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Advancements in polylactic acid (PLA) biocomposites: a review on reinforcement, additive manufacturing, and prospects for a sustainable future

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The environmental threat caused by non-biodegradable plastic wastes necessitates focusing on sustainable material alternatives. Polylactic acid (PLA), also known as polylactide, is a thermoplastic aliphatic polyester produced from renewable feedstocks such as corn starch, sugarcane, or other carbohydrate-rich biomass resources. Owing to its biodegradability, biocompatibility, mechanical strength, and low environmental impact, PLA has emerged as one of the most promising alternatives to conventional petroleum-derived polymers. It can be synthesized by two primary approaches: direct polycondensation of lactic acid monomers and ring-opening polymerization of cyclic lactide intermediates. However, the limitations of neat PLA including brittleness, and low thermal resistance hinder its widespread industrial application. These limitations can be addressed by infusing appropriate functional and natural fibers to PLA. The incorporation of such fibers and transforming them into complex components can be effectively achieved by a conventional additive manufacturing (AM) approach, primarily fused deposition modeling (FDM). This review paper outlines the recent research progress on PLA infused with biocomposites, with a focus on the influence of various natural fibers and nanofillers on its mechanical, thermal, and functional features. The correlation between the process parameters of FDM and the performance features of the printed parts is discussed. Furthermore, the paper explores the application, challenges, and process optimization of the hybrid composites.

Keywords: Non-biodegradable, biodegradable, polylactic acid, natural fibers, additive manufacturing

INTRODUCTION

In recent years, the primary environmental concerns are associated with waste plastic pollution. This consequence is due to the extensive use of petroleum-based polymers. Sustainable alternatives have to be developed to address this challenge [1]. Polylactic acid (PLA) was found to be one of the most sustainable biopolymers due to its biodegradability, and competitive mechanical properties [2]. PLA can be derived from renewable resources like corn starch or sugarcane, making it commercially viable. However, neat PLA suffers from several drawbacks, including inherent brittleness, low transition temperature, poor impact strength, low thermal stability, and slow crystallization rate. A widely adopted technique to improve the features of PLA is the infusion of appropriate reinforcements. Utilizing natural fibers as reinforcement agents is cost-effective; it also offers benefits including biodegradability and significant strength [3]. Furthermore, the integration of nanofillers or metal oxides can improve mechanical and thermal behavior [4]. The most commonly utilized additive manufacturing technology is the fused deposition modeling (FDM) because of its affordability, and capability to produce intricate patterns [5]. Owing to its compatibility, PLA is regarded as a prominent material in FDM. However, the mechanical characteristics of FDM-printed parts are highly anisotropic and highly dependent on process parameters [6]. The present review addresses the impacts of various reinforcements, the significance of FDM, the applications, and the challenges and prospects in this rapidly evolving sector.

Natural fiber reinforcement of polylactic acid

The focus on the development of sustainable materials has led to the incorporation of natural fibers into a biopolymer matrix such as polylactic acid (PLA). These natural reinforcements are typically classified based on their origin as blast fibers (flax and jute), leaf fibers (sisal), seed fibers (cotton), and grass fibers (bamboo). Bamboo fibers comprise a better strength-to-weight ratio. An overview of reinforcement materials in PLA biocomposites is summarized in Table 1.

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Table 1. Overview of reinforcement materials in PLA biocomposites

Reinforcement category	Primary function	Key contribution to sustainability
Natural plant fibers	Enhance stiffness, reduce cost and density	Biodegradability, renewable resource, waste valorization
Bio-derived nanofillers	Improve tensile/barrier properties, thermal stability	High performance from renewable bio-resources
Inorganic minerals	Increase stiffness, heat deflection temperature	Naturally abundant, can improve material efficiency
Synthetic/advanced fibers	Maximize strength and toughness for engineering uses	Promotes circular economy
Bio-based polymers	Enhance biodegradability or impact strength	Creates bio-based blends, compostability

