Plasticized PLA Composites Enhanced with Cellulose Nanoparticles for Improved Mechanical and Thermal Performance

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Editor's note: Polylactic acid (PLA) is a bio-based polyester known for its good mechanical properties and biodegradability. However, its brittleness limits its applications. Nkuna et al. enhanced PLA by incorporating plasticizers such as triacetin (TA) and triethyl citrate (TEC), along with cellulose nanofibers (CNFs). The optimal loadings of CNFs determined in this study were 1 wt% for PLA/TEC and 3 wt% for PLA/TA. The findings revealed that the addition of plasticizers lowered the melting and glass transition temperatures, reduced crystallinity from 65% to 40-50%, and improved thermal stability. These results could facilitate the development of more sustainable polymer products, contributing significantly to reducing global plastic pollution.

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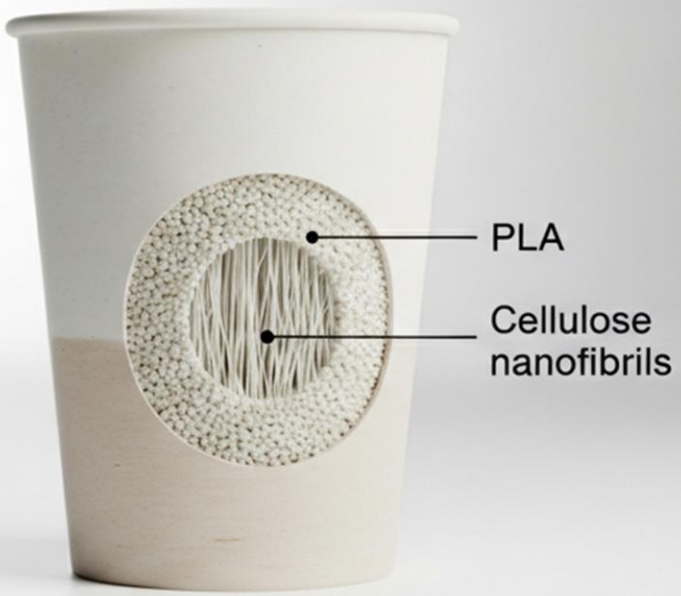

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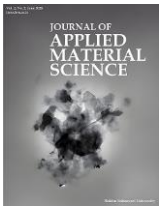

Original Research

Plasticized PLA Composites Enhanced with Cellulose Nanoparticles for Improved Mechanical and Thermal Performance

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Abstract

Polylactic acid (PLA) is a bio-based polyester widely used for its good mechanical properties, biocompatibility, and inherent biodegradability, which enables end-of-life options such as recycling and industrial composting. Despite these advantages, its brittleness, slow crystallization rate, and low crystallinity limit broader application. This study aims to enhance the performance of PLA through the development of biodegradable composite material by incorporating bio-based plasticizers and cellulose nanofibers (CNFs). Triacetin (TA) and triethyl citrate (TEC) were used as plasticizers to improve toughness and processability, while CNFs were added at loadings of 1, 2, and 3 wt% to promote crystallization and thermal stability. The thermal and mechanical properties of the PLA composites were evaluated using differential scanning calorimetry (DSC), thermogravimetric analysis (TGA), and tensile testing. DSC results showed that the addition of TA and TEC slightly reduced the melting temperature by approximately 4 °C and significantly lowered the glass transition temperature (T_g) by about 20 °C, indicating enhanced chain mobility. The combined incorporation of plasticizers and CNFs resulted in smaller or less perfect crystalline structures, reducing the crystallinity index from 65% for neat PLA to 40–50% for the composites. TGA revealed improved thermal stability, with an increase of approximately 10 °C in the onset degradation temperature compared to neat PLA. Mechanically, plasticization reduced tensile strength from 63.75 MPa to around 20 MPa while markedly increasing elongation at break from ~8% to ~400%. Optimal CNF loadings were identified at 1 wt% for PLA/TEC and 3 wt% for PLA/TA systems.

Keywords: Polylactic acid; Biobased plasticizers; Cellulose nanofibers; Bio-nanocomposites; Material properties.

