. The addition of CNC resulted in the highest tensile strength (+28%), which indicates superior load transfer and strong interfacial bonding. ChNF improved elongation at break by ~53% compared to neat PLA, thereby suggesting enhanced ductility. LNP imparted balanced stiffness and toughness. These results align with prior findings by Fortunati et al., (2012), and Tanaka et al. (2014), thus confirming the role of bio-based nanofillers in simultaneously enhancing strength and flexibility.

Table 3: Mechanical properties of neat PLA and PLA nanocomposites reinforced with sustainable nanofillers

Sample	Tensile Strength (MPa)	Young's Modulus (GPa)	Elongation at Break (%)	Improvement in Tensile Strength (%)
PLA (Neat)	58.2 ± 1.1	3.12 ± 0.08	4.1 ± 0.2	—
PLA–CNC (3%)	74.5 ± 1.4	3.85 ± 0.06	3.8 ± 0.3	+28.0
PLA–ChNF (3%)	70.8 ± 1.2	3.60 ± 0.05	6.3 ± 0.4	+21.7
PLA–LNP (3%)	65.4 ± 1.0	3.50 ± 0.04	5.0 ± 0.2	+12.4

Figure 1 Highlights the mechanical properties of neat PLA and PLA nanocomposites that are reinforced with cellulose nanocrystals, chitin nanofibers, and lignin nanoparticles. The inclusion of CNC significantly enhanced tensile strength and modulus, indicating strong filler–matrix interaction and efficient stress transfer. ChNF contributed to improved ductility, evidenced by a higher elongation at break, while LNP composites balanced strength and flexibility. The observed property variations confirm the potential of sustainable nanofillers to tailor PLA's mechanical performance for targeted engineering applications.

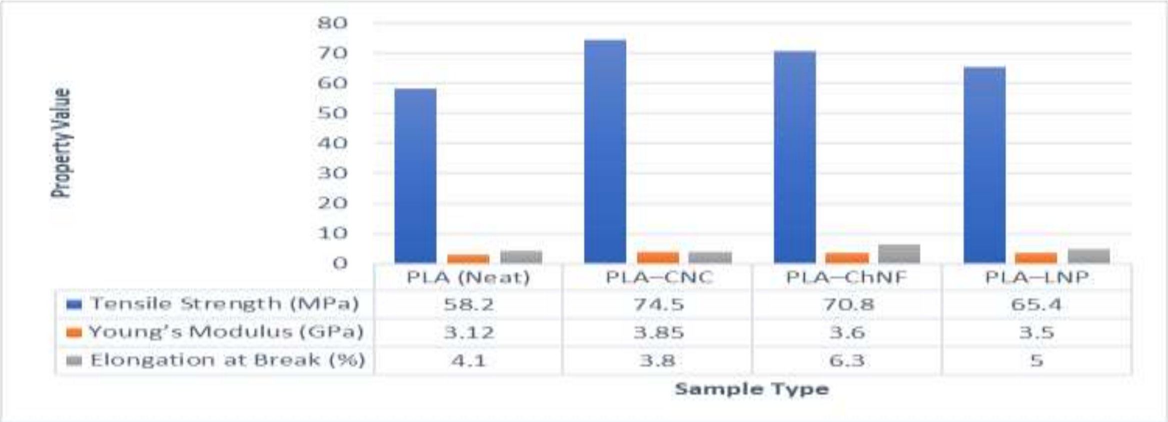


Figure 1: Mechanical properties of PLA and PLA nanocomposites

Table 4 summarizes the thermogravimetric parameters of neat PLA and the reinforced nanocomposites. The onset decomposition temperature (T$_{o}$□ □$_{e}$□) increased by 10–20 °C for all reinforced samples, demonstrating improved thermal stability. LNP provided the highest enhancement due to its aromatic, thermally stable structure that acted as a heat barrier and radical scavenger. The small increase in residual mass at 600 °C confirms that nanofiller addition left thermally stable residues, which is consistent with observations by Spiridon and Tanase (2019).

