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The influence of natural fillers on the mechanical properties of polylactide-based composites produced by fused filament fabrication 3D printing

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ABSTRACT

This review analyzes the effect of various natural fillers on the mechanical properties and process stability of polylactic acid based composites produced using Fused Filament Fabrication (FFF) 3D printing technology. The study synthesizes recent findings involving a wide range of bio-fillers, including hemp hurd, date palm fibers, spent coffee grounds, lyocell, and lignin. Key research highlights include record-high tensile strengths for 20 wt% hemp hurd composites (70 MPa) and a significant doubling of stiffness (7.1 GPa Young's modulus) at 40 wt% loadings. Furthermore, small additions of hemp (2.5 wt%) and date palm fibers (5 wt%) were found to increase tensile strength by 29.6% and 6.53%, respectively, while simultaneously offering weight reduction. The review identifies critical 3D printing challenges, such as moisture-induced porosity and nozzle clogging, and proposes a 'printability window' recommending particle sizes between one-fifth and one-half of the nozzle diameter. Chemical modifications, specifically lignin esterification and the use of coupling agents, are highlighted as essential pathways to improve interfacial adhesion and overcome the inherent brittleness of PLA. Finally, this work formulates general design guidelines for engineers to optimize filler loading and printing parameters, supporting the integration of agricultural waste into the sustainable circular bioeconomy.

Keywords: polylactide (PLA), FFF 3D printing, natural fillers, mechanical properties, biocomposites.

INTRODUCTION

The three-dimensional printing technique using FDM technology was born in the 1980s [1]. We can distinguish several 3D printing technologies, mainly: stereolithography (SLA), fused deposition modeling (FDM) (also known as fused filament fabrication (FFF)), selective laser sintering (SLS) [1, 2]. Reducing material costs and affordability have increased the availability of FFF technology [2, 3]. FFF printing involves applying molten material layer by layer [4] according to commands from g-code, i.e. a program for numerically controlled machines [2, 5].

Additive manufacturing, which includes 3D printing, uses many materials, mainly polymers, but also metals and ceramics [6, 7]. Ecological awareness among printer users and good mechanical properties make polylactide one of the most frequently chosen materials for printing [7]. Polylactic acid (PLA), during degradation, breaks down into natural components, including

water and carbon dioxide [5]. 3D printing is valued for the speed of production of prototypes and designs that can be personalized for a specific customer in low-volume production [6]. Biodegradable materials deserve special attention and are beginning to be tested for their use in 3D FFF printing [5, 6, 8], including derivatives of wood, cellulose [6] and used coffee [4]. Thanks to this, these materials fit into the concept of a circular economy [2, 4, 8]. While PLA is widely used in the packaging and disposable products industry, its widespread adoption in load-bearing engineering components is still limited by its brittleness and low thermal stability [9, 10].

Mixing composites with bio-additives involves the problem of proper homogenization of the polymer with the bio-component [8]. Additives may cause problems with poor material compatibility, which weakens the bonds between surfaces [8]. However, appropriate chemical treatment of additives before combining it with the PLA matrix improves its properties, as is the case with lignin [5]. Furthermore, adding some additives to PLA can help strengthen the composite, but also make it lighter by having a lower specific density of the composite [11].

Attempts to mechanically and thermally strengthen PLA using additives of natural origin are still a challenge [9] - a controlled fiber content and appropriate selection of process parameters are needed to properly produce the filament for 3D printing. This is important due to the appropriate filament diameter, fiber agglomeration, nozzle clogging or uneven extrusion flow. This is important for the performance of biocomposites that are reinforced with natural fibers [12].

FFF technology can be viewed as a micro-extrusion process where scaling down results in significant reductions in extrudate diameter, which complicates the direct translation of results from conventional extrusion to 3D printing. The introduction of natural fillers serves not only as a reinforcement but also as a strategy to reduce production costs and environmental impact, fitting into the circular economy paradigm [13].

This work focuses on checking the possibility of using polylactide additives as an ecological alternative to petroleum-based materials. The work includes some of the most frequently used fillers, as well as a description of the research process and the authors' conclusions. Moreover, this work summarizes the research results and checks which fillers can be successfully used and which worsen the mechanical properties of samples and 3D prints.

CLASSIFICATION OF NATURAL FILLERS

Fibrous fillers

Fibrous fillers represent the most extensively researched group of natural reinforcements for PLA-based composites, primarily due to their high aspect ratio and superior intrinsic mechanical properties compared to particulate waste [2]. This section synthesizes findings on various natural fibers, ranging from traditional bast fibers like hemp to regenerated cellulose (lyocell) and underutilized agricultural residues like date palm and tamarind.

Hemp and bast fibers

Bast fibers, including hemp (Cannabis sativa), flax, and harakeke, are among the most effective natural reinforcements for PLA due to their high cellulose content (typically 68–75%) and complex multi-walled structure. These fibers consist of a hard inner core surrounded by bast fiber bundles (lumen), providing exceptional intrinsic strength and stiffness. Their use supports the circular economy paradigm, as hemp is a carbon-neutral crop that sequesters carbon dioxide during growth and requires minimal chemical fertilizers [20, 21].

The reinforcement efficiency is highly dependent on chemical pre-treatments, specifically alkaline digestion (NaOH) followed by bleaching [9]. This process removes amorphous components such as hemicellulose and lignin, which increases surface roughness and facilitates mechanical interlocking with the PLA matrix. SEM analysis confirms that processed hemp fibers often exhibit a flat, ribbon-like morphology with a network of microcellulose pores, essential for effective stress transfer [9, 14].

Srivastava et al. [14] found that a minimal addition of 2.5 wt% hemp powder results in a 29.6% increase in tensile strength and a 21.7% improvement in Young's modulus compared to pure PLA. A significant benchmark in their study was achieving a 37% reduction in material density at 5 wt% loading, highlighting the potential for lightweight structural applications.

Beg et al. [9] explored higher loadings (20–40 wt%) and reported a record tensile strength of 70 MPa at 20 wt% loading, representing an 8% improvement over neat PLA. At 40 wt%, the

Young's modulus reached a maximum of 7.1 GPa, a two-fold increase over pure polymer. However, concentrations exceeding 20 wt% typically lead to a decline in tensile strength due to increased porosity (up to 16%), fiber agglomeration, and poor wetting by the PLA matrix.

Beyond static mechanics, bast fibers introduce advanced functional properties. Hemp-reinforced PLA exhibits superior vibration damping; tests according to ASTM E1876 show that 2.5 wt% loading provides the highest energy dissipation (loss factor of 0.0834), making it suitable for noise reduction and fatigue-resistant parts [14]. Furthermore, the recyclability of these biocomposites is a critical asset. Studies confirmed that hemp/PLA filaments can be reprocessed for up to three recycling cycles without a catastrophic reduction in mechanical performance, maintaining comparable printability before significant fiber separation occurs [9].

