Design and Optimization of PLA-Based Nanocomposites: Enhancing Performance through Nanofillers and Solvent Systems

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***Abstract*—The transition from conventional fossil-based polymers to biopolymers is essential to promote sustainability and mitigate the environmental impact of persistent thermoplastic waste. Among bio-based alternatives, polylactide (PLA) has emerged as a leading candidate due to its derivation from renewable resources, biodegradability, and favorable mechanical and barrier properties. PLA holds significant promise in fields such as food packaging, biomedical devices, filtration, and technical textiles. However, its brittleness, low melt strength, and slow crystallization hinder its broader applicability. One of the most preferred and economical method to overcome the limitations of PLA is the incorporation of nanofillers to develop PLA-based nanocomposites. A variety of nanofillers—including nanoclay, nanosilica, graphene, carbon nanotubes, and cellulose nanocrystals (CNCs)—have been employed for this purpose. Among these, CNCs—renewable, biodegradable polysaccharide nanoparticles—stand out due to their low density, high specific surface area, and excellent mechanical characteristics. These attributes position CNCs as attractive candidates for the development of environmentally sustainable nanocomposite materials. Owing to their multifunctionality, CNCs have demonstrated significant potential across a broad range of applications, including mechanical reinforcement, barrier films, biomedical devices, drug delivery systems, electrospun nanofibers, and technical textiles. Despite their promise, CNCs tend to aggregate in hydrophobic polymer matrices due to surface hydroxyl groups and sulphate half-esters introduced during acid hydrolysis. Factors such as PLA molecular weight and crystallization behavior significantly influence CNC dispersion and final nanocomposite properties. Moreover, the solution casting method provides superior dispersion compared to melt processing, where CNC agglomeration is often observed. To further enhance CNC dispersion in solution-based systems, approaches such as use of surfactants, surface modification, and selection of highly polar solvents have been revealed.**

This review aims to explore the enhancement of material properties through the incorporation of different types of nanofillers into the polylactide matrix and to assess the key conditions necessary for achieving uniform dispersion of nanoparticles, especially cellulose nanocrystals, within PLA-based polymer systems.

***Keywords*—Dispersion, nanocomposites, nanoparticles, polylactide.**

I. INTRODUCTION

There has been a growing global tendency to replace conventional petroleum-based plastics with biodegradable polymers, driven by increasing environmental concerns, stricter regulations on plastic waste, and the urgent need to reduce carbon footprints. These eco-friendly materials, often derived from renewable resources, not only help minimize dependence on fossil fuels but also offer end-of-life options like composting or controlled biodegradation, thereby reducing persistent pollution. Advances in material science have expanded their mechanical, thermal, and barrier properties, enabling their application across packaging, textiles, biomedical devices, and agricultural products. This tendency is additionally strengthened by growing consumer preference for environmentally friendly products and by industries aiming to adopt circular economy practices, thereby accelerating the shift toward a more sustainable materials economy.

Among biopolymers, polylactide (PLA) is known as a particularly promising candidate owing to its production from renewable resources, i.e., sugarcane and corn starch. Its combination of biocompatibility, non-toxicity, cost-effectiveness, and favorable physicochemical properties such as high modulus and tensile strength, optical clarity, and excellent barrier performance positions PLA as a viable substitute for conventional polymers including polyethylene terephthalate (PET) and polystyrene (PS) in a broad range of applications [1]. Furthermore, the U.S. Food and Drug Administration has designated PLA as “Generally Recognized as Safe” for use in food-contact applications [2]. Beyond its aforementioned advantages, the processing of PLA is constrained by certain limitations such as brittleness [3], slow crystallization rates [4,5], and low melt strength [6,7]. A practical and cost-effective approach to address these drawbacks and enhance its processability involves incorporating nanoparticles into PLA matrix, thereby producing PLA-based nanocomposites [8].

Nanoparticles can substantially enhance the performance of polymer matrices in nanocomposite systems by exploiting their high surface area and interfacial interactions with the polymer chains [9]. Their incorporation can improve mechanical properties through efficient stress transfer [10], enhance barrier performance by inducing a tortuous diffusion pathway and

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increase thermal stability by restricting chain mobility [11]. Besides, nanoparticles act as effective nucleating agents, promoting faster crystallization and higher crystallinity, which in turn benefit mechanical and barrier characteristics [12]. Additionally, nanoparticle–polymer interactions can improve melt strength and processing stability, while surface functionalization enables the introduction of tailored properties, i.e., antioxidant and antimicrobial activity, electrical conductivity, flame retardancy, or UV resistance, thereby expanding the applicability of the resulting materials.