Table 2. Impact of common natural fibers on PLA composite properties

Fiber type	Typical loading (% wt.)	Tensile strength	Tensile modulus	Impact strength	Key processing consideration
Flax/hemp	20-30	Significant increase	Excellent	Increase	High aspect ratio can cause clogging
Wood flour	20-40	Increase	Significant increase	Decrease	Low cost, but high viscosity and brittleness
Bamboo	15-25	Significant increase	Excellent	Increase/ decrease	Good strength-to-weight ratio; moisture sensitive
Recycled cellulose	10-20	Increase/ decrease	Increase	Decrease	Valorizes waste; properties depend on source and purity

Table 2 presents the impact of some typical natural fibers on the mechanical, thermal, and physical properties of PLA composites.

The outcomes of previous research revealed that bamboo fiber-PLA composites displayed significant tensile and flexural moduli, though a primary challenge is to achieve strong bonding between the naturally hydrophilic bamboo and the hydrophobic PLA. This challenge can be overcome by subjecting the composite to chemical treatment like alkalization [7, 8]. The blast fibers, when incorporated in PLA, enhance the stiffness and the thermal stability. The hybrid composites comprising PLA infused with jute and flax display better mechanical features than PLA reinforced with a single-fiber type [9]. The kenaf fiber, a prominent reinforcement material, when incorporated, provides substantial tensile strength and thermal resistance. Furthermore, fibers like sisal, elephant grass, and coir have the capability of improving impact resistance and rate of biodegradation. In contradiction, higher fiber content might lead to agglomeration and reduced tensile strength. The water resistance of the PLA matrix is a primary issue which restricts the reinforcement of hydrophilic natural fibers. However, this limitation can be addressed by chemical modifications including alkali, acetylation, and silane treatments, which significantly modify the fiber surface, facilitate interfacial adhesion, enable efficient stress transfer, and improve the overall mechanical properties of the composite [10].

Nanoscale functional fillers reinforcement of PLA

Though PLA is a biodegradable polymer, its functional features including; poor electrical conductivity, low thermal stability, and brittleness restrict its widespread application. The incorporation of nanoscale functional fillers helps to overcome these shortcomings without compromising its compostable nature. Typically, the nanoscale materials are derived from renewable resources; the most promising cellulosic nanomaterials are nanofibrillated cellulose (NFC) and cellulose nanocrystals (CNC). The exceptional mechanical properties including high stiffness and immense surface area make them ideal for reinforcing the PLA matrix. The PLA/NFC composites exhibit significantly improved tensile strength and stiffness. Lowering the agglomeration of the nanofibers makes them serve as stress concentrators and negatively influence toughness and impact strength. Carbon-based nanofillers like graphene (Gr) and carbon nanotubes (CNTs) are utilized in multiple applications. The tensile strength and modulus of PLA can be significantly increased by evenly distributed graphene reinforcement [11]. Furthermore, such reinforcement promotes properties including UV

shielding and electrical conductivity; makes it a viable candidate for applications in smart packaging and lightweight electronics. Metal and metal oxide nanoparticles are effective in imparting specific bioactive and protective functions. Studies have demonstrated that incorporating nanoparticles like zinc oxide (ZnO) and silver (Ag) can transform PLA films into active packaging materials. ZnO, when infused into PLA, enhances UV barrier properties and provides antibacterial activity [12]. Metal powders (Al, Cu), when employed as reinforcing materials, help to control thermal conductivity; infused milled carbon fibers increase the wear resistance. Moreover, recycled wastes such as spent ground coffee can alter composite properties and promote circular economy [13 – 15]. The key concept is that by meticulously selecting a filler, the characteristics of PLA can be precisely customized, transforming it into a useful material appropriate for specific applications.