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1. Introduction

Since 2019, global plastic production has exceeded 450 million tonnes annually, reflecting the continued dependence on plastics across industrial, commercial, and consumer sectors [1]. This large-scale production generates approximately 350 million tonnes of plastic waste each year, much of which remains inadequately managed [1, 2]. Current estimates indicate that only about 9% of plastic waste is recycled and approximately 12% is incinerated, while the remaining 79% accumulates in landfills or becomes mismanaged in the environment [2]. From this mismanaged fraction, an estimated 1–2 million tonnes of plastic enter the oceans annually, contributing significantly to marine pollution and ecosystem degradation [3-5].

Environmental exposure causes plastics to gradually fragment into microplastics and nano-plastics, which are now detected in terrestrial, freshwater, and marine ecosystems [6]. These particles can be ingested by organisms and subsequently transferred through food webs, raising concerns regarding their potential ecological and human health implications [7, 8]. In addition to environmental contamination, plastics contribute substantially to climate change. The United Nations Environment Programme estimated that the production, use, and disposal of plastics generated approximately 860 million metric tonnes of greenhouse gas emissions globally in 2019, highlighting the need for more sustainable material alternatives [5].

Among emerging alternatives, polylactic acid (PLA) has attracted considerable attention as a biodegradable thermoplastic derived from renewable resources such as corn starch and sugarcane [9, 10]. PLA exhibits favourable mechanical properties, good transparency, and excellent processability, making it suitable for applications including packaging, biomedical devices, and additive manufacturing [10, 11]. However, its inherent brittleness, relatively low thermal resistance, and slow crystallisation kinetics limit its performance in demanding applications [12-14]. Consequently, recent research has focused on the incorporation of nanoscale reinforcements together with bio-based plasticisers to enhance the toughness and flexibility of PLA while maintaining or improving its thermal stability [13, 15, 16].

Cellulose, the most abundant natural polymer on Earth, is a biodegradable polysaccharide composed of β-D-glucopyranose units linked through β-(1→4) glycosidic bonds [17, 18]. Its hierarchical structure contains both ordered crystalline domains and disordered amorphous regions, which strongly influence its mechanical and physicochemical properties [18]. The breakdown of cellulose fibres yields nanoscale derivatives collectively referred to as nanocellulose, which include cellulose nanofibrils (CNFs), cellulose nanocrystals (CNCs), and bacterial cellulose (BC) [18, 19].

CNFs are long, flexible nanostructures containing both crystalline and amorphous regions and can be obtained from various lignocellulosic sources such as wood pulp, cotton, and agricultural residues [18-20]. Typically, CNFs possess diameters ranging from 4 to 50 nm and lengths between 100 and 10 000 nm, with crystallinity values between 54% and 96% depending on the source material and extraction method [18]. Their high aspect ratio, high surface area, and strong hydrogen-bonding capability make them attractive reinforcement agents for biodegradable polymer matrices [18, 19]. As a result, CNFs have been widely incorporated into PLA matrices to enhance mechanical performance and structural integrity [13, 16, 21-25].

Agbakoba investigated PLA/CNF composites containing 1–3 wt% CNFs for fused deposition modelling (FDM) 3D printing, incorporating triacetin as a plasticiser to improve filament flexibility and reduce brittleness. The composite containing 1 wt% CNF exhibited a 12% increase in tensile strength, together with improved tensile modulus and elongation relative to neat PLA. The resulting filaments demonstrated sufficient strength, flexibility, and thermal stability to enable low-temperature 3D printing without warping [11].

Nanocellulose reinforcement has also demonstrated promising performance in other polymeric systems. Mohammadalipour et al. reported electrospun polyhydroxybutyrate (PHB)–chitosan scaffolds reinforced with 1–5 wt% CNCs, which exhibited improved tensile strength (up to 4.52 MPa), enhanced thermal stability (245 °C to 252 °C), increased surface roughness, and reduced water contact angle, indicating potential for cartilage tissue regeneration [26]. Similarly, Mohammadalipour et al. developed electrospun PHB/CNF scaffolds for bone tissue engineering that showed increased crystallinity (≈46% to 53%), improved hydrophilicity, and an approximately 89% increase in toughness compared with pure PHB [27]. In another study, Zhou et al. synthesised polyvinyl alcohol (PVA)/CNF/gelatin hybrid aerogels via freeze-drying,

where gelatin facilitated hydrogen-bonding interactions between PVA and CNFs. The resulting materials exhibited significantly improved mechanical properties, with the PVA/CNF/G3 aerogel demonstrating a modulus of 1.65 MPa, approximately eight times greater than that of PVA/CNF and ninety-one times greater than neat CNF, together with low density and thermal conductivity suitable for insulation applications [28].