With respect to PLA, various nanoparticles, such as graphene, nanoclay, carbon nanotube, nanosilica and cellulose nanocrystals have been used in various studies [13-21]. Cellulose nanocrystals (CNCs) are nanoscale polysaccharides characterized by low density, high surface area, and superior mechanical properties. Their bio-based, biodegradable nature and natural abundance render them highly promising for the development of sustainable nanocomposites [22]. CNCs can be extracted primarily through sulfuric acid hydrolysis [23]. CNCs find extensive applications across polymer matrix reinforcement, barrier films, biomedical implants, drug delivery systems, technical textiles and nanofibrous webs [23, 24]. Nonetheless, the hydroxyl groups on CNC surfaces, coupled with sulfate half-ester groups formed as a result of sulfuric acid hydrolysis, promote agglomeration within hydrophobic polymer matrices. Furthermore, intrinsic matrix characteristics including crystallizability and polymer molecular weight affect nanoparticle dispersion and consequently the performance of PLA-based nanocomposites.

This review aims to investigate the improvement of material properties achieved by incorporating various nanofillers into the polylactide matrix, with a particular focus on evaluating the critical parameters required for attaining homogeneous dispersion of nanoparticles within PLA-based nanocomposites.

II. PROCESSING PLA-BASED NANOCOMPOSITES

A. *Solution Casting*

In the solution casting, a polymer solution prepared in an appropriate solvent or solvent blend is combined with nanoparticles. The solvent is used to pre-disperse nanoparticles, and the resulting dispersion is subsequently mixed with the polymer solution. Vatansever et al. [25] reported that, compared to samples prepared without sonication, those subjected to sonication exhibited a more remarkable increase in complex viscosity at lower frequencies indicating that water-bath sonication enables a more homogeneous CNC dispersion than magnetic stirring. Gao et al. [26] reported that increasing ultrasonication time and power can substantially improve the dispersion of carbon nanotubes/graphene oxide powders in solution, up to the point where a dispersion plateau is achieved.

This method is considered a practical fabrication route for nanocomposites due to the ease of nanoparticle processing in a solvent medium and its suitability for thermally sensitive nanoparticles that would otherwise degrade at elevated temperatures. Moreover, it has been shown to produce superior CNC dispersion in PLA compared to melt processing, in which CNC agglomeration is frequently observed [25, 27]. To further improve CNC dispersion, various strategies have also been proposed, including the use of surfactants, CNC surface modification, and the utilization of polar solvents. For non-polar solvents, i.e., chloroform and dichloromethane (DCM), the studies have employed surfactant addition [28] or nanoparticle surface modification [29] to improve compatibility. In other studies [30, 31], dimethylformamide (DMF), a highly polar solvent having a high dielectric constant, was used to promote uniform CNC dispersion in PLA. Ozdemir and Nofar [30] also demonstrated that dimethyl sulfoxide (DMSO), a non-toxic solvent having a similarly high dielectric constant, was even more successful in improving CNC dispersion than DMF. Palak et al. [32] reported that due to its low dielectric constant, pure DCM resulted in the poorest CNC dispersion; whereas, solvent blends, especially DCM50/DMSO50, achieved the most homogeneous CNC dispersion across various PLA grades. This result is attributed to the higher dielectric constant of DMSO, which promotes stronger interactions with the hydroxyl groups and, in the case of sulfuric acid–hydrolyzed CNCs, the negatively charged sulfate half-esters on the CNC surface. These functional groups interact more favorably with highly polar solvents through hydrogen bonding and electrostatic stabilization, thereby reducing interparticle attractions and minimizing agglomeration.

B. *Melt Process*

Melt process offers notable advantages compared to solution casting method: It eliminates the need for solvents, thereby enhancing environmental sustainability, and the issue of residual solvent negatively affecting the material properties is eliminated in the melt-processing method. Additionally, it is fully compatible with established polymer processing methods, including extrusion and injection molding, which are widely employed in polymer industry.

