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Review

Valorization of Natural Byproducts Through Additive Manufacturing for Ecologically Sustainable Composite Materials: A Literature Review

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Highlights

What are the main findings?

- The integration of sawdust, wood chips, and bark into polymer matrices through AM can enhance thermal stability. Filler contents up to 30 wt.% maintain uniform dispersion while higher concentrations could result in nozzle clogging and degraded mechanical strength.
- Surface modification via coupling agents and controlled fiber morphology can significantly improve interfacial adhesion. These treatments can enhance mechanical performance by 25–30%, provide thermal stability, and improve dimensional accuracy due to improved interfacial bonding.

What is the implication of the main finding?

- Valorization of wood byproducts for the production of polymer-based composites with the aid of additive manufacturing resulted in 30–35% lower energy consumption and 65–70% reduced CO_2 emissions compared to the conventional thermoplastic systems.
- Demonstrated large-scale applications such as the BioHome3D project, confirming the feasibility of translating those findings into industrial applications. The incorporation of smart technologies can lead to efficient sustainable closed loop systems in alignment with the global environmental goals.

Abstract

This review paper explores the influence of natural byproducts on polymer matrices via additive manufacturing (AM), focusing specifically on the development of eco-friendly composite materials. A broad range of lignocellulosic residues—such as sawdust, wood chips, bark, and other related byproducts—are evaluated for their potential incorporation into polymer matrices to create filaments and pastes appropriate for AM techniques. The paper initially examines the features of natural byproducts and their typical uses, then evaluates the benefits that AM presents in comparison to conventional manufacturing techniques. Special emphasis is placed on the physicochemical and mechanical properties of the developed composites, encompassing their thermal characteristics (glass transition temperature, melting point, and stability), density, and mechanical behavior under both static and dynamic loading. Furthermore, the environmental effects of these composites are thoroughly assessed through Life Cycle Assessment (LCA), highlighting their contribution to

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minimizing ecological footprints and promoting circular economy initiatives. Collectively, the findings indicate that the additive manufacturing of composites derived from natural byproducts represent a promising pathway toward sustainable industrial production.

Keywords: sustainability; additive manufacturing; wood byproducts; composites; LCA

1. Introduction

In recent years, the use of natural byproducts, especially wood-based materials, has constantly increased and based on data from the Food and Agriculture Organization of the United Nations, it is projected to increase by 37% globally by the year 2050 [1]. According to data from the European Union, in 2017 approximately 880.0 million cubic meters of wood were consumed and utilized for lumber applications, for energy production, or in the paper mill industry [2,3]. Most of the produced waste was mainly disposed of in landfills and had limited use because the treatment process was deemed as time-consuming, resulting in high carbon dioxide emissions and environmental pollution. Therefore, the need for appropriate exploitation technologies has emerged for the efficient post-treatment of those wastes.

Additive manufacturing (AM) is a rapid manufacturing technique that enables efficient utilization and reuse of these byproducts. The design's flexibility and high accuracy without the need for intermediate steps (e.g., mold construction) promote AM as a reliable alternative solution for innovative sustainable manufacturing compared to other conventional processes. The layer-by-layer deposition of the feedstock material allows for the limitation of the produced waste materials, leading to an almost net-zero waste manufacturing process which is aligned with the European Union's plan for the development of sustainable manufacturing systems, helping in the achievement of Sustainable Development Goal 12 for the development of responsible and sustainable production patterns by the year 2030 [4–8]. The use of reacquired wood byproducts could help elevate the mechanical properties, reduce production costs, and create an esthetically pleasing result, allowing for a smoother surface finish and increased biodegradability of the final products, allowing for further reuse and improved recyclability.

Recyclability issues have already been stated in previous research, and the efficient utilization of wood byproducts has emerged as an efficient solution to satisfy circular economy and sustainable production demands. These solutions include making closed-loop systems where resources and materials are continuously reused and recycled instead of being discarded as waste; industrial symbiosis where various industries come to an agreement so that each one uses the waste of the other as raw materials, reducing production costs; as well as the 'cradle-to-cradle' technique promoting the creation of products that can be reused or safely reintegrated into the environment [9–11]. Zanuttini et al. [12] proposed, as a viable solution, the development of wood-based composites (WBCs) due to the biodegradable nature of the filler and the broad range of non-thermal challenged applications, from household products to structural engineering applications. Miseljic and Olsen [13] applied Life Cycle Assessment (LCA) analysis to five polymer-based everyday products and found that insulation panels made out of polyvinyl chloride (PVC) matrices and wood product fillers presented better mechanical stability and sustainability compared to conventional nanomaterials. Fico et al. [14] utilized olive branches on PLA matrices, creating an all-natural composite via Fuse Filament Fabrication Additive Manufacturing and assessed printability, architectural accuracy, and demanded energy via LCA analysis. It was noted that for every 20% of reacquired wood, an overall 10% reduction in the environmental impact was observed due to significantly less feedstock material and energy demands.

In this context, the aim of this review paper is to provide sufficient feedback regarding the utilization of natural wood byproducts in AM processes, delving into the connection between clean feedstock acquisition and net-zero waste manufacturing processes for the development of sustainable materials and closed-loop manufacturing systems aiming to address environmental footprint issues. In Section 2, an overview of the already used wood byproducts and the source of acquisition is presented, along with the existing possible applications, and the case is made for AM as an efficient way of valorizing those end-of-life materials by comparing the findings with conventional manufacturing techniques. Section 3 consists of the experimental evaluation of the proposed wood-filled composites' physicochemical response and performance evaluation by assessing crucial static and dynamic mechanical properties. Finally, in Section 4, the probability of upcycling is presented with examples of Large-Scale Additive Manufacturing for real-life applications, along with the projected impact on the future environmental goals.

2. Feedstock Analysis, Treatment, and Manufacturing Processes

2.1. Different Types of Raw Materials

2.1.1. Acquisition Source

During wood processing, a multitude of byproducts are generated, which differ in nature and use. These byproducts include sawdust, wood chips, and bark. An important factor that influences the characteristics of these byproducts is the type of wood they come from. These types are divided into two categories: hardwood and softwood. Softwood comes from coniferous trees of the gymnosperm order, such as cedar (Cedrus), pine (Pinus), cypress (Cupressus sempervirens), and fir (Abies). Hardwood is found in trees such as oak (Quercus pubescens), beech (Fagus), chestnut (Castanea sativa), poplar (Populus), and plane (Platanus); these are broad-leaved trees of the angiosperm order [15,16]. An overview of the acquisition sources based on the type of wood and the presence of the seasonal leaf abscission process is presented in Figure 1.

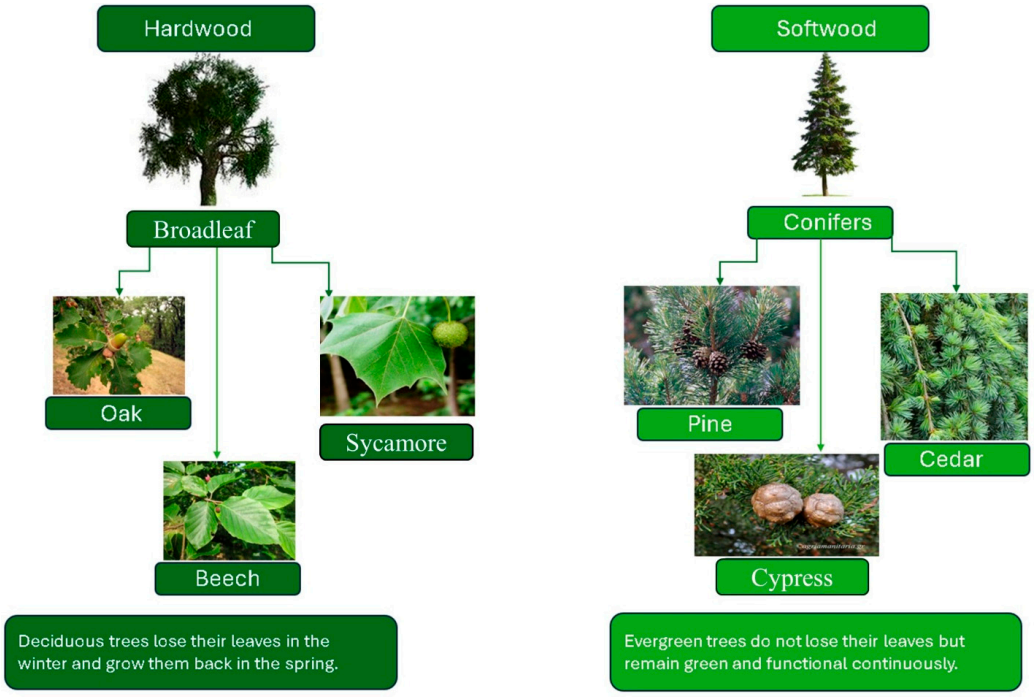


Figure 1. Categorization of feedstock based on the tree type.

Depending on the tree type of Figure 1, differences have also been observed in the cell structure of the acquired wood. Softwood has a simpler cellular organization and lower density, making it lighter and easier to process. In contrast, hardwood has a more complex structure and greater density, giving it increased resistance to wear [17]. The type of wood from which the byproducts are generated also imparts some properties to them. For example, pine (Pinus) sawdust is often chosen for its high resistance to shrinkage, swelling, and vibrations, as well as for its characteristic aroma [18]. On the other hand, oak (Quercus pubescens) sawdust is known for its hardness and durability, as well as its natural antifungal properties, which are due to its high tannin content [19]. Furthermore, oak is one of the main sources of wood for furniture manufacturing in European markets, which implies the generation of significant quantities of byproducts [20]. An overview of the main differences in the bulk and cellular structure between hardwood and softwood is presented in Figure 2.