Despite these advantages, CNFs tend to agglomerate due to strong intermolecular hydrogen bonding between hydroxyl groups on adjacent fibrils [29]. Such aggregation can limit effective dispersion within polymer matrices and reduce reinforcement efficiency. To address this challenge, several surface modification strategies have been explored, including silylation, urethanisation, amidation, and acetylation, as well as the incorporation of plasticisers to improve compatibility between CNFs and polymer matrices [18, 30, 31].

Plasticisers represent an effective approach for modifying the properties of PLA by enhancing polymer chain mobility, reducing intermolecular interactions, and improving processability [15]. Commonly used plasticisers for PLA include polyethylene glycol (PEG), citrate esters, glycerol derivatives, oligomeric lactic acid, and glucose-based compounds [32]. In addition to improving flexibility and ductility, plasticisers can facilitate the dispersion of nanocellulose within the polymer matrix and enhance interfacial interactions between the reinforcing phase and PLA [32]. For instance, Srisawat et al. investigated the effect of PEG plasticiser on PLA/CNF composite and reported a 400% increase in ductility while maintaining CNF reinforcement of tensile strength and modulus [23].

In this study, a dual-modification strategy is proposed that integrates bio-based plasticisers with cellulose nanofibrils within a fully biodegradable PLA matrix. While numerous studies have investigated PLA/nanocellulose composites, relatively few have explored the combined incorporation of bio-based plasticisers such as triacetin (TA) or triethyl citrate (TEC) to simultaneously improve CNF dispersion and matrix flexibility. To address this gap, a CNF-reinforced PLA masterbatch will be synthesised using a solvent-casting [33, 34] approach to promote homogeneous nanofibril distribution and minimise agglomeration. This approach also offers potential scalability for industrial processing. By achieving improved CNF dispersion and balanced mechanical performance, the resulting PLA composites aim to combine enhanced toughness and strength, providing a sustainable material platform for the additive manufacturing of single-use biomedical devices.

2. Experimental

2.1. Materials

PLA; grade LX175; density 1.24 g cm^{-3}; melt flow index 3 g/10 min at 190 °C under a 2.16 kg load) was sourced from Total Energies Corbion (Netherlands). The bio-based plasticizers TEC (Mw = 276.29 g mol^{-1}) and TA (Mw = 218.21 g mol^{-1}), each with a purity of at least 99%, were supplied by Inqaba Biotechnical Industries (Pty) Ltd. Dichloromethane (DCM) was obtained from Associated Chemical Enterprises (ACE). The CNF was sourced from Valida S191C 8% Batch SB-20-0126-01, provided by Sappi.

2.2. PLA/TEC/CNF and PLA/TA/CNF preparation

CNFs were dispersed in an acetone/ethanol/ethyl acetate mixture (3:3:1 v/v) and stirred for 24h to minimize agglomeration [11, 35]. Separately, PLA (10 g) was dissolved in 100 mL DCM, mixed with the CNFs suspension, and sonicated for 5 min at 80 Amp. The blend was cast on aluminium plates and dried for 24 h to form a PLA/CNF masterbatch. The masterbatch was granulated and compounded with neat PLA and bio-based plasticizers (TA or TEC) using a Nanjing Giant A SHJ twin-screw extruder (140–180 °C). The extrudates were pelletized, oven-dried at 60 °C for 24 h, and injection-moulded into test specimens with a TMC30H machine at 130–185 °C. Table 1 lists the formulations of neat PLA and PLA composites containing CNFs and plasticizers.

2.3. Measurements

Material composition was analysed using a PerkinElmer Spectrum Two™ FT-IR spectrometer (ATR mode) with 16 scans over 4000–500 cm^{-1} at 4 cm^{-1} resolution to identify functional groups and confirm molecular interactions. Thermal transitions of neat PLA and PLA composites were examined using a PerkinElmer DSC 6000 under nitrogen (19.8 mL min^{-1}). Samples were heated from −40 °C to 200 °C at 10 °C min^{-1} to determine glass transition (T_g), cold crystallization (T_{cc}), and melting (T_m) temperatures.