Technologically, incorporating these fibers requires precise optimization of printing parameters to prevent nozzle clogging [9, 12, 14]. Common adjustments include increasing the nozzle temperature to 210–220 °C and reducing printing speeds to 35–40 mm/s to ensure stable flow and adequate interlayer adhesion [9, 14]. While some comparative studies find harakeke slightly more effective than hemp at high loadings, both fibers remain the gold standard for sustainable 3D-printed biocomposites [13, 14].

Regenerated cellulose (Lyocell)

Lyocell fibers are a type of human-made regenerated cellulose produced through a sustainable closed-loop process involving the dissolution of cellulose in N-methyl morpholine-N-oxide (NMMO). Unlike natural bast fibers, Lyocell offers uniform dimensions and superior thermal stability, with rapid degradation occurring only above 316 °C, which is well above the typical processing temperatures for PLA (180–220 °C). These characteristics make Lyocell an attractive reinforcement choice for improving the mechanical integrity and toughness of 3D-printed biocomposites [2].

A comprehensive study by Gauss et al. [2] demonstrated that while the high shear forces during twin-screw extrusion significantly reduce the initial fiber length (from 2.93 mm to approximately 0.159 mm), the resulting aspect ratio remains above 10, which is near the critical limit for effective reinforcement. Their findings identified 10 wt% as the optimal filler loading for 3D-printed specimens, yielding an average tensile strength of 64.2 MPa and a significant 40% increase in Young's modulus (to 4.56 GPa) compared to neat PLA. Notably, the 10% formulation also exhibited an increased strain at break (4.93%), suggesting that Lyocell can mitigate the inherent brittleness of PLA more effectively than natural fibers like hemp or kenaf.

The reinforcement efficiency in Lyocell/PLA composites is closely linked to the fiber alignment induced during the FFF process. XRD and microscopic analyses confirmed that shear stresses within the nozzle facilitate the orientation of fibers parallel to the printing direction. This alignment allows for controlled directional stiffening, which is a major advantage of additive manufacturing. However, performance limits were observed at higher concentrations; at 30 wt% loading, the mechanical properties declined due to increased filament porosity (up to 7.88%) and higher stress concentrations, which initiated premature brittle failure.

Technologically, the smooth surface of Lyocell fibers proved beneficial for processability, allowing the 10% composite to be printed with nozzles as small as 0.25 mm without clogging. Furthermore, Lyocell-reinforced PLA exhibits outstanding dimensional stability during post-printing heat treatment. While pure PLA often suffers from significant anisotropic shrinkage when heated to increase crystallinity, Lyocell/PLA composites maintain their shape while achieving a storage modulus at 80 °C that is up to 72 times higher than that of neat PLA. This enables the use of these materials in load-bearing structures with complex geometries for biomedical or engineering applications.

Regional and other bast fibers

The exploration of regional natural fibers allows for the development of sustainable materials tailored to local bio-resources and specific industrial needs. Henequen fibers (Agave fourcroydes), which are endemic to the Yucatan peninsula, have been investigated by Agaliotis et al. [10] as a potential reinforcement for PLA-based composites. In their study, henequen flour with particle sizes ranging from 90 to 250 μm was incorporated in low concentrations (1–5 wt%). The results demonstrated that a 1 wt% loading was optimal for 3D-printed specimens,

achieving a maximum tensile stress of 60.1 MPa (a 5.3% increase over printed neat PLA) and a significantly improved strain to failure of 0.078 (a 53% increase). The presence of these fibers at low concentrations was found to delay crack growth, thereby enhancing both strength and ductility.

However, the effectiveness of henequen reinforcement is highly sensitive to the filler concentration and printing parameters. For flour contents of 2 wt% and higher, the maximum stress declined due to particle agglomeration and increased porosity. SEM analysis revealed the formation of triangle-shaped voids between printed layers and beads, which acted as stress concentrators. To mitigate these issues, the addition of 3 wt% maleic anhydride as a coupling agent was tested, which successfully boosted the maximum stress to 63.9 MPa by improving the interfacial adhesion between the hydrophilic fibers and the hydrophobic PLA matrix. Furthermore, the study confirmed that a 0° raster angle is essential for maximizing mechanical performance, as ±45° and 90° orientations lead to significant reductions in stiffness and strength.

Other regional agricultural residues, such as tamarindus fruit fiber (TFF), also show promise as reinforcements due to their high cellulose content (approximately 67%) and stiffness. Research by Rajaram et al. [15] indicated that TFF can significantly enhance PLA's mechanical properties, with a 40 wt% loading doubling the tensile strength (reaching 54.45 MPa) compared to neat PLA. This substantial improvement was attributed to matrix penetration into the fiber cell walls, which created a strong entangled coherence and effective stress transfer. While these specific TFF results were obtained through compression molding, they establish a high-performance baseline for this filler, suggesting that with optimized particle size and extrusion parameters, TFF could be a viable candidate for high-loading FFF filaments.

Comparing these regional reinforcements highlights a common challenge in additive manufacturing: while traditional processing can accommodate high fiber loadings (up to 40 wt%) for maximum reinforcement, FFF technology often requires lower concentrations (approx. 1–5 wt%) to maintain printability and avoid catastrophic increases in porosity [10, 15].

Particulate fillers

Industrial and agricultural waste

The utilization of industrial and agricultural by-products as fillers in PLA composites is a key strategy for reducing material costs and environmental impact. Among these, spent coffee grounds (SCG) have gained significant attention. Yu W et al. [1] demonstrated that a low concentration of 3 wt.% SCG can slightly improve the tensile strength of PLA from 49.99 MPa to 51.28 MPa, while achieving a maximum modulus of rupture of 109.07 MPa. However, their findings also highlight a critical performance threshold; exceeding 5 wt.% loading leads to a rapid decline in mechanical properties (e.g., strength dropping to 26.45 MPa at 7 wt.%) due to particle aggregation and poor dispersion, which create stress concentration sites and promote crack formation.

While desktop-scale FFF often struggles with high SCG loadings, large-format additive manufacturing (LFAM) offers a different perspective. Paramatti et al. [4] explored the use of coffee masterbatches in a fused granular fabrication (FGF) system equipped with a 3 mm nozzle. Their research revealed that the incorporation of SCGs effectively reduces the complex viscosity of the PLA melt, which facilitates material flow and increases diffusion at the interlayer interface. This rheological advantage resulted in improved layer adhesion and allowed for the successful fabrication of complex, large-scale furniture parts with consistent quality.

Another promising particulate filler is derived from cashew nutshell waste. Paternina Reyes et al. [16] incorporated ground cashew nutshells (0.5–2.0 wt.%) into PLA filaments and reported a notable simultaneous improvement in both strength and ductility. Their results showed that a 2.0 wt.% loading achieved a tensile strength of 64.20 MPa and increased the elongation at break by approximately 3.36 times compared to neat PLA. This unique toughening effect, which contrasts with the brittleness often induced by other particulate fillers, is attributed to the rough and porous surface of the cashew nutshell particles. These morphological features promote strong mechanical interlocking at the filler-matrix interface. Furthermore, the thermal stability of these particles (up to 320 °C) ensures that their

reinforcing characteristics remain intact during high-temperature extrusion and printing.