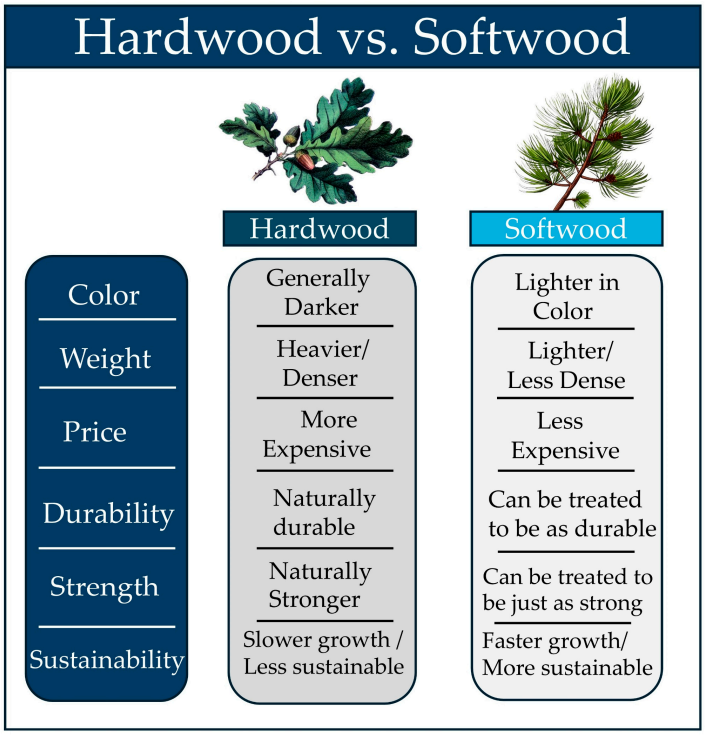


Figure 2. Basic differences between hardwood and softwood.

2.1.2. Structural Characterization of Wood Byproducts

Wood byproducts consist mainly of cellulose (45–50%), lignin (23–30%), hemicellulose (20–30%), and various extracts (acids, resins, oils, and soluble sugars, etc.) in percentages from 1.5 to 5% [21,22]. Sawdust consists of fine wood particles resulting from wood processing, such as sawing, sanding, and milling. Due to the porous nature of wood, sawdust is characterized by high absorbency and resistance to vibrations [23]. It is commonly used for manufacturing Medium-Density Fiberboard (MDF) and Oriented Strand Board (OSB). Wood chips are fragments of wood created during tree cutting or as remnants from the manufacturing process of solid wood products such as plywood. Their size ranges between 1 and 5 cm. They can have a flat, fine, or bulky shape depending on the original material and the production method from which they originated [24]. Wood chips are often used in energy production (pellets) and in the paper industry for the creation of pulp [25]. Bark consists of two main parts, the inner bark, also known as "active bark", which is living tissue and transports the necessary sugars for the tree's nutrition, and the outer bark, which

is dead tissue and acts as a protective layer [26]. Bark is often used as a natural insulation material [27].

2.2. Treatment Methods of Wood Byproducts

Wood byproducts have recently been utilized in the development of biofuels and biomaterials as part of the transition toward sustainable energy. Those second-generation biofuels present high yields per unit of arable land and contribute significantly to reducing pollutants [28]. Through thermochemical conversions such as Pyrolysis and Gasification, wood byproducts can be converted into bio-oil or synthetic gas (syngas), which can be further refined, thus reducing dependence on fossil fuels [29,30].

Wood byproducts can also be used for the development of biopolymers. The two most common types of composite wood are Medium-Density Fiberboard (MDF) and Oriented Strand Board (OSB). MDF is made from fine wood fibers bonded with resins, which are compressed under high pressure and temperature, resulting in the creation of a homogeneous board. It is a material without defects with a smooth surface, which has high resistance to vibrations but is quite vulnerable to moisture if it does not undergo some treatment, which is why it is mainly used in furniture manufacturing [31,32]. Oriented Strand Board (OSB) consists of larger wood fragments that are arranged in layers with orientation to enhance the mechanical strength of the material. Due to its high resistance to moisture and vibrations, it is mainly used in structural constructions [33]. The bonding of both materials is performed with synthetic resins such as Urea-Formaldehyde (UF) for MDF boards and Phenol-Formaldehyde (PF) resins for OSBs. In addition to synthetic resins, the bonding of materials can also be performed using biopolymers, such as Bio-Polyurethane (Bio-PU) and Lignin-based Polyurethane (LPU), which are produced from natural raw materials such as vegetable oils (e.g., rapeseed oil) and polyols from lignin and cellulose, components that can be derived from the processing of wood byproducts [34,35]. Bio-polyurethane constitutes an excellent alternative compared to synthetic binders, such as UF and PF resins, as it is environmentally friendly and safe for humans, given that it does not contain urea. Resins based on urea produce formaldehyde emissions during the bonding of products, which are harmful to humans [36,37]. Furthermore, products bonded with bio-polyurethane can be reused multiple times or safely returned to the environment (Figure 3), as they decompose more easily and safely compared to products that have been bonded using synthetic resins. Moreover, the bonding process with bio-polyurethane is characterized by low energy cost, as it does not require thermal processing [38,39].

The utilization of wood byproducts is also encountered in the paper and pulp industry [40]. Residues such as sawdust, wood chips, and lignin are often used either as reinforcing materials in papermaking or as basic raw materials in pulp production. Depending on the method used to pulp the materials, the properties of the produced paper change. Mechanical pulping (Thermo-Mechanical Pulping and Chemo-Thermo-Mechanical Pulping) allows for the production of paper with high lignin content, imparting strength and rigidity to the final product, as well as high moisture absorbency, while chemical kraft pulping removes lignin, creating a purer cellulose. This results in the production of paper that is more resistant to chemical and mechanical stresses [41]. The technique of adding lignin to thermoformed pulp is another technique that improves the water repellency and mechanical strength of the paper, making it particularly suitable for use in protective packaging [42,43]. In Figure 4, an overview of the possible products from the aforementioned processes is presented.





Properties	Biopolyurethane (BPU)	Urea-formaldehyde (UF)
Origin	Renewable biological sources (e.g., oils, polysaccharides, lignin)	Synthetic resin, consisting mainly of formaldehyde and urea
Biodegradability	High	Low
Mechanical Properties	Moderate strength, flexible	Stronger but brittle
Moisture Resistance	Composition dependent but good overall	Low, absorbs water and degrades easily
Durability	Resistant to wear, moisture and chemicals	Vulnerable to humidity and temperature
Environmental Impact	Environmentally friendly, reduced carbon footprint	Potentially harmful due to formaldehyde emissions
Toxicity	Low, does not emit formaldehyde	Potentially harmful due to formaldehyde emissions
Cost	Higher due to the complex production process	Low, based on established production methods

Figure 3. Comparative analysis of Bio-Polyurethane (BPU) and Urea-Formaldehyde (UF).

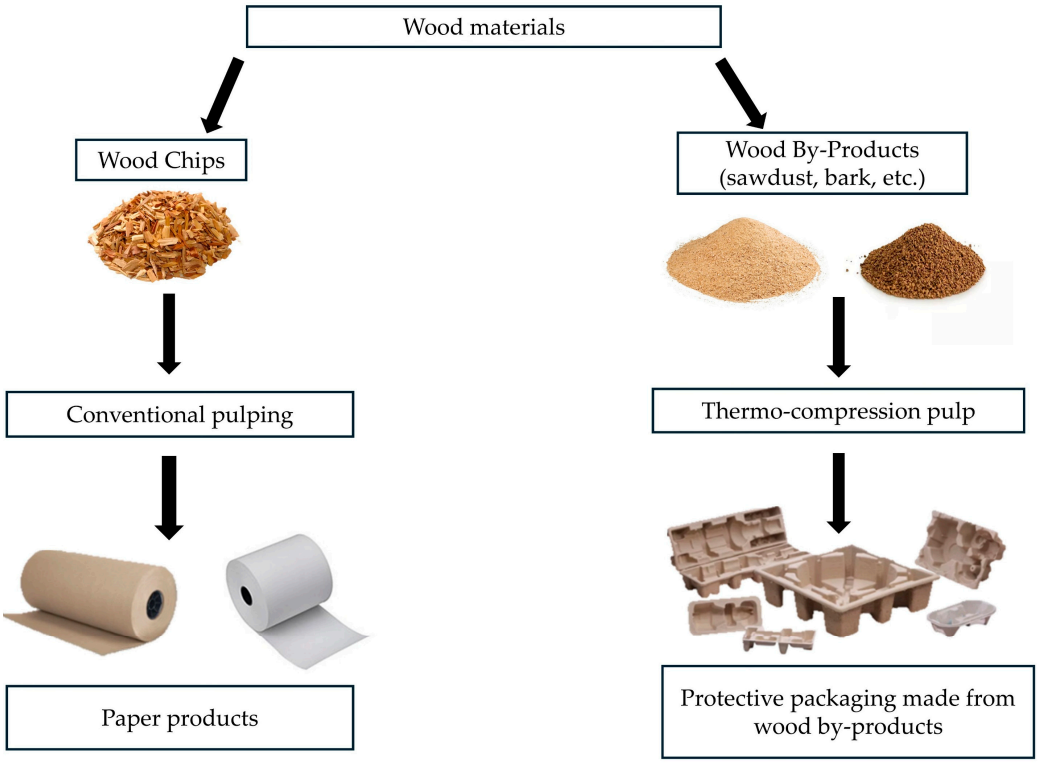


Figure 4. Schematic representation of the application used from wood material and byproducts.

2.3. Utilization of Wood Byproducts in Manufacturing Processes—The Case of 3D Printing

Additive manufacturing constitutes a modern approach for the utilization of wood byproducts and their integration into the manufacturing process. Materials that were

previously considered waste or had very limited uses, are now being redefined as functional and useful raw materials [44,45]. Additive manufacturing minimizes waste production as it uses only the necessary amount of material, in contrast to traditional methods such as machining, where material is removed until the desired product is manufactured [46,47]. The growing interest in their integration into additive manufacturing is due to both their availability and their ecological character. An indicative illustration of AM's significant advantages in sustainable manufacturing compared to other conventional methods is presented in Figure 5.

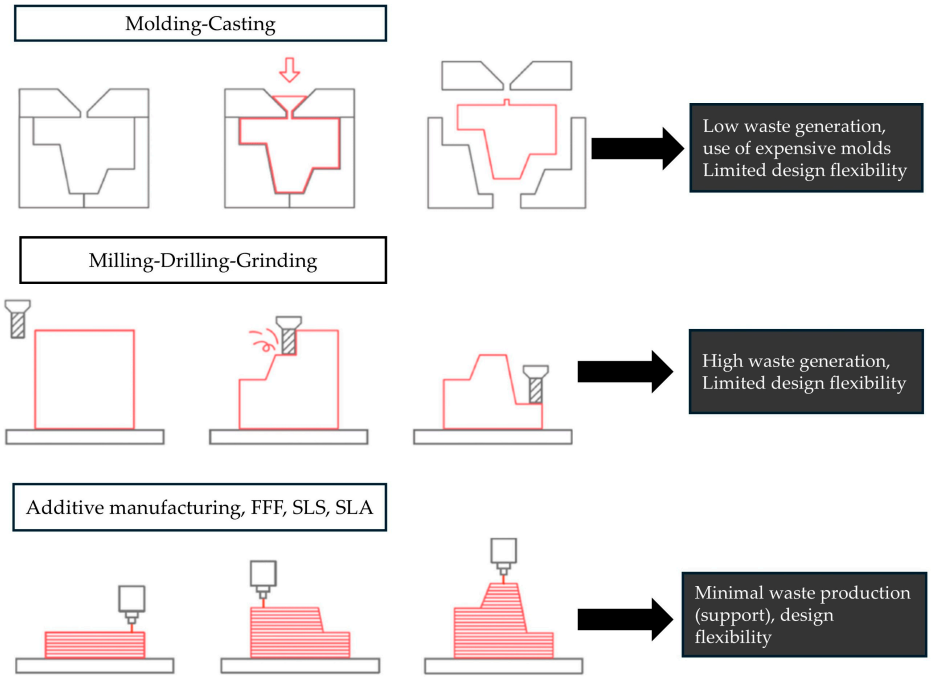


Figure 5. Operating differences between AM and indicative traditional methods and effect on waste production.