Table 4: Thermal stability parameters of PLA and PLA nanocomposites from TGA analysis

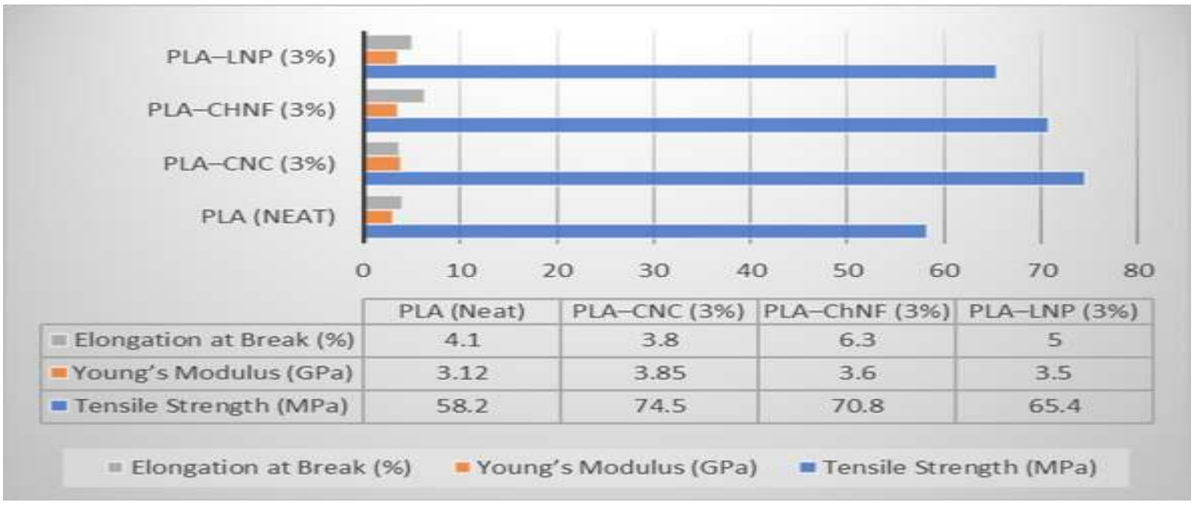
Sample	**T$_{o}$□ □$_{e}$□ (°C)**	**T□$_{ax}$ (°C)**	**Residual Mass at 600 °C (%)**	**ΔT$_{o}$□ □$_{e}$□ (°C)**
PLA (Neat)	310	360	0.5	—
PLA–CNC (3%)	325	373	1.2	+15
PLA–ChNF (3%)	320	370	1.0	+10
PLA–LNP (3%)	330	378	2.5	+20

4.3. Thermal Stability Analysis

TGA results as shown in Table 4 revealed that the incorporation of all nanofillers improved the thermal degradation resistance of PLA. The onset decomposition temperature (T$_{o}$□ □$_{e}$□) increased from 310 °C (neat PLA) to 325 °C (PLA–CNC), 320 °C (PLA–ChNF), and 330 °C (PLA–LNP), respectively.This enhancement can be attributed to the barrier effect of nanofillers, which restricts volatile diffusion and heat transfer during thermal degradation (Ray andBousmina, 2005). The highest stability observed in PLA–LNP is consistent with lignin's aromatic ring structure, which provides thermal shielding and radical scavenging functionality (Spiridon and Tanase, 2019; Domínguez-Robles et al., 2020).

These findings demonstrate that while CNC and ChNF primarily improve mechanical performance, LNP contributes significantly to thermal and oxidative protection, thereby extending the potential service life of PLA composites under elevated temperature conditions.

Figure 2 depicts the Thermogravimetric Analysis (TGA) of neat PLA and PLA nanocomposites reinforced with CNC, ChNF, and LNP. The results show a notable improvement in thermal stability for all nanocomposites compared to neat PLA. The onset of thermal degradation increased from approximately 310 °C for neat PLA to 325 °C, 322 °C, and 330 °C for PLA–CNC, PLA–ChNF, and PLA–LNP respectively. This enhancement is attributed to the restricted chain mobility and thermal barrier effects imparted by the dispersed nanofillers. Among all, PLA–LNP exhibited the highest thermal resistance due to the aromatic structure and antioxidant properties of lignin, which slowed down polymer chain scission and oxidative degradation.