The comparative analysis of these industrial wastes suggests that porosity and surface functionalization are more critical than the chemical origin of the filler. Porous particles, such as those from cashew nutshells or optimized SCG masterbatches, provide better mechanical interlocking and rheological stability, whereas raw, non-porous particulate waste tends to act as a defect that initiates premature failure at higher concentrations [16].

Wood fillers

Wood fillers, typically in the form of sawdust or wood flour, are among the most accessible natural fillers due to their abundance as by-products of the woodworking industry [11]. Their integration into PLA matrices is a well-established strategy to create wood-like aesthetics and reduce material costs, yet it poses significant challenges regarding particle size control and interfacial adhesion [6, 11].

A recent study by Jasiński et al. [11] systematically explored the impact of pine sawdust particle size (ranging from <0.2 mm to 1 mm) on the properties of composites with a 10 wt% filler loading. Their results indicated that at the 3D printing scale, there is no clear correlation between particle size and tensile strength, with average values ranging from 9.21 to 14.28 MPa. However, particle size proved to be the most critical factor for printability. The study established a "printability window," recommending particles between one-fifth and one-half of the nozzle diameter (0.3–0.75 mm for a 1.5 mm nozzle) to prevent nozzle clogging. Interestingly, the smallest fraction (<0.2 mm) also caused partial clogging, which was attributed to the high specific surface area of fine particles leading to the formation of large agglomerates.

The mechanical performance of wood-filled PLA is highly sensitive to the filler concentration. Kariz et al. (2018) demonstrated that while a 10% beech wood content can slightly improve tensile strength, any further increase (up to 50%) leads to a sharp decline in strength (dropping to 30 MPa) and a significant increase in surface roughness and voids. This trend is supported by Liu et al. (2019), who identified 15% as the optimal loading for pine wood flour. Furthermore, Baghia et al. (2020) reported that although 20% wood flour from poplar trees can increase the Young's modulus, it simultaneously reduces both tensile strength and strain at break, reinforcing the inherent brittleness of these biocomposites.

The primary failure mechanism in PLA-wood composites is the formation of extensive voids at the filler-matrix interface. SEM characterization reveals that these voids (ranging from 9.4 to 40 μm) originate from the release of water and air trapped within the capillary spaces of the porous wood particles during high-temperature processing. As noted by Sztorch et al. (2022), the lack of chemical affinity between hydrophilic wood and hydrophobic PLA prevents effective stress transfer, often resulting in fiber pull-out and interlayer delamination. To mitigate these issues, authors suggest that wood particles with more rounded shapes and smoother surfaces, combined with precise de-airing before extrusion, are essential for achieving denser and more reliable 3D-printed parts.

Nano-fillers and bionanocomposites

Nano-fillers, particularly nanocellulose in the form of nanofibrils (CNFs) or nanocrystals (CNCs), represent a modern frontier in developing high-performance PLA bionanocomposites for FFF 3D printing. These materials offer exceptionally high aspect ratios and significant reinforcing potential at very low concentrations (typically <5 wt.%) compared to micro-scale agricultural waste [6, 17].

Recent studies by Agbakoba et al. [17] demonstrated that incorporating just 1 wt.% of cellulose nanofibrils (CNFs) extracted from eucalyptus sawdust, paired with triacetin as a green plasticizer, increased the tensile strength of 3D-printed PLA specimens by 12%. A critical finding was that CNFs act as processing aids, improving filament roundness and reducing thinning during extrusion by increasing melt viscosity. However, increasing the loading to 3 wt.% resulted in properties comparable to neat PLA, highlighting a performance plateau caused by the fundamental hydrophilicity of CNFs, which leads to agglomeration and weak interfacial interactions in the hydrophobic PLA matrix.

The use of cellulose nanocrystals (CNCs) offers even more pronounced thermal and mechanical benefits. Chike Agbakoba et al. [8] reported that CNCs act as powerful nucleating agents, enhancing the crystallization behavior of PLA by up to 15% and improving thermal

stability by approximately 20 °C. Their research achieved a substantial 48% increase in the tensile strength of bionanocomposite filaments through the synergistic use of CNCs and a multifunctional chain extender (Joncryl), which improved melt strength and prevented thermo-hydrolytic degradation during processing. Crucially, these CNC-reinforced materials exhibited a transition from inherent brittleness to ductile fracture behavior, with elongation at break reaching up to 25.5% for some formulations.

Beyond mechanical reinforcement, nanocellulose enables the development of multifunctional filaments. Szymańska-Chargot et al. [18] developed a ternary system using carrot-derived CNFs modified with silver nanoparticles (AgNPs). Their research demonstrated that adding 1–4 wt.% of this modified filler imparted strong antibacterial properties against Escherichia coli and Bacillus cereus. Importantly, the silver nanoparticles remained bonded to the nanocellulose surface, preventing their migration from the composite – a vital safety feature for active food packaging. Furthermore, the addition of carrot-derived CNFs was found to tailor the material's barrier properties by increasing gas permeability, which can be beneficial for the transpiration requirements of fresh produce packaging.

The synthesis of these findings suggests that bionanocomposites overcome the processability hurdles typical of high-loading fibrous fillers. By acting at the molecular level, nanocellulose can simultaneously enhance the mechanical integrity, thermal resistance, and specialized functional characteristics of PLA without compromising its biodegradability or the precision of the 3D printing process [8, 17, 18].

Lignin and its derivatives

Lignin, the second most abundant renewable bio-resource after cellulose, is a highly branched phenolic polymer predominantly considered an industrial byproduct of the pulping industry. Its intrinsic hydrophobicity, antioxidant capacity, and reinforcing potential make it a versatile natural filler for PLA-based composites [3, 19]. However, the incorporation of unmodified (native) lignin often faces challenges related to its high thermal transition temperature and resistance to flow, which can limit printability and induce brittleness.

Research by Zaidi et al. [3] evaluated the stability of filaments produced using a bench-top extruder with organosolv lignin (3–15 wt.%). Their findings indicate that low concentrations (3–5 wt.%) of native lignin can enhance the Young's modulus of 3D-printed specimens by up to 34% through improved interlayer adhesion. However, concentrations reaching 10 wt.% resulted in a significant decline in tensile strength (dropping to 23.38 MPa from 49.25 MPa for neat PLA). This instability at higher loadings is consistently attributed to particle agglomeration and insufficient interfacial bonding between the hydrophilic hydroxyl groups of lignin and the hydrophobic PLA matrix. Similarly, Tanase-Opedal et al. [19] observed that while spruce lignin generally reduces tensile strength, these effects can be partially mitigated by optimizing the 3D printing temperature. For their biocomposites (20–40 wt.% lignin), a temperature of 215 °C proved optimal for enhancing layer fusion and mechanical performance. Furthermore, their study confirmed that lignin acts as an effective nucleating agent, facilitating the crystallization of the PLA matrix.