In Figure 5, it is shown that the AM operating process allows for the use of the exact amount of needed material, limiting material waste. Some concerns have emerged in the filament creation process and the financial and environmental cost of the process [48,49]. Byproducts such as sawdust and cellulose, as well as wood ash can be incorporated into a thermoplastic matrix, aiming to produce composite filaments suitable for use via the Fused Deposition Modeling (FDM) method [50,51]. The most common PLA/wood byproduct ratios in FDM filaments with weight fractions ranged from 10 wt.% to 30 wt.%. The manufactured filament retains its natural texture and color. Furthermore, the use of these filaments reduces the need for non-renewable raw materials, promoting more sustainable ways of production [52,53].

Besides FDM, the Liquid Deposition Modeling (LDM) technique has emerged as an interesting alternative approach for the utilization of wood byproducts, mainly in paste form. This method uses natural binders, such as starch (often modified or enzymatically processed) and other natural resins, such as gum arabic and lignin, in combination with wood byproducts to create a printing material with high biomass content. The main advantage of the LDM technique is the flexibility it offers regarding the type of raw materials that can be used, as it allows for the utilization of materials that are not easily incorporated into thermoplastic filaments [54].

Growing interest in the use of additive manufacturing and wood byproducts is also observed in the construction sector. Specifically, the use of sawdust as an insulation material

in blocks made entirely of polylactic acid (PLA) has been examined. The blocks were constructed with an internal hexagonal structure (honeycomb type), which included cavities designed to function as pockets for insulating materials. After printing was completed, the internal cells were filled with sawdust, which acted as a natural insulating material, significantly reducing the thermal transmittance of the final material by 46.7% compared to the empty block, reaching a value of 0.65 W/m^2K. Furthermore, a more homogeneous thermal distribution was recorded across the surface of the specimen, which was attributed to the uniform distribution of the sawdust within the cavities [55].

The utilization of wood byproducts for the development of composite materials constitutes a significant field of additive manufacturing, particularly when these are combined with other natural raw materials. Byproducts such as sawdust and lignocellulosic fibers have been utilized for the production of mixtures that include starch, natural resins, and lignin, but also agricultural residues (rice husk and corn stalks), aiming to create pastes as well as filaments, suitable for FDM and LDM techniques. Such hybrid compositions enhance the biological basis of the material and can impart properties such as hydrophilicity, filtration capacity, and controlled biodegradation, depending on the specific mixture. Thus, composite materials constitute a dynamic and ecological solution, which can be adapted to different needs of the manufacturing process [56,57]. In Figure 6, an illustration of the three most well-known AM techniques is presented with their compatibility with recycled materials and wood-based materials.

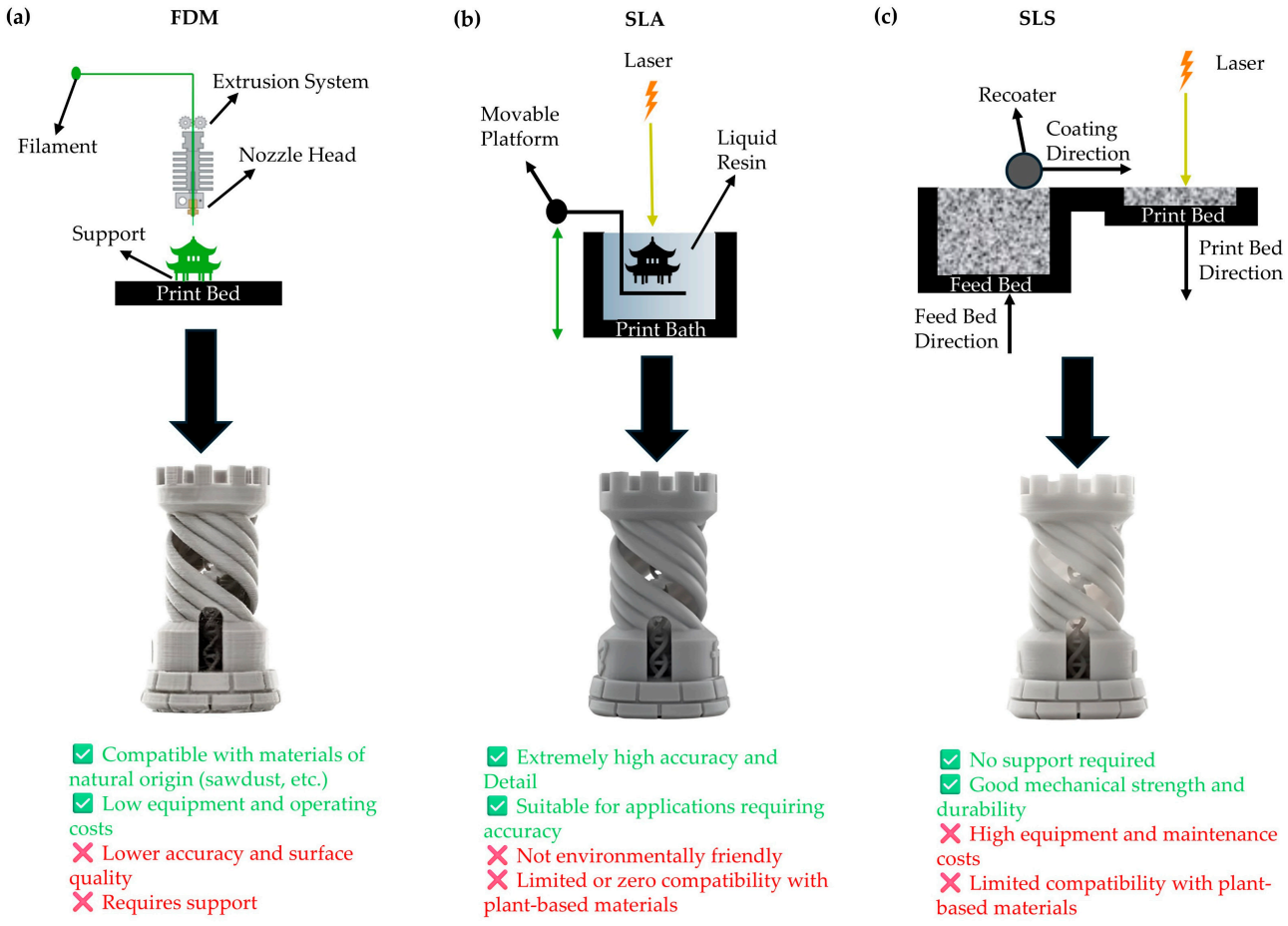


Figure 6. Operating differences between AM techniques and their effect on waste production; (**a**) FDM; (**b**) SLA; (**c**) SLS.

As shown in Figure 6, wood byproducts have great compatibility with material extrusion techniques such as the FDM AM. Custom-made filaments have been widely reported in the literature. Stereolithography presents high accuracy and surface finish, but it is not compatible with sustainable manufacturing processes. On the other hand, Selective Laser Sintering (SLS) has gained interest as a viable alternative for sustainable manufacturing. The high accuracy and the ability to print without support structures can help in the process of rapid manufacturing of functional products. Although wood powders present limited compatibility at the moment, recent studies show increasing interest in the integration of lignocellulosic raw materials into powder sintering technologies [58,59].

2.4. Environmental Perspective of the Valorization of Wood Byproducts via 3D Printing

The integration of wood byproducts as printing materials in additive manufacturing can lead to a significant reduction in the environmental footprint, as they constitute a renewable raw material that is environmentally friendly. The construction of wood-based polymers is compatible with the requirements of three-dimensional printing. This fact reduces the need for the use of conventional petrochemical-based materials such as ABS (Acrylonitrile Butadiene Styrene), PET (Polyethylene Terephthalate), and Nylon-12 (Polyamide), thus contributing to the reduction in carbon dioxide emissions. At the same time, it allows for the reintegration of residues back into the production process, strengthening the principles of the circular economy and the strategy of sustainable development [4,60].

Govedic et al. [61] conducted LCA analysis on the production of three different types of furniture components, one fully metallic, one made entirely of polymer, and one composite of polylactic acid and wood with up to 40% wood content. The composite material presented up to 73% lower Global Warming Potential (GWP) compared to the metallic component, while it also outperformed the pure polymer in terms of overall environmental performance. However, the study focused mainly on carbon dioxide emissions and highlighted the need for a more comprehensive and multi-metric application of LCA that is not limited only to CO_2 emissions but would consider indicators such as land use and water consumption, in order to obtain a more complete picture of the environmental footprint of these materials. In summary, the use of wood byproducts in additive manufacturing offers a more responsible way of production, reducing the environmental footprint of manufacturing processes.

3. Evaluation of the Wood Byproducts Composites via Additive Manufacturing Processes

3.1. Physicochemical Evaluation

3.1.1. Glass Transition and Melting Temperatures

Glass transition temperature (T_g) and melting temperature (T_m) are vital for the printability of the materials. T_g is the temperature at which the material changes from a brittle, "glassy" state to a more flexible and elastic form, while T_m expresses the point where the materials transition from solid to liquid phase. The addition of sawdust on natural polymer matrices resulted in a gradual increase in the melting temperature with the elevation of the sawdust weight fraction [48]. This influence was evident due to the simultaneous effect of sawdust on the crystallinity (increase in the crystallinity temperature, T_c) of the composite, highlighted by the elevated crystallization temperature [62]. This thermal behavior is also visible in Figure 7, where Differential Thermal Analysis (DTA) was conducted on polylactic acid (PLA)–sawdust composites on different sawdust weight fractions.