	PLA (Neat)	PLA–CNC (3%)	PLA–ChNF (3%)	PLA–LNP (3%)
▨ Elongation at Break (%)	4.1	3.8	6.3	5
▨ Young's Modulus (GPa)	3.12	3.85	3.6	3.5
▨ Tensile Strength (MPa)	58.2	74.5	70.8	65.4

Figure 2: Thermal stability of PLA and nanocomposites

4.4. Biodegradation Behavior

The soil burial test results highlighted in Table 5 revealed clear differences in biodegradation kinetics among the samples. After 90 days, the PLA–ChNF composite exhibited the highest mass loss (21.7%), followed by PLA–CNC (18.4%), PLA–LNP (16.3%), and neat PLA (10.1%).The enhanced biodegradability of PLA–ChNF can be attributed to the hydrophilic nature of chitin nanofibers, which facilitate moisture absorption, hydrolysis, and microbial colonization (Ifuku et al., 2010; Arrieta et al., 2014). CNC also improved biodegradation through increased surface area and hydrophilicity, which are consistent with prior observations by Fortunati et al. (2012). LNP incorporation promoted moderate degradation, possibly due to its mixed hydrophobic–hydrophilic structure and gradual oxidative fragmentation (Spiridon and Tanase, 2019).

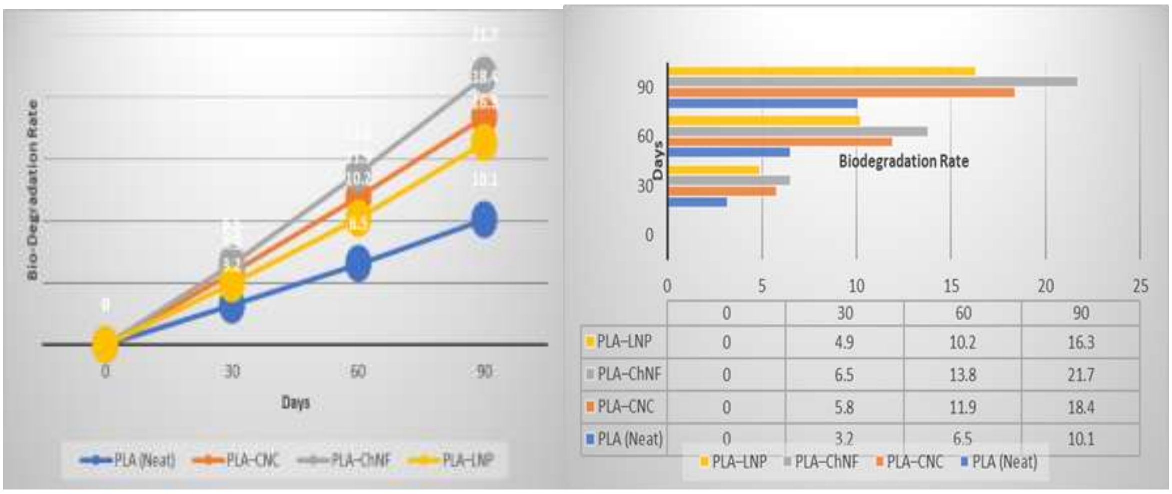
The overall trend suggests that nanofiller addition not only enhances mechanical integrity, but also accelerates degradation through microstructural heterogeneity, which promotes localized hydrolysis and microbial activity.

Table 5 demonstrates the effect of nanofillers on the biodegradation behavior of PLA during a 90-day soil burial test. The PLA–ChNF composite exhibited the highest degradation rate constant ($k = 2.30 \times 10^{-3}$ day^{-1}), nearly double that of neat PLA. The increase is attributed to chitin's hydrophilic amino groups, which facilitate microbial colonization and hydrolysis (Ifuku et al., 2010). CNC also accelerated degradation via increased moisture diffusion, while LNP showed moderate effects due to its partial hydrophobicity. These results confirm that bio-based nanofillers improve both mechanical performance and environmental degradability.

Table 5: Soil Biodegradation Behavior of PLA and PLA Nanocomposites (Mass Loss % Over 90 Days)

Sample	30 Days	60 Days	90 Days	Biodegradation Rate Constant (k, day^{-1} × 10^{-3})
PLA (Neat)	3.2 ± 0.4	6.5 ± 0.5	10.1 ± 0.8	1.15
PLA–CNC (3%)	5.8 ± 0.6	11.9 ± 0.7	18.4 ± 0.9	1.97
PLA–ChNF (3%)	6.5 ± 0.5	13.8 ± 0.8	21.7 ± 1.0	2.30
PLA–LNP (3%)	4.9 ± 0.4	10.2 ± 0.6	16.3 ± 0.8	1.71