To overcome the compatibility issues of native lignin, chemical modifications such as esterification have been explored. Mohan et al. [20] successfully modified organosolv pine lignin with fatty acid esters (C10, C14, and C18). The C14 lignin ester at a 30 wt.% loading emerged as the superior additive, significantly improving strength, formability, and overall structural integrity compared to native lignin. Specifically, samples printed at 220 °C exhibited the highest quality with uniform surface texture and superior layer cohesion. This improvement is attributed to the optimal chain length of the C14 ester, which enhances chain mobility and flexibility without causing phase separation.

Beyond static mechanical reinforcement, lignin imparts valuable multifunctional properties to 3D-printed parts. Ye et al. [5] demonstrated that even extremely low loadings (0.5 wt.%) of lignin can improve the toughness of PLA by 81.8% and tensile strength by 13.5%. Additionally, these lignin-doped materials exhibited remarkable UV-blocking capability (up to 99.9% for UVB) and antioxidant activity (up to 79%), highlighting their potential for sustainable packaging and medical applications.

The synthesis of these findings suggests that while native lignin is effective at low concentrations (up to 5 wt.%), achieving high-performance biocomposites at higher loadings requires

chemical derivatization, such as esterification, to ensure adequate compatibility and structural integrity in FFF-printed components.

PROCESSING AND PRINTABILITY CHALLENGES

Nozzle clogging and particle size control

Nozzle clogging is identified as a primary processing barrier in the FFF printing of PLA biocomposites, as the physical obstruction of the extrusion path directly compromises part quality and process stability [6]. This phenomenon is highly sensitive to the filler's granulometric fraction relative to the nozzle orifice. Jasiński et al. [11] systematically quantified this relationship using pine sawdust, establishing that particles exceeding one-half of the nozzle diameter (e.g., >0.75 mm for a 1.5 mm nozzle) lead to immediate and complete clogging. Interestingly, their study also revealed that the smallest tested fraction (<0.2 mm) could cause partial clogging, a phenomenon attributed to the high specific surface area of fine particles which promotes the formation of large agglomerates. Based on these findings, they proposed a "printability window" recommending particle sizes between one-fifth and one-half of the nozzle diameter to ensure stable flow while maintaining material integrity.

The risk of blockage is further amplified by the filler concentration. Research by Mian et al. [12] and Srivastava et al. [14] consistently identifies an optimal loading range between 2 wt.% and 6 wt.% for various natural fibers, such as date palm and hemp. Exceeding these thresholds often results in uneven extrusion and increased brittleness due to fiber clustering and poor wetting by the PLA matrix. Similarly, Agaliotis et al. [10] observed that henequen flour concentrations above 2 wt.% led to particle agglomeration and the formation of triangle-shaped voids between layers, which further destabilized the printing process.

However, filler morphology also plays a significant role in determining the minimum viable nozzle size. For instance, the smooth surface and uniform dimensions of regenerated cellulose (lyocell) allowed for successful printing with nozzles as small as 0.25 mm at 10 wt.% loading [2]. In contrast, natural micro-scale agricultural wastes typically require larger nozzle diameters to prevent the entrapment of non-uniform particles [2, 4]. Ultimately, as noted by Sharma et al. [6], improper particle size control remains one of the primary reasons for low 3D printing quality in biomass-filled polymers, necessitating rigorous pre-processing steps such as sieving and double extrusion to ensure adequate homogenization.

Rheological behavior and extrusion stability

The introduction of natural fillers fundamentally alters the rheological properties of the PLA melt, which is a critical determinant of extrusion stability and filament quality. FFF technology can be perceived as a micro-extrusion process where scaling down industrial processes results in a significant reduction of the extrudate diameter – often by a factor of 20 to 30. This miniaturization amplifies the impact of melt viscosity variations and the sensitivity of the process to filler-induced inhomogeneities [13].

The impact on viscosity varies significantly depending on the filler type and its morphology. For instance, incorporating spent coffee grounds (SCG) has been shown to reduce the complex viscosity of the PLA melt. This reduction facilitates the melt flow of the extrudate and increases molecular diffusion at the interface between layers, which leads to better interlayer adhesion and reduced internal pores [4]. In contrast, nanocellulose reinforcements generally increase melt viscosity. Studies on cellulose nanocrystals (CNCs) indicate that restricted mobility of PLA chains results in higher viscosity, with composites often exhibiting Newtonian behavior at low shear rates before transitioning to shear-thinning at higher frequencies. While increased viscosity can prevent filament "drooping" and improve the roundness of the extruded material[8], it also heightens the risk of under-extrusion if processing temperatures are not precisely optimized to ensure adequate flow [12].

Extrusion stability is further challenged by inconsistencies in filament diameter and ovality, which directly affect the volume of material deposited. Research on bionanocomposites suggests that diameter variations are often linked to the presence of plasticizers and the tension applied during winding [8, 17]. For instance, triacetin plasticization can lead to "thinning" of the filament as it emerges from the nozzle due to reduced melt strength [17]. However, the addition of cellulose nanofibrils (CNFs) can mitigate this

effect by increasing melt viscosity and enhancing compression during processing, resulting in improved filament roundness and more consistent diameters [8, 17].

A major factor destabilizing the extrusion process is the inherent hydrophilicity of natural fillers. Lignocellulosic particles, such as wood sawdust or hemp fibers, tend to release trapped air and residual moisture during high-temperature extrusion, leading to moisture-induced porosity [2, 12]. These micro-voids not only destabilize the extrudate diameter but also act as stress concentrators in the printed parts, often initiating failure between beads [12]. Furthermore, the lack of chemical affinity between hydrophilic fillers and the hydrophobic PLA matrix often causes phase separation, which manifests as structural discontinuities and irregular flow behavior during the deposition of the molten bead [11].

Parameter optimization and innovative solutions

To overcome the inherent processing difficulties of biocomposites, an iterative optimization of 3D printing parameters is essential. Literature indicates that standard PLA settings (typically 190–210 °C) are often insufficient for filled materials due to incomplete melting and increased melt viscosity. Mian et al. [12] reported that for PLA reinforced with 5 wt% date palm fiber, increasing the nozzle temperature to 220 °C and reducing the printing speed to 63 mm/s were necessary to achieve stable extrusion, as lower temperatures resulted in under-extrusion and clogging. Similarly, Srivastava et al. [14] found that as hemp content increased from 2.5% to 5%, progressive adjustments were required, reaching a nozzle temperature of 220 °C and a reduced speed of 35 mm/s. These adjustments increase the residence time of the material in the heated nozzle, ensuring more uniform melting and reducing the risk of blockage.

Build plate management also requires recalibration to handle the altered thermal properties of biocomposites. Mian et al. [12] successfully mitigated warping and inadequate first-layer adhesion by increasing the platform temperature from 55 °C to 65 °C. For lignin-based composites, Tanase-Opedal et al. [19] identified 215 °C as the optimal printing temperature to enhance layer fusion. In more advanced systems using modified lignin esters, the highest print quality and strongest interlayer cohesion were achieved at 220 °C, highlighting the synergy between chemical modification and thermal optimization [20].