Table 1. Formulation of PLA reinforced with CNC and biobased plasticizers

Samples	PLA (wt. %)	Plasticisers (wt. %)		CNF (wt. %)
		TA	TEC	
A	100	-	-	-
A(TA)_1C	86	15	-	1
A(TA)_2C	85	15	-	2
A(TA)_3C	84	15	-	3
A(TEC)_1C	86	-	15	1
A(TEC)_2C	85	-	15	2
A(TEC)_3C	84	-	15	3

Crystallinity (X_c) was calculated after the second heating using:

$$X_c = \frac{\Delta H_m - \Delta H_{cc}}{w\Delta H_m^{\circ}} \times 100 \quad (1)$$

where ΔH_m and ΔH_{cc} are the melting and cold-crystallization enthalpies, w is the PLA weight fraction, and ΔH_m° is the enthalpy of a 100% crystalline polymer (PLA: 93 J g^{-1}; PBAT: 114 J g^{-1}) [9]. Thermal stability was evaluated using a PerkinElmer TGA-4000. Samples (5–10 mg) were heated from room temperature to 600 °C at 10 °C min^{-1} under nitrogen. Tensile properties were measured on a Lloyds RX EZ50 tester at room temperature according to ASTM D638 and ISO 527-2. Specimens (165 × 10 × 4 mm) were tested at 50 mm min^{-1}, and results were averaged over five samples.

3. Results and discussion

3.1. FTIR Spectroscopy

The FTIR spectra of neat PLA and PLA/CNF/plasticizer composites showed no significant differences, indicating that the chemical structure of PLA was preserved after incorporation of CNFs and bio-based plasticizers, as shown in Figure 1. CNF showed a broad –OH stretching band at 3338 cm^{-1} and 2900cm^{-1}, typical of cellulose hydroxyl groups and C-H stretching (both asymmetric and symmetric) within the cellulose backbone, respectively [26, 27, 35-38]. The CNF also displayed C-O stretching vibration in the glycosidic linkages, CH_2 scissoring and rocking vibrations, and bending vibrations of absorbed water as