On the other hand, dispersion of nanoparticles within polymer matrices during melt mixing presents a considerable challenge due to both thermodynamic incompatibility and processing limitations. The disparity in surface chemistry between hydrophilic nanoparticles and hydrophobic polymers results into poor interfacial adhesion and a strong thermodynamic tendency for nanoparticles to agglomerate. The problem is exacerbated by the high surface energy of nanoparticles and the strong intermolecular interactions between them, which resist breakup under conventional shear forces [33]. Although melt mixing relies on mechanical shear to disperse fillers, the viscous nature of polymer melts hinders the penetration of shear forces into tightly bound agglomerates, while excessive shear can degrade either the polymer or the nanoparticles. Even when partial dispersion is achieved, the absence of effective interfacial stabilization allows re-agglomeration to occur during processing or cooling. Consequently, without surface modification, compatibilizers, or alternative preparation techniques, achieving a stable and homogeneous nanoparticle dispersion in polymer melts remains inherently difficult. In the study of Zhou et al. [16], PLA was modified with poly(butylene adipate-co-terephthalate) (PBAT) and carbon nanotubes (CNTs–COOH) via melt process, while a compatibilizer, facilitating the bonding of CNTs–COOH to the PLA/PBAT matrix through interactions between the epoxy groups of E-BA-GMA and the carboxyl groups on the nanoparticles, was used. Zhu et al. [34] demonstrated that oleic acid functionalization of silica nanoparticles improved the rheological behavior of PLA/silica nanocomposites prepared via melt mixing, an effect attributed to the enhanced interfacial adhesion provided by the functionalized nanoparticles.

As an alternative preparation approach, the solution casting method can be combined with melt mixing, wherein a solution-cast polymer/nanoparticle masterbatch is subsequently diluted through melt mixing [27, 35, 36].

III. PERFORMANCE OF PLA-BASED NANOCOMPOSITES

A. Mechanical Performance

The nanoparticle incorporation into the PLA matrix can significantly enhance its mechanical performance when appropriate dispersion and loading levels are achieved. Well-dispersed nanoparticles act as nucleating agents, fostering crystallization and thereby increasing stiffness and strength. In addition, the large interfacial area between the nanoparticles and PLA enables efficient stress transfer, contributing to improved tensile properties. The restricted mobility of polymer chains around the nanoparticles further enhances rigidity, while their uniform distribution hinders crack initiation and propagation, improving toughness. However, these benefits are typically observed at low to moderate nanoparticle contents, as excessive loadings may lead to agglomeration and reduced chain mobility, ultimately impairing mechanical performance. In the study of Younus et al. [37], tensile testing was conducted to assess the mechanical properties of PLA and its CNT-reinforced nanocomposites, and the results revealed a substantial improvement in toughness with CNT incorporation, reflected by the remarkable increase in elongation at break. Compared to neat PLA (11%), the nanocomposites exhibited elongation values of 24.4, 30.3, 51.8, and 42.7% for 0.5, 1, 3, and 5 wt% CNTs, respectively. In contrast, the tensile strength and Young’s modulus decreased with CNT addition. This suggests that although CNTs reinforce PLA by enhancing ductility, their incorporation also introduces structural defects that compromise strength and stiffness. Both Lai et al. [38] and Acik et al. [39] demonstrated that the incorporation of small amounts of organoclay into PLA markedly enhances ductility, with Lai et al. reporting an elongation at break exceeding 200% at 1 phr C30B and Acik et al. observing improvements at 1–3 wt% loadings. However, in both studies, higher organoclay contents (≥3–4 wt%) led to a pronounced reduction in elongation, ultimately falling below that of neat PLA. In another study [27], the incorporation of CNC did not lead to a significant enhancement in the tensile strength of PLA, which has been attributed to the formation of CNC aggregates acting as stress concentrators. While a slight increase in modulus was observed with CNC addition, both elongation at break and energy absorption decreased, likely due to the introduction of the stiffer CNCs. Mohammadi et al. [31] reported that well-dispersed CNCs and the formation of an interconnected nanoparticle network in PLAs improved the mechanical properties of the solution-cast nanocomposites, leading to higher modulus and tensile strength compared to the neat PLAs. At 3 wt% CNC loading, the modulus increased by 24%, 15%, for semicrystalline PLA, and amorphous PLA, respectively.

B. Thermal Properties

Due to its inherently slow crystallization kinetics, PLA often fails to crystallize under rapid or even moderate cooling conditions [29]. Consequently, achieving sufficient crystallinity during processing methods that involve fast cooling remains a challenge. The nanoparticle incorporation into the PLA can influence its crystallization behavior by acting as nucleating agents, provided that a homogeneous dispersion is attained. Crystallinity enhancement is usually more pronounced at low nanoparticle loadings; however, when the concentration surpasses a certain threshold, the reduced molecular mobility of PLA can hinder chain folding and consequently restrict the growth of lamellar crystals formed at nucleation sites [9].