Beyond software adjustments, hardware innovations provide promising solutions to biomass-related printability issues. Sharma et al. [6] proposed a novel nozzle design featuring selective variable extrusion and multiple interchangeable dies. This mechanism can accommodate various filler shapes and granularities, significantly reducing the risk of blockages while improving production speed and surface quality. Furthermore, the application of large-format additive manufacturing (LFAM) with direct pellet extrusion (Fused Granular Fabrication, FGF) represents a major scaling breakthrough. Paramatti et al. [4] demonstrated that using a 3 mm nozzle and a single-screw pellet extruder bypassed the clogging problems typical of desktop FFF, allowed the use of coarser particles (up to 900 μm), and minimized thermomechanical degradation by eliminating the intermediate filament extrusion step.

The synthesis of these findings suggests that while desktop FFF requires narrow "printability windows" and precise tuning of thermal parameters, emerging hardware technologies like FGF and specialized nozzles offer more robust pathways for the industrial implementation of natural filler-reinforced PLA.

PLA SPECIFIC PARAMETERS

Natural fillers concentrations in the composite range from 1% to 40% (in the case of lignin ester and hemp hurd). The 3D printing temperature of the samples ranged from 150°C (Jasiński et al.) to 230 °C (Tanase-Opedal et al.). Due to the additives in the composite, printing speeds are low, ranging from 10 to 63 mm/s. According to available studies, the bed temperature does not affect the mechanical strength of the samples, but rather the adhesion of the samples to the printing bed. This prevents the samples from detaching from the bed. The numerical data for Young's modulus and tensile strength are summarized in Table 3 and Table 4, respectively. These tables include only the specific values explicitly stated in the source texts by the original authors. In cases where research findings were presented solely through graphical representations without precise numerical data, those values were omitted

to ensure the highest degree of accuracy and to avoid potential errors associated with manual estimation from charts.

The Young's modulus values reported by Beg et al. are highest at a 40% additive concentration, but no values are provided for other concentrations. The Mian et al. study provides information on Young's modulus but does not calculate its value.

Tensile strength was reported by the authors for all concentrations in most cases. In the work of Jasiński et al., maximum strength values were given depending on the length of the sawdust used in the study. Again, in the work of Beg et al., only the highest strength value obtained overall (the highest was for the 20% concentration).

Reinforcement efficiency – fibrous vs. particulate fillers

A comparison of the data indicates that fibrous fillers consistently provide superior reinforcement compared to particulate waste, particularly regarding ultimate tensile strength (UTS). The primary reason for this divergence is the high aspect ratio of fibers, which facilitates more effective stress transfer from the PLA matrix to the reinforcement [2, 9].

High-performance reinforcements such as hemp hurd fibers (20 wt.%) have achieved a record tensile strength of 70 MPa [9]. Similarly, regenerated cellulose (Lyocell) at 10 wt.% loading yielded

Table 1. Bio-additives used, PLA type and natural filler concentration values in the composite

References	Filler	PLA type	Filler concentration in the composite
Yu W et al. [1]	Coffee	PLA Ingeo 4043D	0%, 1%, 3%, 5%, 7%
Paramatti et al. [4]	Coffee	PLA Ingeo 3D850	0%, 5%, 10%
Gauss et al.[2]	Lyocell	PLA Ingeo 2003D	0%, 10%, 20%, 30%
Zaidi et al. [3]	Organosolv lignin	PLA Ingeo 3052	0%, 3%, 5%, 10%
Mohan et al. [20]	C14 lignin ester	PLA Ingeo 3D850	0%, 10%, 20%, 30%, 40%
Jasiński W et al. [11]	Wood (sawdust)	PLA Ingeo 4043D	0%, 10%
Beg et al. [9]	Hemp fibers (hemp hurd)	PLA Ingeo 2003D	0%, 20%, 30%, 40%
Mian SH et al. [12]	Date palm	PLA (not specified)	0%, 5%
Srivastava et al. [14]	Ground and sieved hemp powder	PLA (not specified)	0%, 2.5%, 5%
Agaliotis et al. [10]	Henequen fiber flour	PLA Ingeo 2003D	0%, 1%, 2%, 3%, 4%, 5%
Agbakoba et al. [17]	Cellulose nanofibrils (CNFs)	PLA Ingeo 6202D	0%, 1%, 3%
Chike Agbakoba et al. [8]	Cellulose nanocrystals (CNCs)	PLA Ingeo 6202D	0%, 0.1%, 0.5%, 1%
Tanase-Opedal et al. [19]	Spruce lignin	PLA (not specified)	0%, 20%, 40%

Table 2. 3D printing process parameters used in individual studies

References	Nozzle diameter [mm]	Printing temperature [°C]	Bed temperature [°C]	Print speed [mm/s]
Yu W et al. [1]	n/a	195	-	60
Paramatti et al.[4]	3.0	190	80	10, 15, 20, 30
Gauss et al. [2]	0.5	215	70	30
Zaidi et al. [3]	0.4	215	60	60
Mohan et al. [20]	0.4	220	70	15
Jasiński W et al. [11]	1.5	150	-	-
Beg et al. [9]	0.75	210	70	30
Mian SH et al. [12]	0.4	210	65	63
Srivastava et al. [14]	0.4	200–220	60–70	35–45
Agaliotis et al. [10]	0.8	215	60	30
Agbakoba et al. [17]	n/a	175	n/a	60
Chike Agbakoba et al. [8]	0.4	185–190	50	60
Tanase-Opedal et al. [19]	0.4	205–230	n/a	35

Table 3. Young's modulus value for different natural filler concentration values [MPa]

References	Material used in the biocomposite	0%	1%	2%	2.5%	3%	4%	5%	10%	20%	30%	40%
Yu W et al. [1]	Coffee (grounds)	2336	-	-	-	3604	-	2044	-	-	-	-
Paramatti et al. [4]	Coffee masterbatch	3192.6	-	-	-	-	-	2983.7	2880.7	-	-	-
Gauss et al. [2]	Lyocell fibers	3260	-	-	-	-	-	-	4560	4580	4900	-
Zaidi et al. [3]	Lignin	488	-	-	-	654	-	548	524	-	-	-
Mohan et al. [20]	C14 lignin ester	-	-	-	-	-	-	-	-	-	786.14	-
Jasiński W et al. [11]	Wood (sawdust)	1580	-	-	-	-	-	-	1014	-	-	-
Beg et al. [9]	Hemp fibers (hemp hurd)	-	-	-	-	-	-	-	-	-	-	7100
Srivastava et al. [14]	Hemp powder	3200	-	-	3760	-	-	4090	-	-	-	-
Agaliotis et al. [10]	Henequen flour	1930	1480	1620	-	1430	1580	1490	-	-	-	-
Agbakoba et al. [17]	CNFs + plasticizer	2115.5	2378.75	-	-	2542.33	-	-	-	-	-	-
Tanase-Opedal et al. [19]	Spruce lignin	2890	-	-	-	-	-	-	-	2460	-	2695

Table 4. Tensile strength values depending on the natural filler concentration [MPa]