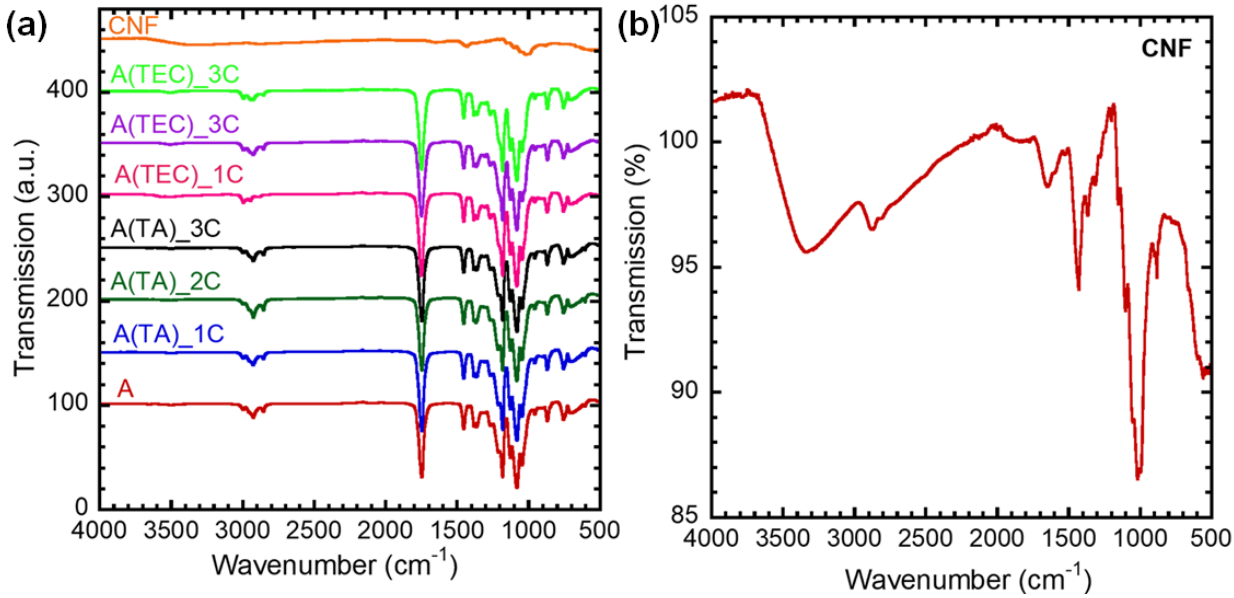

Figure 1. FTIR spectrum for (a) neat PLA, PLA/TEC/CNF, and PLA/TA/CNF, and (b) CNF.

seen at wavenumbers 1050, 1400, and 1600 cm^{-1}, respectively [26, 27, 35-39]. The CH_2 stretching vibrations at ~2900 cm^{-1} and the C–O stretching band at ~1100 cm^{-1} remained unchanged across all samples, confirming the stability of the PLA backbone [40]. The characteristic carbonyl (C=O) stretching band appeared at ~1700 cm^{-1} for all materials; however, its intensity increased in the PLA/CNF/plasticizer composites compared to neat PLA. This increase suggests enhanced intermolecular interactions, such as hydrogen bonding between PLA carbonyl groups and hydroxyl groups from the additives [40]. No new absorption bands or peak shifts were detected, indicating the absence of new covalent bond formation. Overall, the FTIR results confirm that CNFs and plasticizers are incorporated through physical interactions rather than chemical modification of PLA.

3.2. TGA analysis

TGA was used to evaluate the influence of CNFs and bio-based plasticizers on the thermal stability of the PLA composites, as seen in Figure 2. Neat PLA exhibited a typical single-step degradation profile with an onset degradation temperature (T_{onset}) of approximately 350 °C and a maximum degradation temperature (T_{max}) of 372 °C, consistent with previously reported PLA thermal behaviour [9, 14, 16]. PLA degradation is mainly associated with random chain scission, intramolecular transesterification, and depolymerization reactions that produce lactide and oligomeric species at elevated temperatures [41, 42]. The incorporation of CNFs and plasticizers generally improved thermal stability [13], with T_{on}set increasing by about 10 °C for most composite formulations. Similarly, T_{max} shifted to higher values in the range of 380–385 °C, indicating delayed thermal decomposition. This improvement is primarily attributed to the reinforcing and barrier effects of CNFs within the polymer matrix [43, 44]. Due to their high aspect ratio and strong hydrogen-bonding capability, CNFs can form an interconnected network that restricts polymer chain mobility and slows the diffusion of volatile degradation products [13]. Furthermore, citrate-based plasticizers such as TA and TEC can enhance nanofibril dispersion by increasing chain mobility during processing, thereby improving interfacial interactions between CNFs and PLA [29].

However, the A(TA)_3C formulation deviated from this trend, exhibiting only a marginal increase in T_{onset} (~1 °C) and a T_{max} of 374 °C. The reduced thermal stability at higher CNF loading may be attributed to partial nanofibril agglomeration caused by strong intermolecular hydrogen bonding between adjacent fibrils [13, 16, 32, 45, 46]. Such aggregation reduces effective interfacial interactions with the PLA matrix and can act as localized defects that accelerate degradation. The residual mass at 600 °C further illustrates the reinforcing role of CNFs in the composites. Neat PLA