Fused deposition modelling (FDM)

In recent years, FDM has become a viable processing technique for PLA biocomposites. The production of filament with superior quality is the primary challenge of this technique. Typically, it involves the use of a screw extruder to achieve homogeneous dispersion of the filler within the PLA matrix [16]. The flow index of the polymer undergoes a change in presence of fibers or particles, leading to nozzle clogging. The use of compatibilizer and plasticizers like polyethylene glycol (PEG) often facilitates smooth printing [17, 18]. The specific printing parameter has a significant influence on the mechanical and physical properties of parts manufactured through fused deposition modelling (FDM) [19, 20]. It is documented in previous studies that raster angle configurations of 0°/90° or ±45° and on-edge orientations result in higher tensile strength compared to other angles [21]. Furthermore, a smaller layer thickness increases the number of layers and the interlayer bonding area, which often enhances tensile strength at the cost of longer print times [22]. The infill density and pattern also play a significant role: higher infill density increases the mechanical strength, and the selection of patterns like cubic or gyroid can provide optimal balance between strength and material efficiency [23]. Furthermore, thermal parameters have significant impact on layer adhesion. The better layer adhesion relies on an optimum nozzle temperature whereas too high or too low nozzle temperatures might cause thermal degradation [24]. Therefore, optimizing these parameters is essential for mitigating the anisotropic nature of FDM particles and achieving performance suitable for functional applications. Table 3 summarizes the FDM processing parameters and their influence on quality and performance.

Table 3. FDM processing parameters and their optimization for PLA biocomposites

Processing parameter	Typical range for neat PLA	Effect of reinforcement	Recommended adjustment for composites
Nozzle temperature	190-220 °C	Fillers can increase melt viscosity	Increase by 5-15 °C to improve flow
Build plate temperature	50-70 °C	Often improves adhesion	Generally maintained or slightly increased
Print speed	40-80 mm/s	High filler content can hinder layer deposition	Reduce by 20-40% to ensure proper extrusion
Filament drying	Optional (recom-mended)	Natural fibers are highly hygroscopic	4-8 h at 50-60 °C before printing
Nozzle type	Brass	Abrasive fillers (e.g., carbon fiber, glass) cause rapid wear	Switch to hardened or stainless steel nozzle

Comparison of the mechanical property

PLA with incorporated fillers and reinforcements is compatible for a variety of applications due to strategic enhancement of properties. In common, PLA is ideal for low-stress applications; inducing reinforcements or filler materials can significantly alter its mechanical and thermal limitations. In particular, introducing micro-scale natural fibers typically increases material stiffness and thermal resistance with reduction in tensile strength and impact toughness. This is primarily attributed to concerns with adhesion between fiber and matrix. Moreover, by developing a better interfacial bond within the polymer matrix, synthetic fibers or nanoscale fillers like cellulose nanocrystals can facilitate a significant enhancement of strength, stiffness, and thermal stability, typically at lower loading levels. Integrating PLA with biodegradable polymers is a useful approach that enhances ductility and impact resistance rather than strength and rigidity, producing a harder, more flexible material. Choosing and incorporating an appropriate filler system customizes the property of the matrix to reach the customized ideal balance for its intended use. Table 4 compares the mechanical properties of PLA biocomposites developed for additive manufacturing.

Table 4. Comparison of the mechanical properties of PLA biocomposites for AM

Material formulation	Tensile strength (MPa)	Young's modulus (GPa)	Impact strength (J/m)	Heat deflection temp. (°C)
Neat PLA	50-70	3.0-3.5	20-30	55-60
PLA + 20% wood flour	45-55	4.0-5.0	15-20	60-65
PLA + 15% carbon fiber	80-100	7.0-9.0	35-45	100-110
PLA + 5% CNC	65-80	4.5-5.5	25-35	65-70
PLA/PHA blend	40-50	2.0-2.5	50-70	50-55

Applications of PLA biocomposites

Owing to the enhanced performance characteristics of PLA biocomposites, they are utilized for widespread industrial and commercial applications. PLA films reinforced with nanofillers such as ZnO reveal improved thermal barrier characteristics, making it compatible for sustainable applications [25, 26]. These materials are being used for non-structural interior parts due to their light weight and biodegradable benefits. PLA with the addition of hydroxyapatite (HAp) or Mg particles displays enhanced osteo conductivity and biodegradability, making it suitable for tissue engineering scaffolds, drug delivery and surgical systems, [27, 28]. The utilization of PLA biocomposites as a feedstock for FDM facilitates the production of functional prototypes, custom tools, and end-use parts [29, 30]. The extent of application continues to expand into construction, encompassing biodegradable products [31].