References	Material used in the biocomposite	0%	1%	2%	2.5%	3%	4%	5%	7%	10%	20%	30%	40%
Yu W et al. [1]	Coffee (grounds)	49.99	-	-	-	51.28	-	-	26.45	-	-	-	-
Paramatti et al. [4]	Coffee (grounds)	-	-	-	-	-	-	49	-	47	-	-	-
Gauss et al. [2]	Lyocell	62.8	-	-	-	-	-	-	-	64.2	57.1	47.5	-
Zaidi et al. [3]	Lignin (printed samples)	49.25	-	-	-	31.48	-	30.23	-	23.38	-	-	-
Mohan et al. [20]	C14 lignin ester	-	-	-	-	-	-	-	-	-	-	19.2	-
Jasiński W et al. [11]	Wood (sawdust)	33.43	-	-	-	-	-	-	-	14.28	-	-	-
Beg et al. [9]	Hemp fibers (hemp hurd)	-	-	-	-	-	-	-	-	-	70	-	-
Mian SH et al. [12]	Date fibers	50.4	-	-	-	-	-	53.69	-	-	-	-	-
Agaliotis et al. [10]	Henequen flour	56.7	60.1	48.9	-	41.4	40.4	35.5	-	-	-	-	-
Agbakoba et al. [17]	CNFs + plasticizer	22.7	25.43	-	-	22.41	-	-	-	-	-	-	-
Tanase-Opedal et al. [19]	Spruce lignin	58.45	-	-	-	-	-	-	-	-	39.35	-	45.65

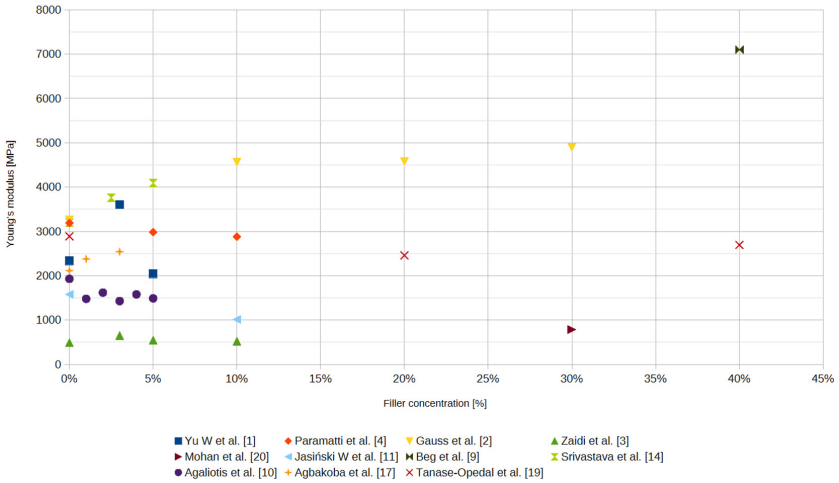

Figure 1. Young's modulus value for different natural filler concentration values [MPa]

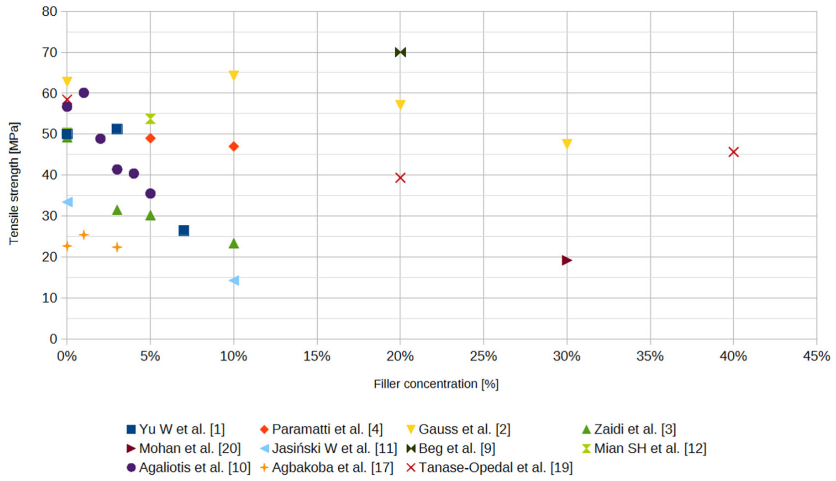

Figure 2. Tensile strength values depending on the natural filler concentration [MPa]

a UTS of 64.2 MPa [2]. Even at low concentrations, fibers show significant benefits; Srivastava et al. [14] reported a 29.6% increase in tensile strength with just 2.5 wt.% hemp powder, while Mian et al. observed a 6.53% improvement (reaching 53.69 MPa) using 5 wt.% date palm fibers [12].

In contrast, particulate fillers such as spent coffee grounds (SCG) and wood sawdust typically lead to a degradation of UTS as concentrations increase. Yu W et al. [1] found that while 3 wt.% SCG could slightly enhance strength to 51.28 MPa, exceeding 5 wt.% caused a rapid drop to 26.45 MPa at 7 wt.% loading. Similarly, Jasiński et al. [11] reported that 10 wt.% pine sawdust resulted in relatively low UTS values ranging from 9.21 to 14.28 MPa, with no clear correlation to particle size at the 3D printing scale.

The efficiency of fibers is further enhanced by the alignment induced during extrusion. XRD and microscopic characterization of Lyocell and hemp composites confirm that fibers orient themselves parallel to the printing direction (0° raster), allowing for controlled directional stiffening [2]. Conversely, isometric particles (e.g., coffee grounds, wood dust, native lignin) often act as stress concentrators or inclusions that disrupt matrix continuity [1, 3]. Without high aspect ratios to bridge cracks, these particulates initiate premature failure at the filler-matrix interface [3].

The synthesis of these results suggests that for applications requiring high structural integrity, fibrous fillers are the optimal choice, whereas particulate fillers are better suited for cost reduction or aesthetic purposes at very low loadings (typically <5 wt.%) to avoid catastrophic loss of strength.

The stiffness-strength trade-off

A universal trend observed across nearly all studied natural fillers is the marked increase in stiffness (Young's modulus) accompanied by a progressive loss of tensile strength and ductility (strain at break) as filler loading increases [21]. This "stiffness-strength trade-off" is primarily governed by the restricted mobility of PLA polymer chains induced by the inclusion of rigid bio-derived particles or fibers [3, 21].

The addition of rigid fillers acts as a reinforcement that enhances the modulus of elasticity according to the rule of mixtures [21]. For instance, adding native organosolv lignin at low concentrations (3–5 wt.%) can enhance the Young's modulus by up to 34% [3]. In high-loading fibrous composites, Beg et al. [9] reported that a 40 wt.% hemp hurd content achieved a record Young's modulus of 7.1 GPa, representing a two-fold increase over neat PLA. Similar stiffening effects were noted for cellulose nanocrystals (CNCs), where restricted chain mobility resulted in higher storage moduli and improved thermal stability [8, 17].

While stiffness continues to rise with filler content, UTS often reaches a peak at low concentrations before declining sharply [1]. Native lignin at 10 wt.% loading causes the UTS to drop from 49.25 MPa to just 23.38 MPa [3]. Similarly, wood-filled PLA exhibits a 10% strength increase at 10 wt.% beech wood loading, but drops to 30 MPa when the content is increased to 50 wt.% [10, 13].