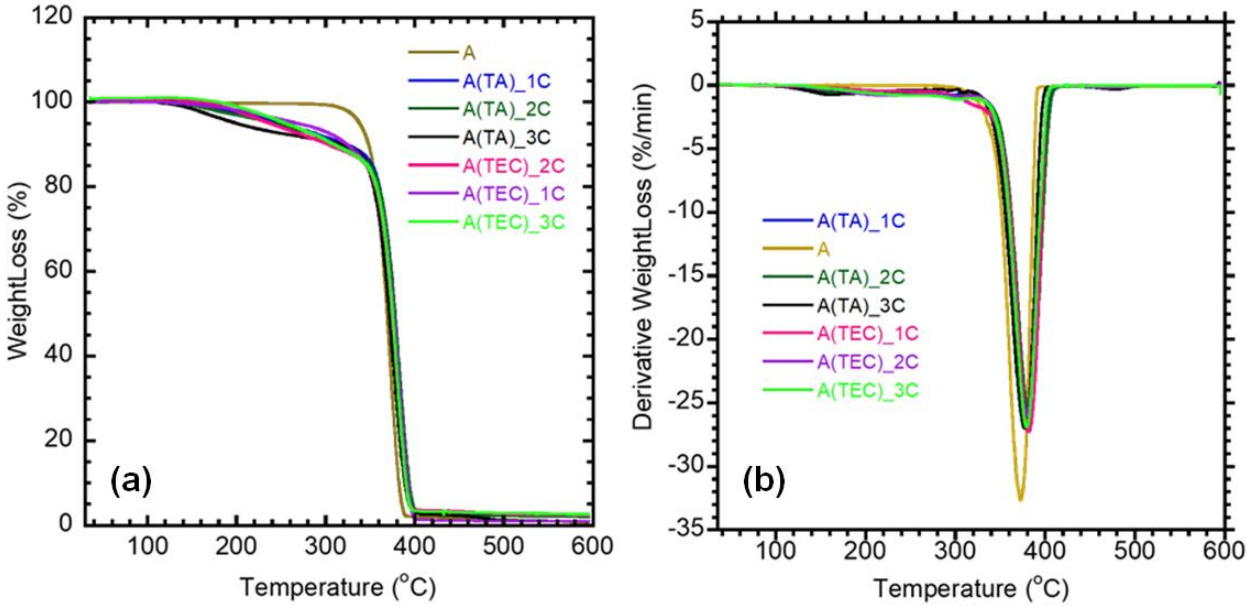

Figure 2. FTIR spectrum for (a) neat PLA, PLA/TEC/CNF, and PLA/TA/CNF, and (b) CNF.

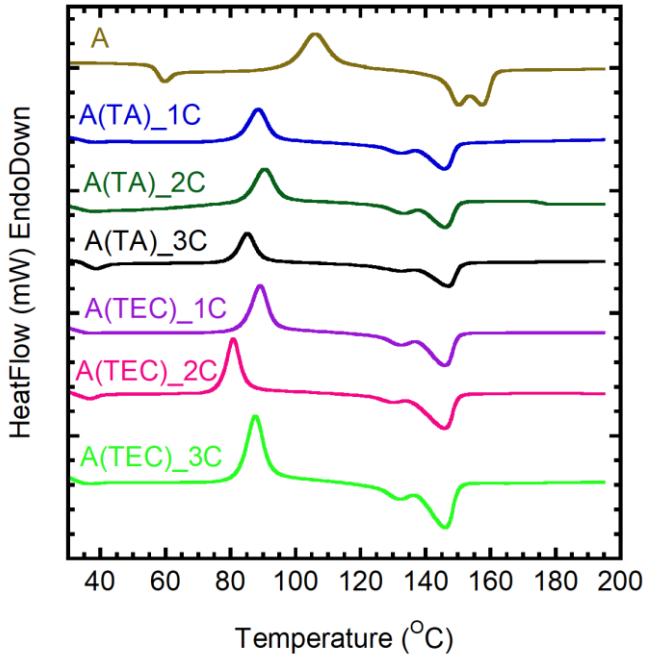

Figure 3. DCS thermograms for PLA reinforced with CNF and biobased plasticizers.

showed a very low char residue of ~0.69%, whereas most composite formulations exhibited higher residues of around 2.7%. This increase is attributed to the carbonization of nanocellulose during thermal decomposition, which forms thermally stable carbonaceous structures that act as protective barriers against heat and mass transfer [24, 30]. A slightly lower residue observed for A(TEC)_1C may result from the stronger plasticizing effect of TEC, which enhances chain mobility and promotes more complete volatilization during degradation. Among the formulations, A(TA)_2C exhibited the best thermal performance with a T_{onset} of 364 °C and a T_{max} of 384 °C, indicating an optimal balance between CNF reinforcement and plasticizer content.