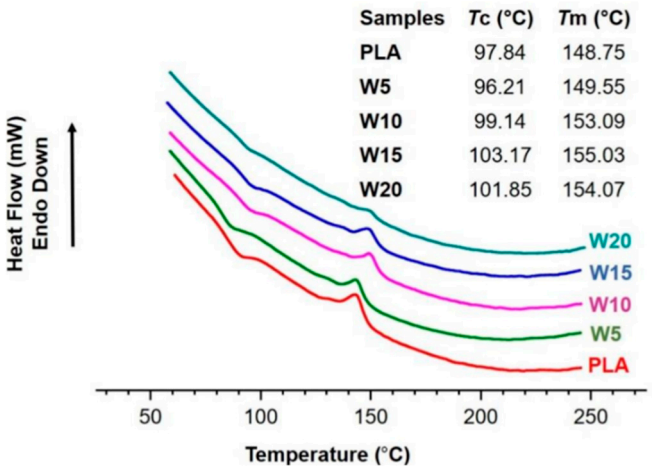


Figure 7. Evaluation of the influence of wood sawdust filler on PLA matrix regarding the crystallinity and the melting temperature [48].

Despite the affection in T_m and T_c, the insertion of wood sawdust did not result in a significant deviation in T_g. This indicates that the mobility of polymer chains in the amorphous phase is not significantly affected by the addition of sawdust, and the final dispersion is homogeneous in weight fractions up to 30 wt.% [63]. Higher weight fractions resulted in poor dispersion, leading to reduced thermal stability. Sawdust particle size and morphology also played a pivotal role in structural integrity. Bulkier particles resulted in nozzle clogging. The findings suggested that the sawdust's particle diameter has to be below 50% of the utilized nozzle diameter to avoid continuity disruption and nozzle clogging [64]. The main findings regarding the weight fraction, the fillers' morphology, and the printing conditions regarding the printability of the composites are summarized in Table 1.

Table 1. Main findings regarding the influence of wood fillers on natural polymers [48,62,64].

Content	T_g (°C)	T_m(°C)	Main Findings
Elevated weight Fraction (up to 30 wt.%)	Stable	Increase	Steady molecular mobility, increased crystallinity
Fine particle size	Stable	Stable	Optimal thermal properties, good dispersion
Bulk particles	Stable	Stable	Thermal stability, printability issues due to structural disruption

3.1.2. Composites' Density and Infill Density's Influence on Printability

The composites' density is relevant to the wood byproducts' nature (e.g., hardwoods tend to be bulkier) but also relevant to the printing conditions regarding the infill density. Infill density was shown to have an immediate effect on the printability and the final dispersion of the composites. Agaliotis et al. [65] used wood flour from the henequen plant (agave), with a content of 1–5 wt.%, and a gradual increase in density was observed from 1.24 g/cm^3 for pure polymer up to 1.31 g/cm^3 at maximum reinforcement. The increase was attributed to the higher density of fibers and their good dispersion within the polymer matrix. Buschmann et al. [66] printed wood composites using individual layer fabrication (ILF); the density of the composites was found to range between 704.45 and 878.45 kg/m^3, for wood contents from 82.95 to 89.70 wt.%, with this variation being related both to the amount of wood incorporated and to the printing parameters. Better interlayer bonding

resulted in higher final density [67]. Print speed also influenced density, with high speeds resulting in a decrease in pore formation between layers. Modifying printing conditions can result in controlled porous formation leading. Desired porous formation can result in lightweight materials with adequate structural formation (architected materials) [68]. Overall, the addition of wood byproducts such as sawdust and wood flour tend to reduce nominal material density, due to the lower density of wood compared to polymer. However, the final, actual density of the printed object is more affected by printing parameters such as infill density and printing speed, which determine interlayer bonding [67]. In Figure 8, the effect of various wood byproducts on the PLA matrix regarding the composites' density is illustrated.

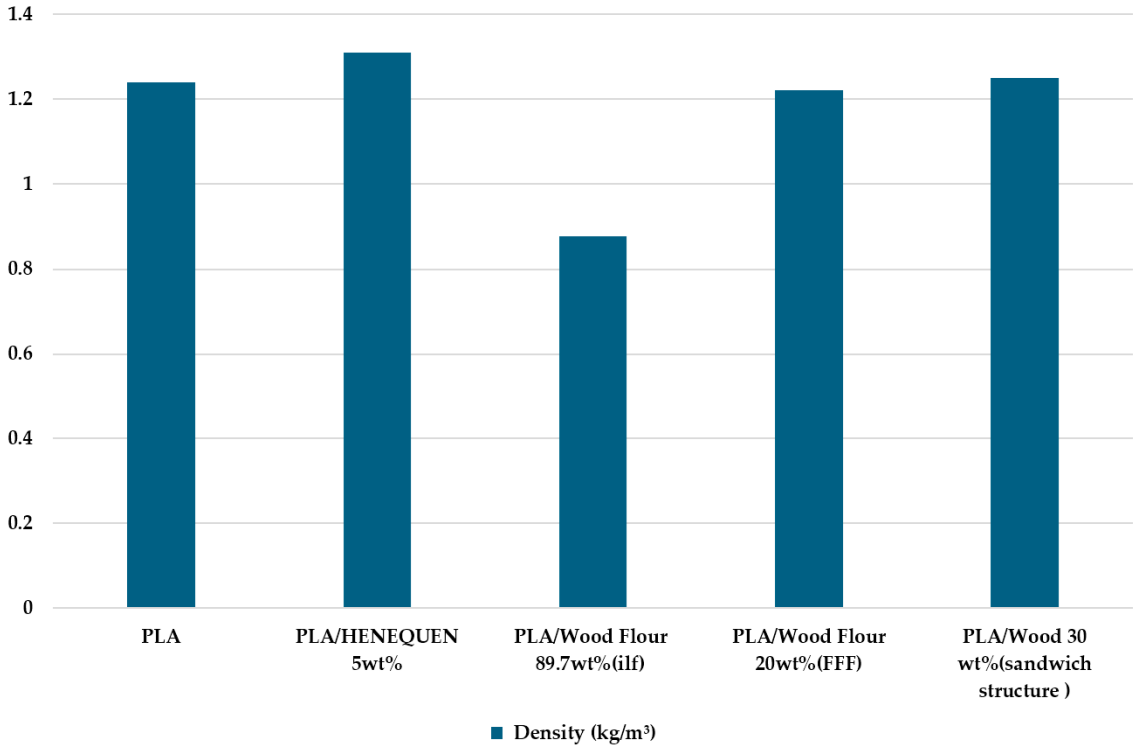


Figure 8. Density (kg/m^3) of pure PLA and PLA/wood composites with different reinforcement percentages.

3.1.3. Thermal Stability

Thermal Stability evaluation is vital for a numerous of industrial applications. Thermogravimetric analysis (TGA) and DTA were utilized for the assessment of the wood composite's response to thermal stresses. Wood filler insertion resulted in lower degradation temperatures, which could lead to easier decomposition and enhance recyclability, aligned with the recent environmental goals [48,69]. This shift in the degradation temperature with the gradual increase in the weight fraction of wood byproduct is presented in Figure 9.

In addition to the influence on degradation temperature, an increased char residue was observed in the wood composites. This existence could be useful in applications where flame retardancy or heat resistance are required. Also, char residue could be utilized via pyrolysis for the development of biomaterials or biofuels [6,48].

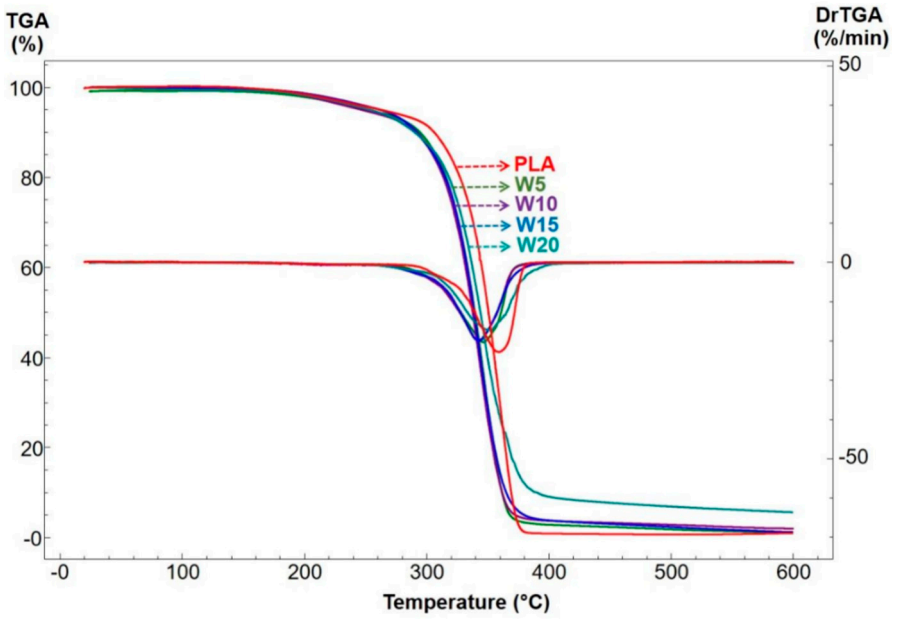


Figure 9. TGA and DrTGA thermograms of the wood/PLA composite filaments [48].

3.1.4. Intermolecular Interactions

The interfacial cohesion between polymer matrix and wood byproducts (sawdust and wood flour, etc.) is largely influenced by intermolecular interactions between these two phases (polymer/wood). The most significant are Van der Waals forces and hydrogen bonds, which affect the mechanical, thermal, and processing properties of the composite. The surface of wood particles contains hydroxyl (–OH) polar groups, which often form hydrogen bonds with carboxyl (–COOH) and ester groups of the thermoplastic. Carboxyl groups are located at polymer chain ends, while ester bonds form the main structural unit of the polyester chain. However, due to the hydrophilic nature of wood and the hydrophobic nature of PLA, natural compatibility is limited, resulting in low interfacial adhesion and micro-voids at the contact zone [70] (Figure 10).