The degradation of strength at higher loadings is consistently attributed to three factors: particle agglomeration, increased porosity, and poor interfacial wetting [1, 9, 10]. As the filler

content increases, the PLA matrix can no longer fully encapsulate every particle or fiber, leading to the formation of extensive internal voids – reported as high as 16% for some hemp and wood composites [9]. These voids and clusters act as stress concentrators that initiate premature brittle failure, preventing the effective transfer of loads from the matrix to the reinforcement [3, 10, 21].

The synthesis of these findings highlights a critical "performance plateau" for most biocomposites; while natural fillers are highly effective at increasing the rigidity of PLA, their use in structural applications is limited to a narrow concentration window (typically 3–10 wt.%) unless chemical modifications are employed to preserve material integrity [14, 20].

Role of interfacial adhesion and chemical modification

The fundamental challenge in developing high-performance PLA biocomposites is the inherent chemical incompatibility between the hydrophilic lignocellulosic fillers and the hydrophobic polymer matrix. This mismatch leads to poor interfacial adhesion, resulting in the formation of micro-voids at the boundary layers that prevent effective stress transfer and act as crack initiation sites [9, 10]. Synthesis of recent findings indicates that while physical interactions can provide modest reinforcement at very low concentrations, chemical modification is the critical pathway for breaking performance barriers at higher loadings.

Zaidi et al. [3] reported that incorporating 10 wt.% of native organosolv lignin caused a drastic reduction in tensile strength, dropping from 49.25 MPa for neat PLA to just 23.38 MPa. This instability is attributed to insufficient interfacial bonding and the tendency of unmodified hydroxyl groups to promote particle agglomeration.

In contrast, Mohan et al. [20] demonstrated that esterifying organosolv lignin with tetradecanoic acid (C14) allows for a successful loading of up to 30 wt.%, achieving the highest strength and toughness in the study. The long-chain aliphatic groups of the C14 ester enhance compatibility and wetting by the PLA matrix, which SEM analysis confirms leads to superior layer cohesion and a uniform surface texture.

The utilization of coupling agents offers another effective strategy for bridging the filler-matrix interface. Agaliotis et al. [10] found that adding 3 wt.% maleic anhydride to a 1 wt.% henequen-reinforced composite successfully boosted the maximum tensile stress from 60.1 MPa to 63.9 MPa. This improvement is fostered by the coupling agent's ability to create a chemical bridge between the filler and matrix, thereby delaying crack growth and enhancing ductility.

Furthermore, pre-treatments such as alkaline digestion (NaOH) followed by bleaching or silanization play a dual role: they remove amorphous components like hemicellulose and lignin while simultaneously increasing surface roughness [9]. This modified morphology facilitates mechanical interlocking with the PLA matrix, a factor identified as essential for the effective stress transfer observed in high-strength Lyocell and treated hemp composites [2, 13]. Collectively, these results suggest that chemical functionalization not only preserves material integrity at high filler contents but also allows for the customization of functional properties like thermal stability and UV-blocking without compromising mechanical performance [5, 20].

AM-specific failure mechanisms

A critical analysis of the literature reveals that the mechanical performance of PLA biocomposites is often dominated by structural defects unique to the Fused Filament Fabrication (FFF) process, rather than the intrinsic properties of the material constituents alone. The layer-by-layer nature of additive manufacturing introduces specific failure modes, primarily centered around anisotropic porosity and interfacial discontinuities.

One of the most prevalent defects in 3D-printed biocomposites is the formation of triangle-shaped voids at the junctions between adjacent beads and layers. These voids, which typically result in a baseline porosity of 4–5% even in neat PLA, act as primary stress concentrators that initiate crack propagation [10]. In natural filler-reinforced composites, this phenomenon is often exacerbated by the filler's presence, with reported porosity levels climbing as high as 16% for hemp and wood-filled filaments [9]. The presence of these macro-pores leads to premature brittle failure, as the effective load-bearing cross-section is significantly reduced [10].

Unlike traditional injection molding, the hot-end extrusion in FFF triggers the rapid release of air and water vapor trapped within the capillary spaces of hydrophilic fillers like wood sawdust or hemp fibers. This "outgassing" during the molten state creates micro-voids and phase separation at

the filler-matrix interface. SEM characterization by Jasiński et al. [11] and others confirms that these moisture-induced discontinuities range from 9.4 to 40 µm and do not correlate with particle size, suggesting they are a fundamental byproduct of processing porous biomass at high temperatures.

The strength of 3D-printed parts is heavily dependent on the cohesion between deposited beads. Research on date palm biocomposites shows that a tighter raster packing (approx. 133 µm) allows for a more uniform stress distribution compared to neat PLA (approx. 212 µm), which often exhibits larger gaps and signs of interlayer debonding [12]. While inlayer fracture (breaking of individual rasters) remains the dominant mechanism in high-quality prints, weak interfacial bonding – often caused by filler agglomeration at layer boundaries – frequently shifts the failure mode to interlayer delamination or fiber pull-out[10, 12].

An exceptional mechanical shift is observed in nanofillers (CNC/CNF) reinforced systems. While most micro-scale natural fillers increase the brittleness of PLA, the addition of cellulose nanocrystals can induce a transition from brittle to ductile fracture behavior. This is evidenced by the "whitening effect" during tensile testing, which results from crazing (micro-void formation) and strain-induced crystallization. For instance, bionanocomposites containing 1 wt.% CNFs and triacetin achieved an elongation at break of 105%, highlighting how molecular-scale reinforcement can densify the structure and mitigate the sudden failure typical of 3D-printed parts [8, 17].

In summary, failure in 3D-printed PLA biocomposites is a multi-scale phenomenon where AM-specific geometric voids and process-induced porosity often override the reinforcing potential of the fillers, necessitating precise control over printing parameters and filler drying to achieve structural reliability.

CONCLUSIONS

The selection of filler concentration represents a fundamental trade-off between mechanical stiffening and process stability.

To achieve a balance between strength and stiffness, fibrous reinforcements such as Lyocell or treated hemp should be utilized at concentrations of 10–20 wt.%. While higher loadings (up to 40 wt.%) can double the Young's modulus, they often lead to a sharp decline in tensile strength due to porosity levels reaching 16%.

For industrial and agricultural waste (e.g., spent coffee grounds or wood flour), the optimal loading window is much narrower, typically 1–5 wt.%. Concentrations exceeding 7 wt.% usually act as structural defects, promoting crack initiation and significant loss of material continuity. Bionanocomposites utilizing cellulose nanocrystals (CNCs) or nanofibrils (CNFs) offer the most efficient reinforcement, providing significant improvements in crystallinity and ductility at extremely low loadings of 1–3 wt.%.