In the study of Palak et al [32], the cold crystallization temperature (Tcc) of neat PLA was consistently higher than that of PLA/CNC nanocomposites, confirming the nucleating role of CNCs in facilitating crystallization by reducing Tcc and enhancing crystallinity. Kang and Kim [40] reported that both the crystallinity and crystallization rate increased with CNC content, reaching a maximum at 105 °C regardless of CNC loading.

Incorporation of nanoparticles can enhance the thermal stability of PLA nanocomposites, depending on particle type, dispersion, and surface chemistry. Well-dispersed nanoparticles act as physical barriers, restrict polymer chain mobility, and can promote crystallization, all of which delay thermal degradation. However, excessive loading or poor dispersion may induce defects that compromise thermal resistance. For example, it was previously reported that incorporation of 1 wt% CNC slightly effected the degradation temperatures, whereas increasing the CNC content to 3 wt% led to lower thermal stability, indicating that CNC addition could not enhance the thermal resistance of PLA at a high CNC loading level [32]. On the other hand, a 0.5 wt% PLA–graphene nanocomposite exhibited an increase in degradation onset temperature of approximately 6 °C compared to neat PLA [41].

C. Barrier Properties

Incorporation of nanoparticles into PLA nanocomposites can markedly enhance barrier properties by creating a tortuous diffusion pathway that slows the transport of gases and vapors through the polymer matrix. Platelet- and sheet-like nanoparticles such as nanoclay, graphene derivatives, and CNCs are particularly effective, as their high aspect ratio increases the length and complexity of diffusion paths. Additionally, strong interfacial interactions between nanoparticles and polymer chains reduce free volume, while nanoparticle-induced crystallinity further restricts molecular permeability [42]. These combined effects often lead to significant reductions in water vapor and oxygen and transmission rates, although the extent of improvement depends strongly on nanoparticle type, dispersion quality, surface modification, and loading level. In the study of Fortunati et al. [43], it was reported that the barrier performance demonstrated a 34% reduction in water vapor permeability with 1 wt.% CNC and consistently improved oxygen barrier performance across all nanocomposites, emphasizing the reinforcing role of cellulose. Another study [44] investigated the barrier characteristic PLA/ graphene oxide nanosheets (GONSs) nanocomposites, and showed that the permeability coefficients of O_2 and CO_2 were reduced by approximately 45% and 68%, respectively. The enhanced gas barrier properties were mainly attributed to the intrinsic impermeability of GONSs, their complete exfoliation and homogeneous dispersion, and the strong interfacial adhesion with the PLA matrix, rather than to changes in PLA crystallinity or crystalline morphology. The study of Rhim et al. [45] indicated that the type of nanoclay had a significant effect on water vapor permeability (WVP): incorporation of organically modified clays reduced permeability, whereas unmodified clay slightly increased it due to its hydrophilicity. Among the modified clays, Cloisite 20A indicated the greatest reduction in WVP, reflecting its higher hydrophobicity and better compatibility with the PLA matrix. The improved barrier performance of PLA/clay nanocomposites was attributed to the tortuous diffusion pathway created by the well-dispersed impermeable clay layers, which increased the effective path length for water vapor transport.

IV. CONCLUSION

PLA-based nanocomposites represent a promising class of sustainable materials capable of overcoming the intrinsic drawbacks of neat PLA. The incorporation of nanoparticles whether processed via melt compounding or solution casting has been shown to substantially enhance the mechanical, thermal, and barrier properties of PLA by exploiting the high surface area and strong interfacial interactions of nanoscale fillers. Among the various nanofillers explored, CNCs stand out due to their abundance, renewability, biodegradability, and superior reinforcement capability. Nevertheless, their hydrophilic nature and tendency to agglomerate within the hydrophobic matrix remain major challenges, emphasizing the importance of surface modification strategies and optimized processing conditions to achieve homogeneous dispersion. Overall, nanoparticle incorporation provides a versatile route to tailor PLA for advanced applications in packaging, textiles, and biomedical fields, thereby supporting the transition toward sustainable, high-performance polymer systems. Future research should focus on scaling up environmentally benign processing techniques, enhancing nanoparticle–polymer compatibility, and systematically correlating dispersion mechanisms with end-use performance to fully exploit the potential of PLA nanocomposites in a circular materials economy.

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