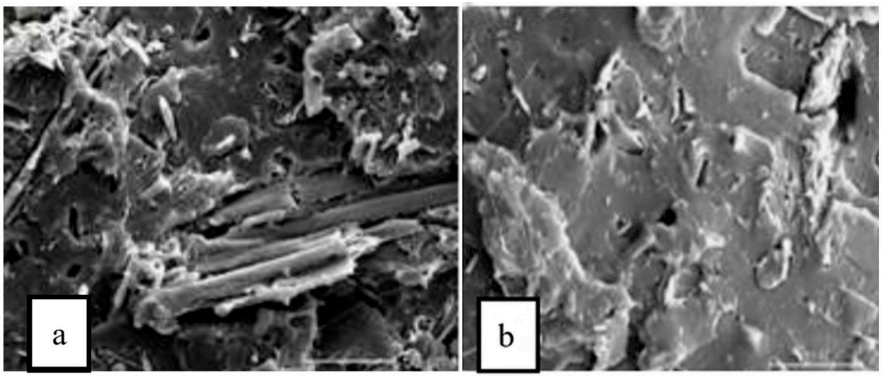


Figure 10. SEM for filaments: (**a**) 4% glycerol (**b**) 25 glycerol + 2% TBC (tributyl citrate) [70].

Chemical modification can be applied to enhance interfacial cohesion. Silane treatment of wood flour with coupling agent KH-550 can increase tensile strength by 25.3% (from 35.6 MPa to 44.6 MPa) and flexural strength by 24.2% (from 70.8 MPa to 87.9 MPa). As seen in Figure 10b, the material's surface is clearly more homogeneous with indications of stronger adhesion between phases. Furthermore, through Fourier-transform infrared spectroscopy (FTIR), the modification of the filler is evidenced by monitoring the changes in the spectral peaks of the hydroxyl groups (Figure 11).

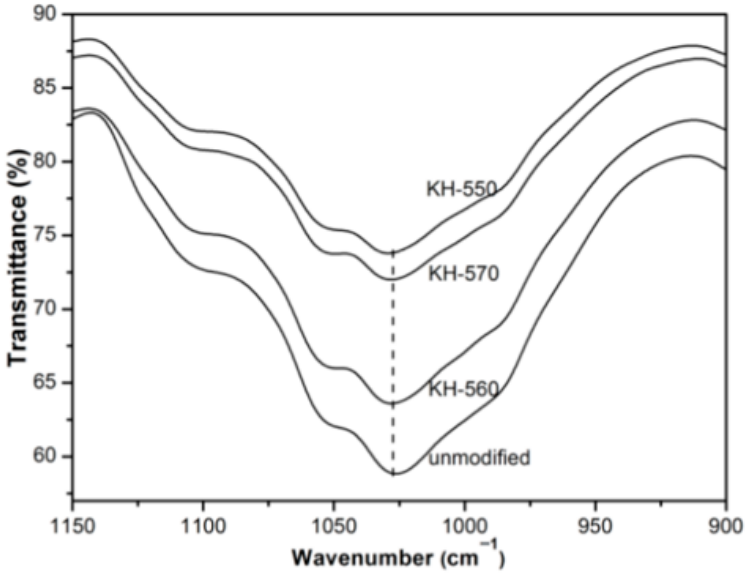


Figure 11. FTIR spectra in the 900–1150 cm^{-1} region for PLA/wood flour composites with and without silane modification [71].

In the 3200–3400 cm^{-1} region, the characteristic hydroxyl absorption (–OH) of wood is observed, which is related to the symmetric and asymmetric stretching of the O–H bonds. In KH-550-treated samples, this peak decreases significantly compared to untreated samples (Figure 11). This reduction indicates a reaction between silane groups and wood hydroxyls, reducing free polar groups and enhancing compatibility with the PLA matrix [70,71]. Similar results were recorded with teak wood flour chemically modified with APTES (3-aminopropyltriethoxysilane), which acted as an intermediary bond between wood hydroxyls and polymer polar groups, improving interfacial cohesion. This modification increased Young's modulus by 31.6% and maximum tensile stress by 18.4% compared to unmodified material [72]. Although weaker than hydrogen bonds, Van der Waals forces significantly contribute to the cohesion of the composite, especially when particles are well-dispersed and surface-modified. Adding plasticizers such as glycerin and tributyl citrate (TBC) in composites with 27 wt.% wood can improve dispersion and strengthen intermolecular interactions. With 4% TBC, tensile strength increased from 18.53 MPa to 24.60 MPa, and elongation after fracture from 1.14% to 1.77%, confirming Van der Waals' effectiveness [70,73]. Finally, the quality of the intermolecular bonds affects not only the mechanical performance of the material but also its flow and printability. The improved interfacial cohesion reduces the risk of nozzle clogging and increases the accuracy of the layers [63,72].

3.2. Influence of Size and Morphology on Printability

Particle distribution and size of wood fillers in the polymer matrix directly affect filament quality, extrusion flow, and final object quality. Particle diameter is especially critical, and size is closely related to flow and clogging risk during printing. In tests using five particle size ranges (0.2–1.0 mm) with a 0.6 mm nozzle, the extrusion current increased from 0.79 A (for <0.2 mm) to 1.27 A (for 0.6–0.8 mm), while printing completely failed for >0.8 mm. The best results were recorded for 0.2–0.4 mm particles [64], as shown in Figure 12.

Homogeneous dispersion and final object integrity are not only dependent on filler particle size. Functional groups of fillers and their compatibility with the matrix also determine the quality of integration. Both chemical modification (e.g., silane treatment) and physical modification (e.g., thermal treatment) have proven effective in improving

distribution and compatibility. Tomec et al. [74] found that the thermal treatment of wood particles at 200 °C reduced the mean size from 242 µm to 163 µm and the standard deviation from 210 µm to 166 µm. Thermally modified particles (TM) also showed up to 28% lower surface roughness (Sa) and reduced porosity (0.063 cm^3/g compared to 0.653 cm^3/g for untreated, with 20% filler). In Figure 13, the SEM images of these materials show that the modified samples (right) exhibit a more homogeneous structure, without visible agglomeration regions or large voids, while the cross-section of the filaments displays a more rounded shape and reduced surface roughness.

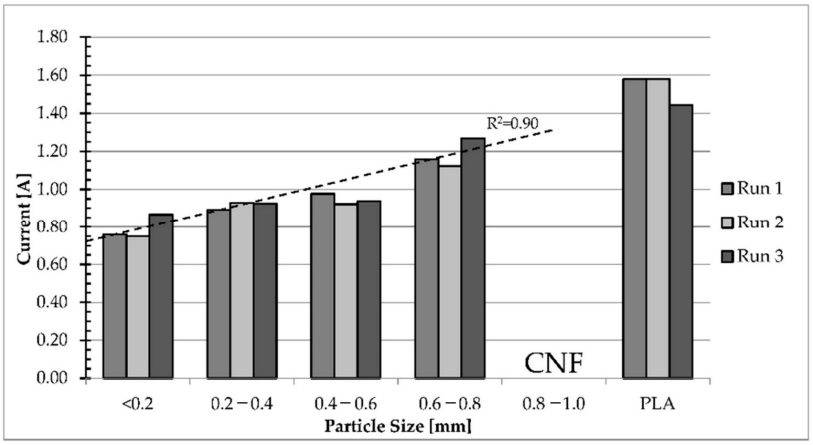


Figure 12. Relationship between particle size and extrusion current [64].

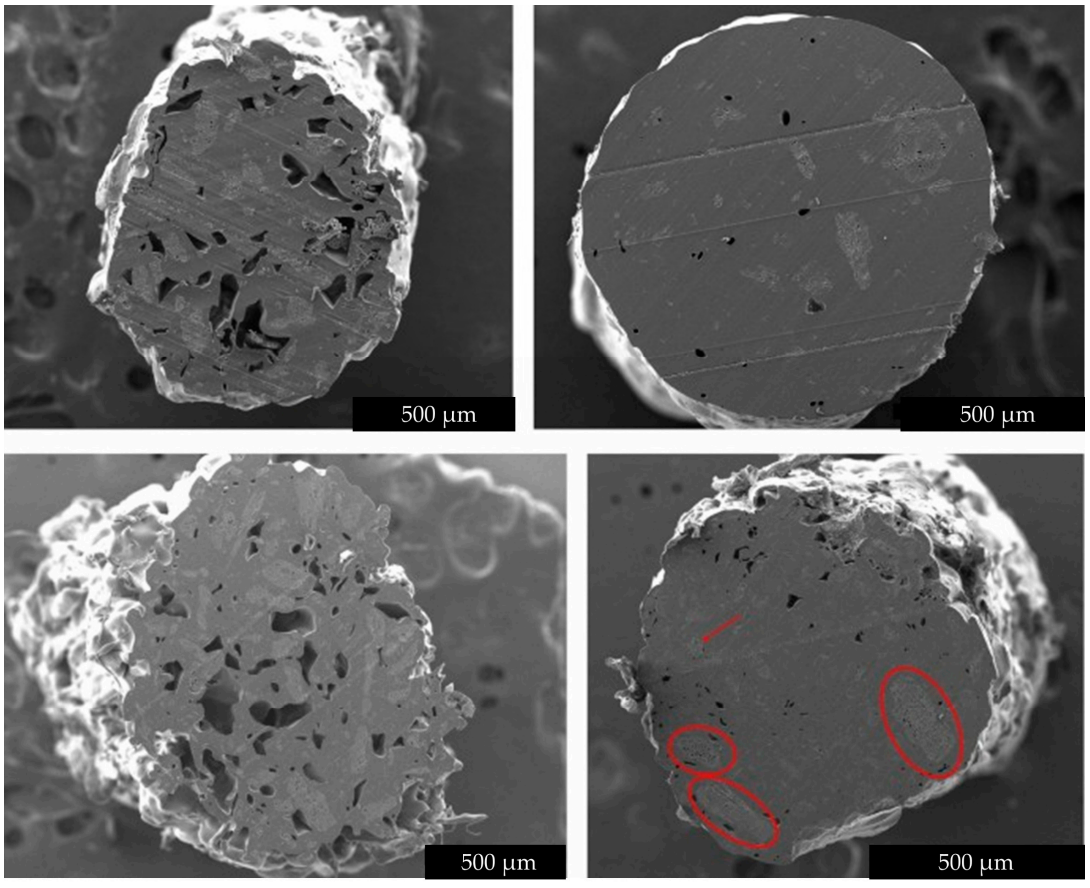


Figure 13. SEM images of PLA/wood filament cross-sections with thermally modified (**right**) and unmodified (**left**) particles. The circles and the arrow point to wood particles that are well-embedded in PLA [74].

Chemical modification of fillers with silane can also improve flow and printability for PLA/teak wood composites [72]. The chemical modification of fillers with silane can also improve flow and printability. For PLA/teak wood composites, silane-treated particles (<75 μm) enhanced extrusion flow and surface quality. Similarly, in composites with 10% silane-modified micro cellulose fibers (S-MCF10) [75] samples printed via a 0.6 mm nozzle at 40 mm/s and 210 °C with 100% infill showed smoother surfaces, full layer adhesion, and no clogging or roughness issues.