3.3. DCS analysis

DSC analysis revealed a strong influence of plasticizers and CNFs on the thermal transitions of the PLA matrix, as shown in Figure 3 and Table 2. The T_g decreased significantly from approximately 65 °C for neat PLA to around 40 °C for the modified composites, confirming the effective plasticization of the polymer system. This reduction in T_g is primarily attributed to the incorporation of low–molecular–weight plasticizers such as TA and TEC, which increase the free volume within the polymer matrix and weaken intermolecular interactions between PLA chains [11]. As a result, segmental mobility of the polymer chains increases, allowing molecular rearrangements to occur at lower temperatures [11, 29]. The presence of CNFs may also contribute to this behaviour by disrupting the continuity of the PLA matrix at the nanoscale, particularly when the nanofibrils are well dispersed. Furthermore, the T_{cc} shifted to lower values, indicating that the enhanced chain mobility enables PLA chains to reorganize into ordered crystalline structures more readily during heating. Similar behaviour has been reported in plasticized PLA systems where increased molecular mobility promotes earlier crystallization during thermal scanning [29, 47].

The T_m also decreased from approximately 160 °C to around 130 °C following the incorporation of plasticizers and CNFs, suggesting modifications in the crystalline structure of the material. Plasticizers such as TA and TEC interfere with the regular packing of PLA chains, leading to the formation of crystals with reduced lamellar thickness and lower structural perfection [48, 49]. At the same time, CNFs can act as heterogeneous nucleation sites for PLA crystallization, promoting the formation of crystalline domains while simultaneously restricting crystal growth when present at higher

Table 2. Thermal properties of PLA reinforced with CNF and biobased plasticizers

Sample	T_g (°C)	T_{cc} (°C)	ΔH_{cc} (J/g)	T_{m1} (°C)	T_{m2} (°C)	ΔH_m (J/g)	X_c (%)
A	61.8	108.7	-27.6	150.4	157.9	32.5	64.6
A(TA)_1C	37.0	88.5	-17.2	132.1	145.9	20.1	47.8
A(TA)_2C	34.0	78.2	-19.2	127.6	145.1	23.8	55.8
A(TA)_3C	38.6	85.2	-16.6	132.0	147.1	21.1	49.4
A(TEC)_1C	36.1	89.2	-19.3	132.3	145.9	22.1	53.0
A(TEC)_2C	36.1	80.9	-19.3	129.3	145.9	23.5	55.5
A(TEC)_3C	38.1	80.7	-20.7	129.4	146.2	27.8	63.6

concentrations [21, 22]. This dual effect often results in a greater number of smaller or less perfect crystals, which can lower the T_m while still influencing the overall crystallization behaviour of the polymer matrix [13, 46]. The combined effects of plasticization and nanofiller incorporation, therefore, modify both the crystallization kinetics and crystalline morphology of PLA [29]. These changes in thermal transitions indicate that the introduction of TA, TEC, and CNFs effectively tailors the thermal response of the PLA matrix, enabling improved processability and flexibility while maintaining the structural characteristics required for composite applications [13, 29].

3.4. Mechanical Properties

The tensile behaviour of the composites was predominantly controlled by the presence of plasticizers, which reduced the reinforcing effect typically associated with CNFs [50, 51]. Figure 4 presents representative tensile stress–strain curves, showing that neat PLA exhibited typical brittle fracture with abrupt failure at low strain, whereas the incorporation of polymeric plasticizers markedly improved its ductility [29]. The decrease in tensile strength is from approximately 65 MPa for neat PLA to about 30 MPa for the plasticized composites (Table 3). This reduction is characteristic of plasticized PLA systems and is attributed to the increased chain mobility induced by low–molecular–weight plasticizers [29, 32]. Both TA and TEC act by inserting between PLA chains, weakening intermolecular interactions and increasing free volume within the polymer matrix [29, 32]. This plasticization mechanism facilitates chain slippage under tensile loading, resulting in reduced strength but improved flexibility [29]. The effect is further supported by the