Engineers should increase the nozzle temperature to 210–220 °C and reduce printing speeds to 35–40 mm/s. These adjustments increase the residence time of the material in the hot-end, ensuring more uniform melting and reducing the risk of nozzle blockage. To prevent clogging, filler particle size must be strictly controlled between one-fifth and one-half of the nozzle diameter (e.g., 0.3–0.75 mm for a 1.5 mm nozzle). Raising the build plate temperature to 65–70°C is recommended to mitigate the warping issues commonly associated with the moisture-sensitive nature of bio-fillers.

Esterification of fillers (e.g., using C14 tetradecanoic acid) is highly recommended for amorphous fillers like lignin, as it improves interfacial wetting and allows for loadings of up to 30 wt.% without sacrificing toughness. The addition of approximately 3 wt.% maleic anhydride as a coupling agent can effectively bridge the filler-matrix interface, delaying crack growth and enhancing ductility.

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REFERENCES

1. Yu W, Yuan T, Yao Y, Deng Y, Wang X. PLA/coffee grounds composite for 3D printing and its properties. Forests. 2023 Feb;14(2):367. https://doi.org/10.3390/f14020367
2. Gauss C, Pickering KL, Tshuma J, McDonald-Wharry J. Production and assessment of poly(lactic acid) matrix composites reinforced with regenerated cellulose fibres for fused deposition modelling. Polymers. 2022 Jan;14(19):3991.

https://doi.org/10.3390/polym14193991

3. Zaidi SAS, Kwan CE, Mohan D, Harun S, Luthfi AAI, Sajab MS. Evaluating the stability of PLA-lignin filament produced by bench-top extruder for sustainable 3D printing. Materials. 2023 Jan;16(5):1793. https://doi.org/10.3390/ma16051793
4. Paramatti M, Romani A, Pugliese G, Levi M. PLA feedstock filled with spent coffee grounds for new product applications with large-format material extrusion additive manufacturing. ACS Omega. 2024 Feb 13;9(6):6423–31. https://doi.org/10.1021/acsomega.3c05669
5. Ye H, He Y, Li H, You T, Xu F. 3D-printed polylactic acid/lignin films with great mechanical properties and tunable functionalities towards superior UV-shielding, haze, and antioxidant properties. Polymers. 2023 Jan;15(13):2806. https://doi.org/10.3390/polym15132806
6. Sharma V, Roozbahani H, Alizadeh M, Handroos H. 3D printing of plant-derived compounds and a proposed nozzle design for the more effective 3D FDM printing. IEEE Access. 2021;9:57107–19. https://doi.org/10.1109/ACCESS.2021.3071459
7. Valvez S, Santos P, Parente JM, Silva MP, Reis PNB. 3D printed continuous carbon fiber reinforced PLA composites: A short review. Procedia Struct Integr. 2020 Jan 1;1st Virtual Conference on Structural Integrity - VCSI125:394–9. https://doi.org/ https://doi.org/10.1016/j.prostr.2020.04.056
8. Chike Agbakoba V, Hlangothi P, Andrew J, Jacob John M. Preparation of cellulose nanocrystal (CNCs) reinforced polylactic acid (PLA) bionanocomposites filaments using biobased additives for 3D printing applications. Nanoscale Adv. 2023;5(17):4447–63. https://doi.org/10.1039/D3NA00281K
9. Beg MDH, Pickering KL, Akindoyo JO, Gauss C. Recyclable hemp Hurd fibre-reinforced PLA composites for 3D printing. J Mater Res Technol. 2024 Nov 1;33:4439–47. https://doi.org/10.1016/j.jmrt.2024.10.082
10. Agaliotis EM, Ake-Concha BD, May-Pat A, Morales-Arias JP, Bernal C, Valadez-Gonzalez A, et al. Tensile behavior of 3D printed polylactic acid (PLA) based composites reinforced with natural fiber. Polymers. 2022 Jan;14(19):3976. https://doi.org/10.3390/polym14193976
11. Jasiński W, Szymanowski K, Nasiłowska B, Barlak M, Betlej I, Prokopiuk A, et al. 3D printing wood–PLA composites: The impact of wood particle size. Polymers. 2025 Jan;17(9):1165. https://doi.org/10.3390/polym17091165
12. Mian SH, bin Jumah A, Saleh M, Mohammed JA. Fabrication of PLA–date fiber biocomposite via extrusion filament maker for 3D printing and its characterization for eco-friendly and sustainable applications. Polymers. 2025 Jan;17(19):2707. https://doi.org/10.3390/polym17192707
13. Sztorch B, Brząkalski D, Pakuła D, Frydrych M, Špitalský Z, Przekop RE. Natural and synthetic polymer fillers for applications in 3D printing – FDM technology area. Solids. 2022 Sep;3(3):508–48. https://doi.org/10.3390/solids3030034
14. Srivastava P, Arumugam AB, Agrawal AP, Ntumba ZN. Development of hemp fiber reinforced PLA composites for sustainable 3D printing: Mechanical and microstructural properties. J Nat Fibers. 2025 Dec 31;22(1):2558214. https://doi.org/10.1080/15440478.2025.2558214
15. Rajaram S, Subbiah T, Arockiasamy FS, Iyyadurai J. Transforming agricultural waste into sustainable composite materials: mechanical properties of tamarindus fruit fiber (TFF)-reinforced polylactic acid composites. Eng Proc. 2024;61(1):32. https://doi.org/10.3390/engproc2024061032
16. Paternina Reyes MJ, Unfried Silgado J, Santa Marín JF, Colorado Lopera HA, Espitia Sanjuán LA. Cashew nutshells: A promising filler for 3D printing filaments. Polymers. 2023 Nov 7;15(22):4347. https://doi.org/10.3390/polym15224347
17. Agbakoba VC, Mokhena TC, Ferg EE, Hlangothi SP, John MJ. PLA bio-nanocomposites reinforced with cellulose nanofibrils (CNFs) for 3D printing applications. Cellulose. 2023 Dec 1;30(18):11537–59. https://doi.org/10.1007/s10570-023-05549-2
18. Szymańska-Chargot M, Chylińska M, Pieczywek PM, Walkiewicz A, Pertile G, Frąc M, et al. Evaluation of nanocomposite made of polylactic acid and nanocellulose from carrot pomace modified with silver nanoparticles. Polymers. 2020 Apr;12(4):812. https://doi.org/10.3390/polym12040812
19. Tanase-Opedal M, Espinosa E, Rodríguez A, Chinga-Carrasco G. Lignin: A biopolymer from forestry biomass for biocomposites and 3D printing. Materials. 2019 Jan;12(18):3006. https://doi.org/10.3390/ma12183006
20. Mohan MK, Krasnou I, Lukk T, Karpichev Y. Novel softwood lignin esters as advanced filler to PLA for 3D printing. ACS Omega. 2024 Nov 5;9(44):44559–67. https://doi.org/10.1021/acsomega.4c06680
21. Zhang L, Chai W, Li W, Semple K, Yin N, Zhang W, et al. intumescent-grafted bamboo charcoal: a natural nontoxic fire-retardant filler for polylactic acid (PLA) composites. ACS Omega. 2021 Oct 19;6(41):26990–7006. https://doi.org/10.1021/acsomega.1c03393