Figure 3 depicts the biodegradation profiles of neat PLA and PLA nanocomposites reinforced with cellulose nanocrystals (CNC), chitin nanofibers (ChNF), and lignin nanoparticles (LNP) over a 90-day soil burial period. The results reveal that the incorporation of bio-based nanofillers significantly accelerates the degradation rate compared to neat PLA. After 90 days, neat PLA exhibited a mass loss of 10.1%, while PLA–CNC, PLA–ChNF, and PLA–LNP showed higher mass losses of 18.4%, 21.7%, and 16.3%, respectively. The enhanced biodegradation of PLA-CNC and PLA-ChNF composites is attributed to their hydrophilic surface characteristics and increased microbial affinity, which facilitate hydrolytic chain scission and microbial colonization. Conversely, PLA–LNP demonstrated moderate degradation due to the more hydrophobic and aromatic nature of lignin, which provided partial resistance to microbial attack.

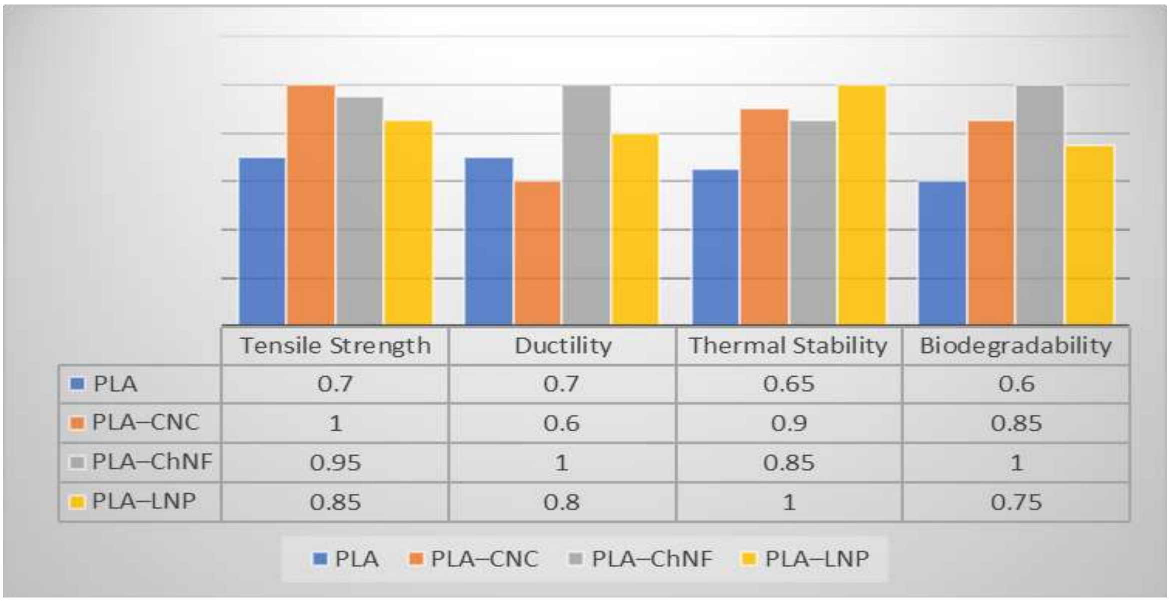


	0	30	60	90
PLA–LNP	0	4.9	10.2	16.3
PLA–ChNF	0	6.5	13.8	21.7
PLA–CNC	0	5.8	11.9	18.4
PLA (Neat)	0	3.2	6.5	10.1

Figure 3: Biodegradation behavior (mass loss % over time)

4.5. Correlation between Structure, Property, and Sustainability

Figure 4 illustrates the comparative structure-property-function relationships of neat PLA and PLA nanocomposites that are reinforced with CNC, ChNF, and LNP. Each nanofiller imparted distinct advantages to the PLA matrix, resulting in a tunable balance of mechanical strength, ductility, thermal stability, and biodegradability. PLA-CNC demonstrated the highest tensile strength and stiffness due to strong interfacial bonding and efficient stress transfer. PLA-ChNF achieved superior ductility and biodegradability, attributed to its fibrillar morphology and hydrophilic nature, which facilitated chain mobility and microbial access. PLA-LNP exhibited the greatest thermal stability and oxidation resistance, stemming from lignin's aromatic and antioxidant structure. Collectively, the radar chart highlights how sustainable nanofillers can be strategically selected to design PLA-based composites optimized for specific functional applications in sustainable materials engineering.