3.3. Static Mechanical Evaluation

For the static evaluation, results from tensile, flexural, and compression testing were taken into consideration. The filler's content, the porosity presence, and the fillers all influenced the final structure's integrity. Printing parameters also played a pivotal role in the optimization of the mechanical properties.

3.3.1. Tensile Strength

The feedstock's nature, dispersion, and printing quality have an immediate effect on the tensile properties of the final structure. Kartal et al. [6] utilized waste beech sawdust (WBS) and reported a gradual decrease in the Ultimate Tensile Strength (UTS) up to 40–45% in wood weight fractions of 20 wt.%. Pinewood sawdust also led to a moderate reduction in UTS, but this drop was evident mainly due to the presence of porous and micro-voids [48]. The insertion of chemical stabilizers and modifiers helped to address minor printability issues and resulted in a slight increase in UTS and Young's modulus [75]. In composites where maleic anhydride (MA) was inserted as a chemical stabilizer, a noticeable 15% improvement of the UTS was observed compared to the untreated samples [76,77]. MA presence aids in the formation of hydroxyl groups (-OH) on the filler's surface, therefore improving interfacial adhesion and resulting in unified stress transfer throughout the composite structure [78,79]. Design optimization also positively influenced the mechanical response. Kechagias et al. [68] developed sandwich structures with a matrix of PLA and PLA/wood layers. This distribution resulted in 100% of the UTS and a 60% increase in the Young's modulus. The effect of wood fillers on the polymer matrix, with the PLA matrix used as a reference point, is presented in Figure 14.

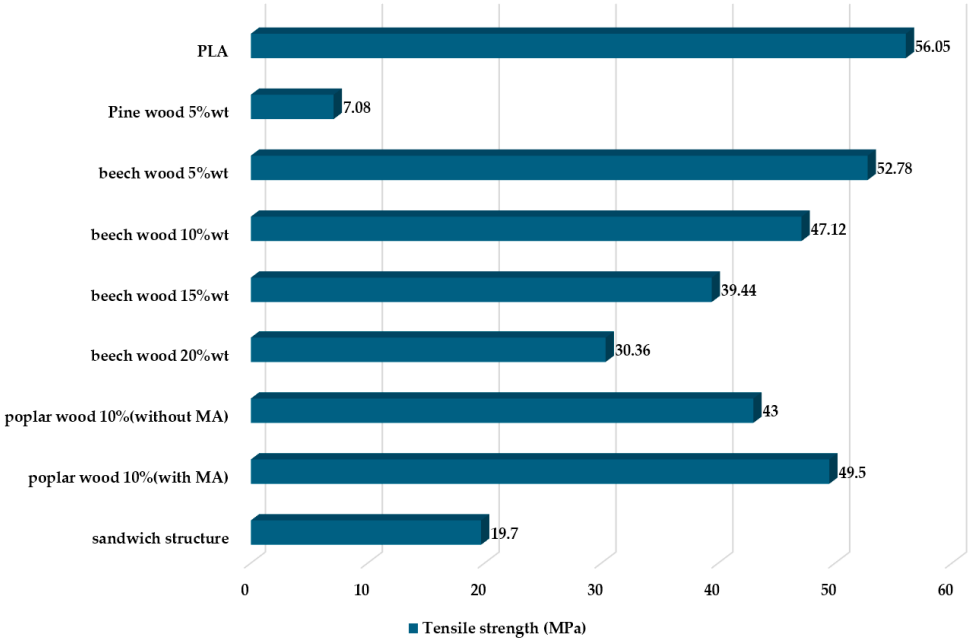


Figure 14. Effect of wood fillers and chemical modifications on UTS [48,68,75–77,79].

3.3.2. Flexural Strength

Flexural strength's value was determined again by the nature of the feedstock (hardwood or softwood), along with the feedstock and printing characteristics. WBS inserted in polymer matrix (65.12 MPa) resulted in a slight decrease in the flexural strength at 10 wt.% (55.58 MPa), but this drop was rapidly increased when the insertion was elevated to 20 wt.% (25.90 MPa), leading to the conclusion that WBS had minimal suitability with the polymer and too much filler can lead to unstable structures [6]. This drop was more controlled in the case of poplar wood sawdust, where adequate properties were maintained in weight fractions up to 30 wt.% [63], while pine wood sawdust resulted in higher flexural strength but inadequate dispersion was observed in concentrations higher than 15%, which led to instabilities and a drop in the absolute value but it was still higher than the neat polymer [48]. Overall, without the presence of chemical modifiers, wood fillers can help maintain or slightly improve flexural strength but in relatively low weight fractions (5–15 wt.%), to ensure unified filler distribution and optimal properties. Chemical modifications with Microcrystalline Cellulose Fibers (MCFs) helped to address instability issues and positively influenced the overall flexural strength. The existence of MCFs influenced the degree of homogeneity and resulted in a unified stress distribution into the structure while simultaneously resulting in the minimization of unwanted porosity or micro-voids [63,77]. In Figure 15, the flexural strength's deviation regarding the wood's concentration and the existence of chemical modifiers is presented.

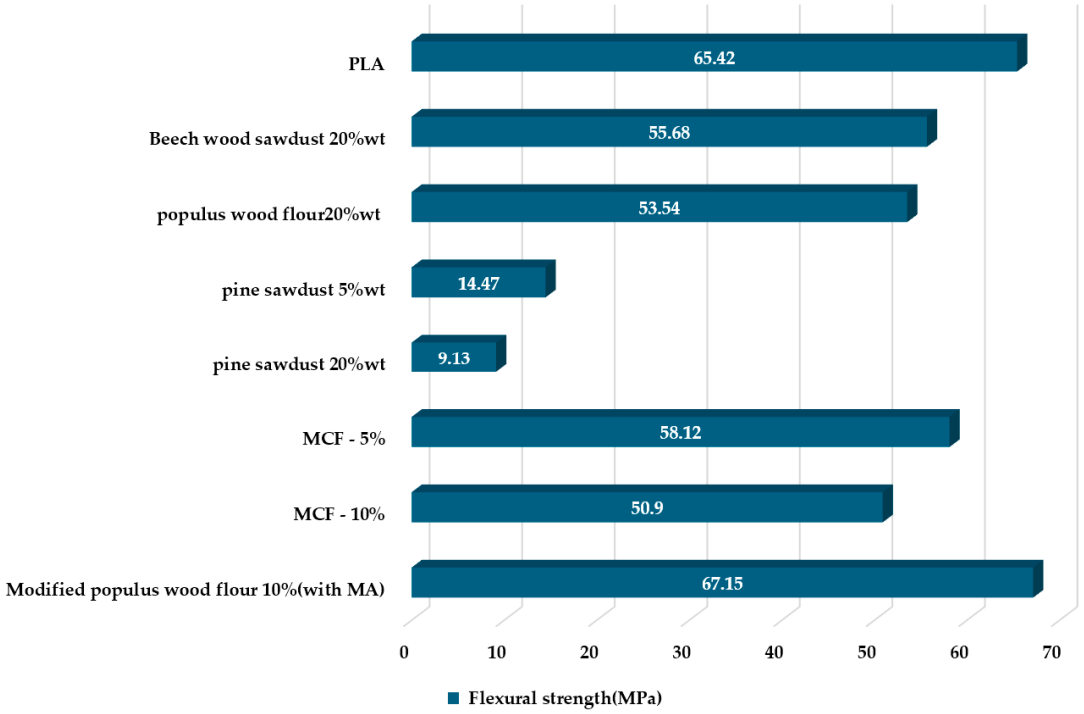


Figure 15. Effect of wood fillers and chemical modifications on flexural strength [6,48,63,75].

3.3.3. Compressive Strength

Regarding the compressive response of the wood–polymer composites, all the aforementioned factors regarding filler's concentration, printing parameters, and printing quality overall, a more positive response was observed in all types of woods and the ability to form high weight fraction (>20 wt.%) composites. In fact, concentrations around 20 wt.% were deemed optimal in terms of achieving structures with the highest compressive strength [80]. Structures with wood fractions higher than 50 wt.% presented significant defects and a rapid decrease in their overall response. This drop was evident due to the fact that in

most cases, no stabilizers were utilized and because at higher fractions, the hydrophobic polymer matrix did not present adequate compatibility with the hydrophilic wood filler [81]. Printing speed also played a pivotal part in the optimization of the properties, with low printing speed resulting in better structural integrity due to lower heat diffusion time, leading to improper interlayer welding. Silane-based chemical modifiers are deemed the optimal solution for the maximization of the compressive behavior of wood–polymer composites [82,83]. Besides FMD AM, Liquid Deposition Modeling (LDM) AM was also utilized in this case, but the results were heavily influenced by the wood's weight fractions and morphological characteristics, highlighting the relationship between the wood's filler nature and the proper treatment of the feedstock. Overall, promising results regarding the mechanical influence were extracted, highlighting that various types of wood can be utilized and provide tailored properties on the final structure [84]. An overview of the final compressive properties of various wood byproducts–polymeric composites is presented in Figure 16.

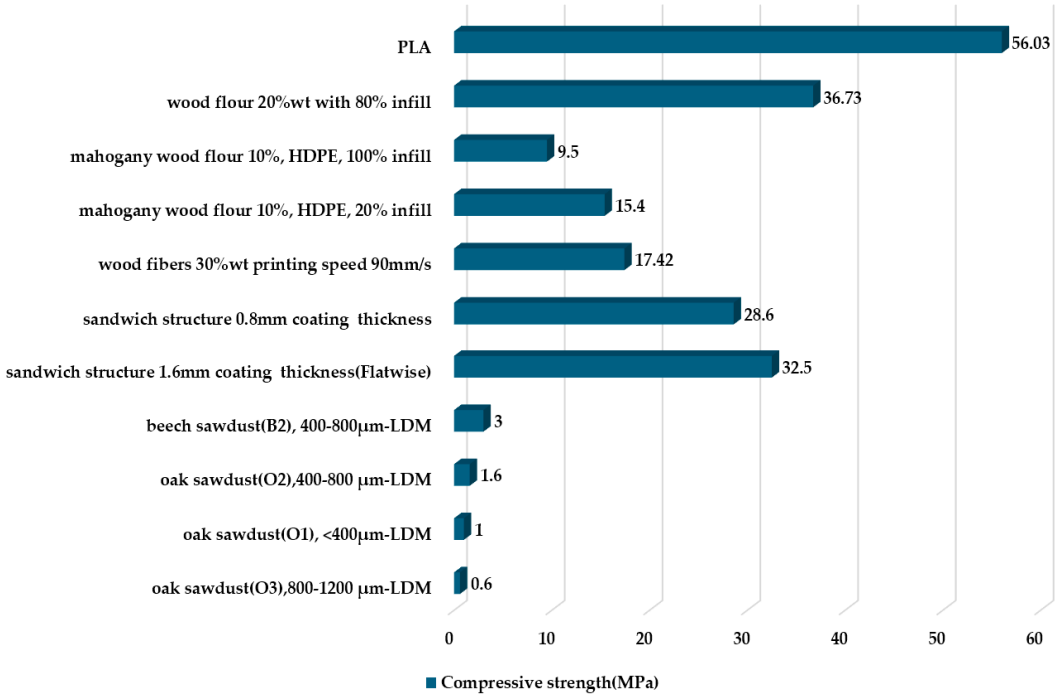


Figure 16. Effect of wood fillers and chemical modifications on compressive strength [80–84].