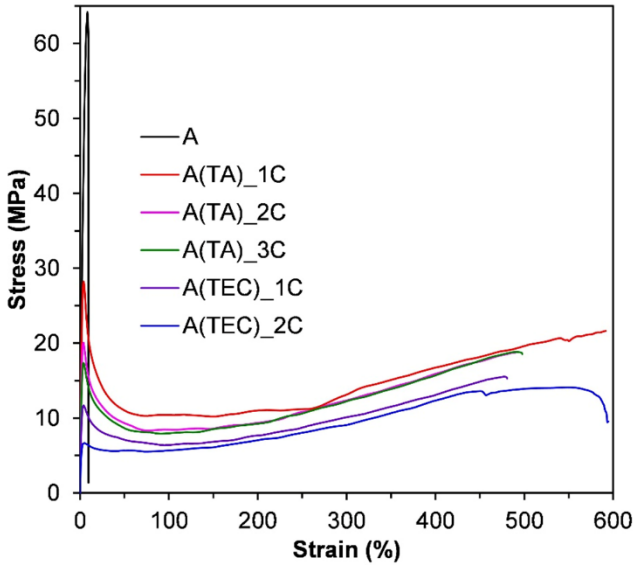

Figure 4. Stress-strain curves for neat PLA and PLA reinforced with CNF and biobased plasticizers.

Table 3. Mechanical properties of PLA reinforced with CNF and biobased plasticizers

Sample	Tensile Strength (MPa)	Young Modulus (MPa)	Elongation at break (%)
A	63.8 ±1.1	1751.5 ±74	7.6 ±0.9
A(TA)_1C	15 ±0.5	844.3 ±130	471.7 ±5.4
A(TA)_2C	13.6 ±0.5	493.2 ±198	519.7 ±63.3
A(TA)_3C	32.8 ±2.8	1530.2 ±130	231 ±186
A(TEC)_1C	29.1 ±4.8	1465.8 ±105	467.8 ±157
A(TEC)_2C	19.7 ±1.4	1253.3 ±66	487.5 ±22
A(TEC)_3C	17.9 ±1.4	1053.5 ±85	545.8 ±80

observed decrease in T_g from 65 °C to around 40 °C, confirming enhanced molecular mobility in the plasticized systems [11].

The crystallinity index also decreased from approximately 62% for neat PLA to around 45% in the composites, indicating that plasticization disrupted the regular packing of polymer chains during crystallization. Reduced crystallinity generally leads to lower stiffness and strength but contributes to greater ductility due to the larger amorphous fraction that allows easier chain movement [4]. Although CNFs are known to act as nucleating agents that promote heterogeneous crystallization and improve mechanical reinforcement in PLA matrices, the strong plasticization effect appears to have limited polymer–filler interactions in most formulations [13, 16, 24, 27, 29, 46, 52, 53]. Nevertheless, partial recovery of stiffness and tensile strength was observed for A(TA)_3C and A(TEC)_1C, which may be attributed to improved CNF dispersion and stronger interfacial hydrogen bonding between the hydroxyl groups of cellulose and the carbonyl groups of PLA, enabling more efficient stress transfer within the composite structure [29]. At higher CNF loadings, PLA plasticized with TEC exhibited reduced strength compared with TA-plasticized composites due to the stronger plasticizing effect of TEC, which increases free volume and chain mobility within the PLA matrix [15, 29, 42, 54]. This increased mobility may promote CNF agglomeration and weaken stress transfer between the nanofibers and the matrix [29], resulting in a lower modulus compared with A(TA)_3C.

4. Conclusions

This study demonstrates that the combined incorporation of bio-based plasticizers and cellulose nanofibers effectively tailors the thermal and mechanical behaviour of PLA without altering its chemical structure. Plasticization significantly enhanced chain mobility, leading to reduced glass transition, melting, and T_{cc} temperatures, improved flexibility, and increased elongation at break, although at the expense of tensile strength and stiffness. While the reinforcing potential of CNFs was largely suppressed by the dominant plasticizing effect, selected formulations (A(TEC)_1C and A(TA)_3C) showed partial recovery of mechanical and thermal performance, highlighting the importance of composition and dispersion. Thermal degradation resistance and char yield were generally improved in the composites, indicating enhanced thermal stability. FTIR analysis confirmed that these changes arose, explaining the physical interactions rather than chemical modification. Overall, the results underline the potential of bio-based plasticizers and CNFs to produce more flexible and thermally stable PLA-based materials, provided that an optimal balance between plasticization and reinforcement is achieved.

Acknowledgments

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Conflict of Interest

The authors declare no conflict of interest.

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