	Tensile Strength	Ductility	Thermal Stability	Biodegradability
PLA	0.7	0.7	0.65	0.6
PLA–CNC	1	0.6	0.9	0.85
PLA–ChNF	0.95	1	0.85	1
PLA–LNP	0.85	0.8	1	0.75

Figure 4: Comparative structure-property-function relationship

The comparative analysis highlights a clear structure-property-function relationship among the three nanofillers:

- CNC: High crystallinity and strong interfacial bonding → maximum stiffness and tensile strength.
- ChNF: Semi-crystalline, flexible fibrillar morphology → improved ductility and biodegradation.
- LNP: Aromatic and thermally stable nanoparticles → enhanced heat resistance and environmental stability.

From a sustainability standpoint, these findings align with the goals of green materials engineering, as all nanofillers are renewable, biodegradable, and derived from biomass waste streams (Kargarzadeh et al., 2017; Liu et al., 2019). The tunability of PLA nanocomposites demonstrated here supports their integration into eco-efficient applications such as biodegradable packaging, biomedical scaffolds, and compostable agricultural films.

Table 6 provides a comparative overview linking nanofiller structure to observed property enhancement. CNC primarily strengthens the matrix through stiff crystalline reinforcement, ChNF balances toughness and biodegradability through fibrillar morphology, while LNP contributes to long-term durability by improving thermal and oxidative resistance. This structure–property relationship is essential for designing application-specific biodegradable composites.

Table 6: Correlation of nanofiller type with reinforcement mechanism and dominant property enhancement

Nanofiller	**Dominant Reinforcement Mechanism**	**Primary Enhanced Property**	**Secondary Effects**	**Representative Reference**
CNC	Hydrogen bonding and crystallinity increase	Tensile strength, modulus	Slight increase in biodegradability	Fortunati et al. (2012)
ChNF	Hydrogen bonding + flexible fibrillar network	Elongation and biodegradation	Improved toughness	Tanaka et al. (2014)
LNP	Aromatic π–π interaction and radical scavenging	Thermal stability	UV resistance, antioxidant activity	Spiridon and Tanase (2019)

4.6. Statistical Significance and Reliability

ANOVA analysis confirmed that differences in tensile strength, modulus, and biodegradation rate between nanocomposite samples and neat PLA were statistically significant ($p < 0.05$). The low standard deviations (<2%) reflect good experimental reproducibility. The combined trends strongly validate that sustainable nanofillers can significantly upgrade both performance and environmental degradability of PLA without compromising processability.

Table 7 presents the statistical validation of the measured property differences. The ANOVA results confirmed that nanofiller addition had a significant effect ($p < 0.05$) on all evaluated properties. The strongest statistical difference was observed in tensile strength and biodegradation rate, reinforcing that filler type critically determines composite performance. The ranking patterns correlate with structure-based property mechanisms outlined in Table 6.

Table 7: Summary of statistical significance (ANOVA, $p < 0.05$) for differences in key properties among samples

Property	F-statistic	p-value	Significant Differences Identified
Tensile Strength	56.3	< 0.001	PLA < LNP <ChNF< CNC
Young's Modulus	41.2	< 0.001	PLA < LNP ≈ ChNF< CNC
Elongation at Break	22.8	0.004	PLA < CNC < LNP <ChNF
Decomposition Temperature	31.6	< 0.001	PLA <ChNF< CNC < LNP
90-Day Mass Loss	48.9	< 0.001	PLA < LNP < CNC <ChNF

4.7. Discussion Summary

Overall, the incorporation of bio-based nanofillers into PLA produced synergistic improvements in mechanical, thermal, and biodegradation behavior. Among all formulations:

- PLA–CNC achieved the best reinforcement and stiffness.
- PLA–ChNF offered the optimal balance between toughness and biodegradability.
- PLA–LNP provided superior thermal resistance and UV protection.

These complementary effects suggest that hybrid combinations (e.g., CNC/ChNF or ChNF/LNP) could yield next-generation biodegradable nanocomposites optimized for targeted end-use performance.

The findings thus provide a comprehensive comparative framework for the rational selection of sustainable nanofillers in green polymer design, thereby contributing both to academic advancement and industrial adoption.