3.4. Dynamic Mechanical Evaluation

Dynamic characterization of materials is crucial for the assessment of the mechanical integrity in systems where multiple factors are influential to the overall response, standing as an important intermediary between the coupon level and real-life applications. Those tests are mainly thermal or frequency-dependent Dynamic Mechanical Analysis (DMA), Fatigue testing, or Impact testing (Charpy, Izod, and Drop Impact).

3.4.1. Dynamic Mechanical Analysis (DMA)—Thermomechanical Response

DMA testing examines the mechanical response in frequency or temperature changes by calculating the amount of stored energy (Storage Modulus, E′), the energy losses (Loss Modulus, E″), and the overall damping coefficient (tan δ = E″/E′). The insertion of wood sawdust regardless of the type of the wood's microstructures was linked with a positive influence on the Storage Modulus extracted from isothermal frequency sweeps. Regarding the glass transition temperature, thermal sweeps indicated a minor increase compared to neat polymers [85]. This transition is mainly evident due to the mobility between

the molecular chains, due to the presence of wood fibers, which inhibits intermolecular relaxation. An indicative example of this response is the insertion of pine wood sawdust into the PLA matrix, with the final composite (f-PLA) having a significant 20% increase in the storage modulus with a stable glass transition temperature compared to the natural polymer (n-PLA) [86]. This change is also evident in the temperature-dependent sweep of E′ and E″ in Figure 17.

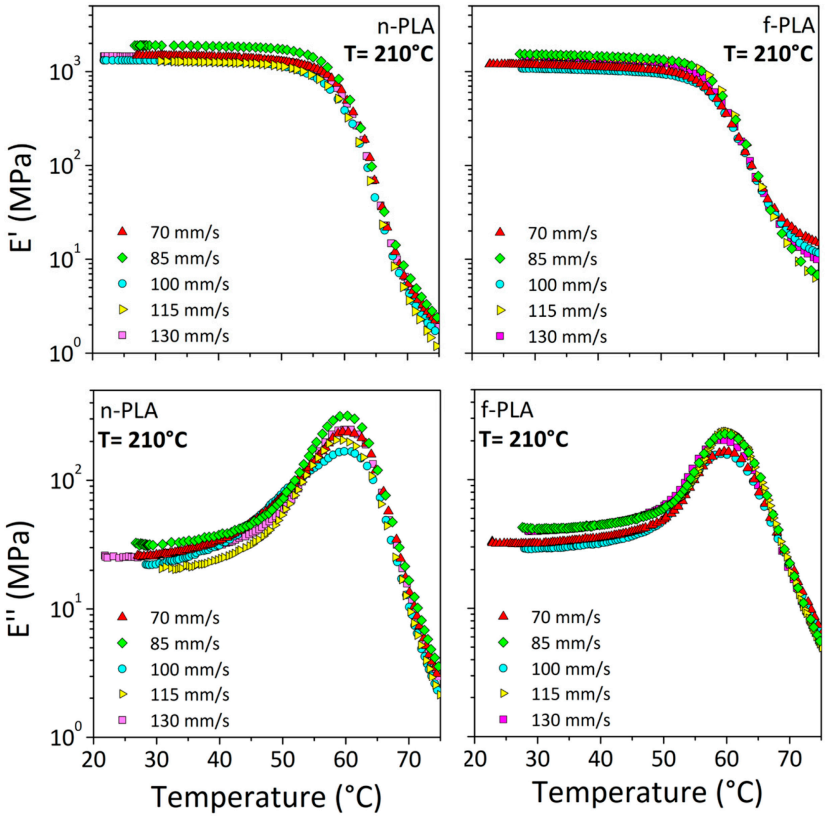


Figure 17. Representative curves of storage (E′) and loss (E″) moduli of 3D specimens made from neat–PLA (n-PLA) and composite (f-PLA) filaments printed at 210 ∘C and varied printing speeds [86].

Mazurchevici et al. [87] conducted DMA testing on biodegradable composites with wood fibers (WFs) and polyester reinforcement (BDP) on 20 wt.% concentrations. Both presented a positive influence on the dynamic thermal capabilities with a greater increase in the BDP composite. WF composites presented greater thermal stability observed by the increased TG, while BDP presented higher energy storage capabilities. The comparative results are presented in Figure 18.

Besides FDM printing, efforts were conducted with wood–resin composites through SLA AM. In contrast to the results of FMD printing, the developed composites presented a gradual decrease in the thermal properties and the energy storage capacity. The main reason behind this declining behavior was due to defects of the photopolymerization process caused by the existence of wood fibers. Sharp peaks on the loss modulus sweeps were flattened and less structural integrity was observed, confirming the unsuitability of wood-based byproducts with stereolithography AM [88]. An overall presentation of the main findings regarding the DMA response of wood–polymer composites is presented in Table 2.

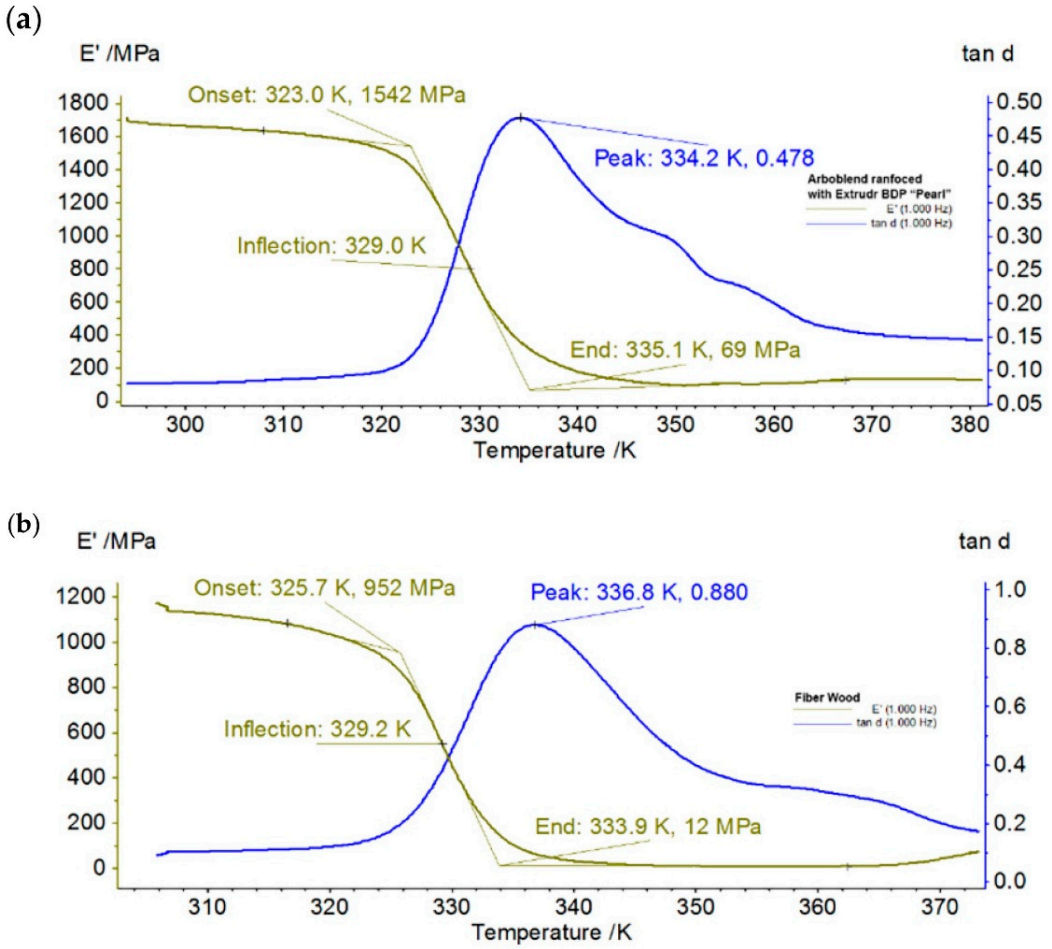


Figure 18. Thermal sweeps of E′ and E″; (**a**) PLA-BDP; (**b**) PLA-WF [87].

Table 2. Comparative results of the thermomechanical response extracted from DMA testing [85–88].

Composite	T_g (°C)	E′ (MPa)	tan δ	Main Findings
Wood Sawdust/PLA	Increase	Increase	Increase	25.7% energy storage increase, slightly increased T_g
Pine Fibers/PLA	Decrease	Increase	Decrease	Enhanced energy storage capabilities, slight decrease in T_g
Polyester/PLA	Decrease	Increase	Decrease	Higher strength, less damping
Wood Flour/Resin	Decrease	Unknown	Decrease	Poor compatibility, performance drop

3.4.2. Fatigue Response

Fatigue analysis in wood–polymer composites is vital for assessing the probability of stress concentration and material failure after repetitive loading cycles due to material defects occurring by the wood nature or the printing conditions. Wood flour insertion on the polymer matrix resulted in a significant 87.5% decrease in durability in cyclic loading with two different total stress applications (7.5–15 MPa). On the contrary, cyclic loading with steady maximum stress application resulted in the ability of the structure to tolerate over 1,000,000 loading cycles at an overall stress of around 9 MPa. The S–N curves recorded for the same composition are described by Basquin's relationship, with indicative values of the coefficient $\sigma_0 f \approx 295$–353 MPa and the fatigue exponent $b \approx -0.27$ to -0.32 [89,90]. A presentation of the crack propagation after cyclic loading is presented in Figure 19.