Challenges and future perspectives

Along with PLA significant advancements, there are certain limitations while utilizing biocomposites. The primary drawback is associated with achieving optimal and significant interfacial adhesion between the polymer matrix and the reinforcement, which is typically difficult with natural fibers. The recent research efforts will concentrate on developing novel, eco-friendly reinforcing agents and advanced surface modification techniques to address this limitation. Furthermore, printing of composite filaments necessitates precise parameter adjustment, a need for standardized procedures in manufacturing of filament and printing of component to ensure reproducibility and dependability [32]. PLA is biodegradable; therefore recycling PLA by chemical means saves materials and energy, [33]. The future research will be focused on using recycled PLA and waste materials, to make new composite filaments [34]. The future of FDM relies on 4D printing and multi-material printing. These enable PLA composites to be configured for modification in response to heat or moisture, forming a novel category of smart materials [35]. A promising approach is to develop hybrid composites which incorporate two or more filler materials, integrating natural fibers with nanoparticles. This approach intends to provide an integrated impact that addresses the drawbacks of distinct reinforcement systems and enables the exact modification of features for particular applications [36, 37]. Table 5 summarizes the functional applications of PLA biocomposites enabled by various additives. Table 6 outlines the current challenges associated with PLA biocomposites.

Table 5. Functional applications of PLA biocomposites enabled by additives

Target functionality	Typical additive	Mechanism	Potential application
Antimicrobial	Chitosan, silver nano-particles, zinc oxide	Disrupts microbial cell walls/ membranes	Medical implants, food packaging, hygienic devices
Electrical conductivity	Graphene, carbon nanotubes, carbon black	Forms a conductive percolation network	Static-dissipative packaging, sensors, flexible electronics
Flame retardancy	Aluminum tri-hydroxide, biobased phosphates	Endo-thermic decomposi-tion, char formation	Automotive interiors, electronics housings, building materials
Enhanced bio-degradation	Thermo-plastic starch	Increases hydrophili-city and microbial attack sites	Single-use agriculture products, compostable items

Table 6. Current challenges and future research directions in PLA biocomposites

Challenge category	Specific issue	Promising research direction
Material Performance	Poor interfacial adhesion between fiber and matrix	Development of novel bio-based coupling agents and surface treatments
Processability	Nozzle clogging and filament brittleness in FDM	Optimization of fiber aspect ratio and exploration of pellet-based 3D printing
Durability & aging	Hydrolytic degradation and UV sensitivity	Investigation of sustainable stabilizers and protective coatings
Standardization	Lack of material data and printing standards for composites	Establishing standardized test protocols and material databases for AM

CONCLUSION

This review points out that the primary limitations of neat PLA, which include brittleness and low thermal stability, can be significantly addressed by incorporating natural fibers and functional nanofillers, thereby customizing its properties for a wide range of applications. By addressing these constraints, PLA biocomposites can be able to make significant contributions to the transition from a conventional petroleum-based framework to a renewable, bio-based one. The use of polylactic acid (PLA) biocomposites is a significant and potent approach that successfully addresses the distinction between desirable engineering requirements and environmental sustainability. However, the associated challenges including interfacial adhesion necessitate advanced approaches. The introduction of biocomposites and additive manufacturing technologies, primarily FDM, facilitates the potential for creating complex, distinctive and lightweight components. Attaining significant interfacial adhesion between the water-resistant PLA matrix and the water-soluble natural fibers, maintaining optimum consistency in FDM and developing standardized procedures for reproducibility, are some of the primary obstacles. Future research should focus on developing novel, sustainable reinforcement agents, improving the features of composite for additive manufacturing, and integrating approaches using recycled PLA and recyclable waste fillers. PLA ensures the significant transition from a conventional, petroleum-centered economy to a renewable, bio-based approach. The research and development of PLA significantly contribute to sustainability in manufacturing and a more sustainable future.

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