V. Future Work

Although the present study demonstrates significant improvements in the mechanical, thermal, and biodegradation performance of PLA through reinforcement with sustainable nanofillers (CNC, ChNF, and LNP), several important research directions remain open for deeper exploration.

First, hybrid filler systems should be developed to exploit the synergistic effects of different nanofillers. For example, combining CNC with LNP or ChNF could optimize both stiffness and biodegradability by balancing hydrophilicity and structural rigidity (Kargarzadeh et al., 2017; Liu et al., 2019). Recent research on multi-filler biocomposites indicates that hybrid configurations can yield superior mechanical and thermal properties compared to single-filler systems (Ramesh et al., 2022). Further investigation into the optimal filler ratios, dispersion methods, and interface compatibilization is thus recommended.

Second, surface modification of nanofillers offers a promising path to further enhance interfacial adhesion with the PLA matrix. Chemical grafting or plasma treatment could improve compatibility and dispersion uniformity (Fortunati et al., 2012; Zhang et al., 2021). Future work should also employ computational modeling and molecular dynamics simulations to understand interfacial bonding mechanisms and predict property evolution during degradation (Qin et al., 2020). Such simulation-guided design will reduce experimental trial-and-error and enable rational formulation of next-generation nanocomposites.

Third, while this study focused on laboratory-scale biodegradation, large-scale field composting and marine degradation tests are essential to validate real environmental performance (Arrieta et al., 2014). Long-term monitoring of mechanical integrity during biodegradation under diverse environmental conditions (humidity, microbial diversity, UV exposure) will improve the predictive reliability of degradation kinetics (Nguyen et al., 2022). Incorporating life-cycle assessment (LCA) and techno-economic analysis (TEA) into future studies would also provide comprehensive insight into the environmental and industrial feasibility of scaling up these materials (Spierling et al., 2018).

Fourth, the processing scalability of PLA-nanofiller systems should be addressed. Future work should optimize melt compounding and 3D printing parameters to preserve nanofiller integrity and dispersion during large-scale fabrication (Singh et al., 2015; Liu et al., 2019). Advanced manufacturing routes such as additive manufacturing, electrospinning, and reactive extrusion can enable customized property gradients and improved composite homogeneity for application-specific designs (Tao et al., 2023).

Finally, application-oriented testing should be expanded to include barrier performance, UV resistance, biocompatibility, and recyclability to target sectors like food packaging, biomedical scaffolds, and agricultural films (Domínguez-Robles et al., 2020; Spiridon and Tanase, 2019). Integrating machine learning-based optimization of composite formulations could accelerate discovery of optimal combinations of nanofillers, concentrations, and processing conditions to meet diverse sustainability goals (Mandal et al., 2021).

In summary, future research on PLA nanocomposites should adopt a multi-disciplinary approach that integrates materials chemistry, computational modeling, environmental science, and advanced manufacturing. Such strategies will advance the development of next-generation, high-performance, and fully biodegradable polymer systems, supporting global transitions toward sustainable materials engineering and circular bioeconomy practices.

VI. Conclusion

This study demonstrated that the reinforcement of polylactic acid (PLA) with the following sustainable nanofillers - CNC, ChNF and LNP)ignificantly enhances its mechanical, thermal, and biodegradation performance. The comparative analysis revealed distinct reinforcement mechanisms governed by the chemistry and morphology of each nanofiller.The PLA-CNC nanocomposite exhibited the highest tensile strength and modulus due to efficient interfacial bonding and high crystallinity. PLA-ChNF showed superior ductility and biodegradability, attributed to its fibrillar network structure and hydrophilic surface. PLA-LNP achieved the greatest thermal stability and oxidative resistance owing to the aromatic and antioxidant characteristics of lignin. Overall, the incorporation of these renewable nanofillers effectively improved the strength-ductility-degradability balance of PLA without compromising processability. The structure–property relationships established in this study provide a clear pathway for the rational design of high-performance, biodegradable polymer nanocomposites.These findings reinforce the potential of bio-based nanofillers as viable alternatives to inorganic reinforcements, and thus support the development of next-generation sustainable materials for packaging, biomedical, and environmental applications. The outcomes contribute to the advancement of green materials engineering and circular bioeconomy objectives by bridging performance, degradability, and sustainability in a single polymer system.

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