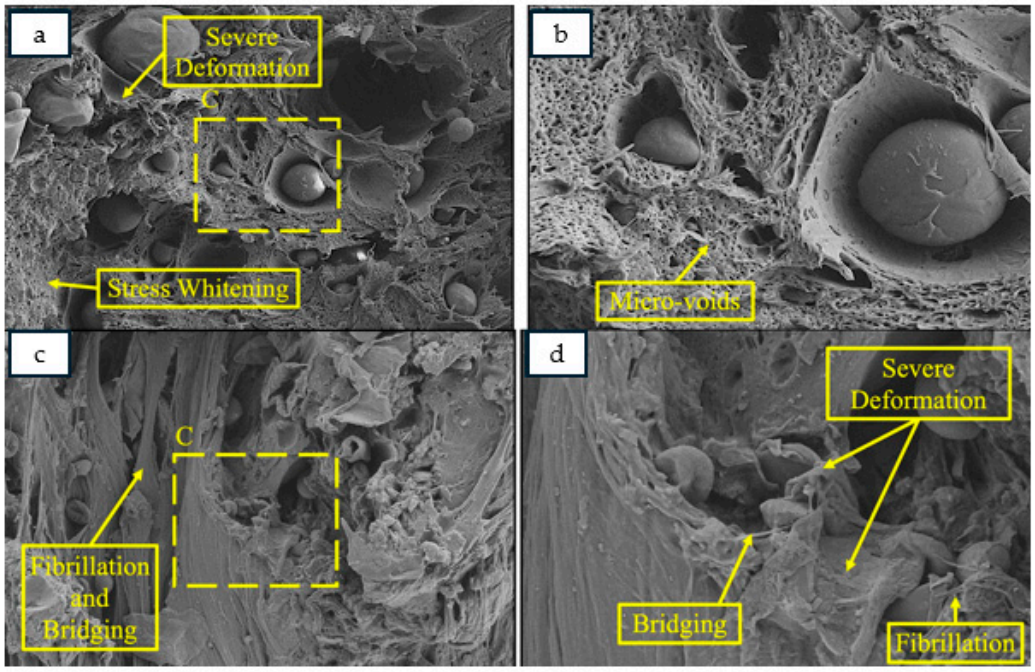


Figure 19. Morphological analysis in PLA-WF composites after cyclic loading; (**a**) 7.5 MPa; (**b**) 7.5 MPa; (**c**) 15 MPa; (**d**) 15 MPa [89].

The results of Figure 19 indicate that plastic deformation was observed around the wood particles, while at higher stresses (15 MPa), extensive microcrack zones and debonding appeared at the composite interface. In addition, internal voids and delamination phenomena in areas perpendicular to the printing direction suggest that the presence of filler (wood flour) enhances stress concentration and reduces interlaminar cohesion, making the material more prone to dynamic failure. Pine wood-reinforced polymers presented limited durability even at low to moderate stress applications. On the other hand, bamboo-based fibers show exceptional durability after 1000 loading cycles, even at stress closer to the yield point, highlighting the importance of the nature of the feedstock in the mechanical response. Bamboo fibers present a great strength-to-weight ratio and are deemed as a suitable natural alternative to high-strength conventional fibers (e.g., glass fibers) [91]. The Palmgren–Milner criterium was applied to estimate the life cycle duration of unknown wood composites. The relation between the composite fatigue and the number of cycles does not present linear correlation, therefore explaining the increased risk of crack propagation at unstable loads, even at stress regions lower than the material's yield or failure point. Overall, wood composites presented improved durability in a higher number of cycles under stable loading cycles or at low regions of stresses compared to the neat polymers, but in higher stresses, the durability was decreased due to the creation of stress concentration points at the interface between the fiber and matrix [92].

3.4.3. Impact Response

Impact testing is vital for further upcycling. The Charpy impact is mainly utilized for metals, while the Izod impact is used for ceramics. For polymeric materials, both testing techniques are utilized. Another efficient technique for the identification of energy absorption and the crashworthiness of a structure is drop impact testing. Wood flour sawdust in concentrations up to 10 wt.% proved to provide a slight increase in the energy capacity of the composite, regardless of the use of chemical compatibilizers, due to proper dispersion and reinforcement of the interfacial mechanical interlocking, while at concentrations higher than 20 wt.% the effect was negated due to known structural instability issues (porosity

and micro-voids) [93]. Wood leachate filler provided a positive influence on the impact resistance value with a numerical peak at 9 wt.% [94]. In temperatures way below 0 OC, brittle-like behavior was observed regardless of the filler's nature and a significant drop in the final impact resistance was also observed [93]. Overall, thermal treatment negatively influenced the mechanical response of the final structures during Charpy impact testing [95].

In Izod impact testing, a similar response was observed in concentrations up to 10 wt.% due to proper dispersion, and a numerical peak was again observed in this range. In higher concentrations, declining behavior was again observed due to unstable filler distribution [96]. Although the behavior was declining in terms of absolute impact resistance, composites with walnut shell fibers in high concentrations presented minimal deviations in the impact property's evaluation, meaning that adequate synthesis was conducted [97]. Overall, the dynamic evaluation of unknown acquired waste materials is a field with limited covered research and emerges as an important literature gap. Drop impact testing stands as the most accurate technique to calculate energy storage capacity under real-life conditions. Drop impact testing in lignocellulosic-filled wood–PLA composites indicated that the impact properties of the final structure can be greatly enhanced and even exceed the performance of the conventional PLA-CF (carbon fiber) filaments [98,99]. Those findings indicate that with proper treatment and by assuring the maximum printability of wood, byproducts can work as a sustainable alternative for the rapid manufacturing of functional products with high crashworthiness.

3.5. Life Cycle Assessment (LCA) Analysis

Life Cycle Assessment analysis is vital for the evaluation of the crucial environmental and financial sustainability factors for long-term efficiency in the manufacturing sector. The analysis considers every stage of the life cycle of a product and assesses the environmental impact throughout the life span. Cradle-to-gate LCA covers the stages from feedstock collection to the final product, while cradle-to-grave covers the whole life cycle up to the disposal of the product as waste. The gate-to-gate approach covers the manufacturing cycles without an evaluation of the feedstock collection and production process effects [100,101]. In the context of additive manufacturing using composite materials made from polymers and wood byproducts, the application of life cycle analysis is particularly important, as it assesses the environmental performance of these materials, which combine renewable raw materials and technologies with a low environmental footprint [102]. In cradle-to-grave LCA wood–polymer composites presented 30–35% less energy demands, 30–35% less carbon emissions, and 60–70% lower freshwater ecotoxicity and human toxicity [61]. In cradle-to-gate LCA, the insertion of wood flour from olive wood branches resulted in 10–20% less Global Warming Potential (GWP) and a narrow 10% decrease in the total energy demands and the overall carbon emissions. Another interesting factor is that the Photochemical Ozone Creation Potential (POCP) was also reduced by roughly 10% [14]. Overall, the existing LCA analyses indicated that there is room for the potential inclusion of wood byproducts as a viable long-term sustainable solution.

4. Large-Scale Applications

The utilization of wood byproducts through additive manufacturing constitutes an emerging and highly promising field, especially when considered for large-scale applications. The transition from the laboratory level to more practical and commercial applications is feasible to a degree, as it requires addressing certain challenges due to the nature of the material. However, there are studies showing that their use in larger-scale applications is feasible and viable. An excellent example of a large-scale application of wood composite

materials through additive manufacturing is the University of Maine's BioHome3D project. BioHome3D is the first home in the world printed entirely from bio-based materials (wood fibers) using the world's largest 3D printer. The printing was performed using the Fused Filament Fabrication (FFF) technique. The home, with an area of approximately 56 square meters, was designed to be a fully recyclable, eco-friendly, and sustainable housing solution. This project highlights the potential of additive manufacturing in utilizing wood byproducts to address social and environmental challenges [103]. Another example of an application is the development of Radicant panels, the panels constructed using the technique of viscous mixture extrusion with the aid of a cobot robotic arm. The printing process was carried out with the support of the Italian organization WASP (World's Advanced Saving Project), which specializes in developing innovative large-scale 3D printing solutions with a focus on sustainability. The use of the panels was primarily decorative and constituted a pilot approach [104]. Another approach is the utilization of binder jetting technology to produce objects from recycled wood powder from the furniture industry, combined with bio-resins to produce objects with complex geometry and detail, such as chairs, lighting fixtures, table surfaces, decorative panels, and custom wall coverings. These objects combine the esthetic of natural wood with the customization capabilities offered by 3D printing. The utilization of recycled raw materials presents positive effects on environmental sustainability. The natural texture and esthetic of the final structures manufactured with this method have proved that wood byproducts stand as a viable alternative to natural wood, in applications such as interior architecture and furniture design [105]. The above examples confirm that additive manufacturing with wood byproducts is not limited to a laboratory scale but can be extended to broader architectural and design fields. Despite the technical challenges, the flexibility offered by additive manufacturing, both in terms of printing methods and the adaptability of materials, creates new possibilities for the utilization of wood byproducts in large-scale applications. Further research, combined with industrial adoption, could transform these composites into competitive alternatives to conventional materials, thereby strengthening strategies for the circular economy and green growth in the construction sector.

5. Research Gaps and Solutions

Overall, wood byproducts and bioproducts stand out as promising sustainable alternatives to conventional non-recyclable materials. Although a certain degree of compatibility has been observed, it is also proven that in most cases, some sort of chemical modification is vital and positively influences the mechanical integrity of the final composite. A critical research gap exists in developing safer methods for improving interfacial bonding and neutralizing the difference between the hydrophilic wood byproducts and the mainly hydrophobic polymers. Enzymatic modification processes, plasma treatment, or the development of bio-based coupling agents could stand as a viable sustainable solution. In addition to intermolecular bonding, printing parameters are shown to be pivotal for the efficient development of composite structures. Kariz et al. [106] studied the effect of plasma treatment methods on the bonding of wood with the PLA matrix while Donate et al. [107] highlighted the ability of property modification with oxygen plasma treatment. Since differences on the thermal properties are evident and the acceptable range of key printing parameters in such cases tend to become narrowed down; the external optimization with Artificial Intelligence (AI) algorithms or the insertion of advanced analytical equations could help enhance the printing success rate for unknown weight fractions or filler materials. The combination of bio-based materials and AI optimization could heavily influence the manufacturing field. Miehe et al. [108] highlighted the probability of producing smart biomanufacturing devices as closed-loop systems that utilize biotechnology,

additive manufacturing, and AI optimization. Another important research gap seems to be the limited feedstocks that have already been utilized. Most studies investigated the use of pine wood, olive branches, and other lumber mill woods. The development of feedstock material from agricultural waste is an overlooked but important field. Polymers from packaging waste, natural fibers from various seeds or plants, and even biowaste from farming processes could be reacquired and processed into new feedstocks, new sources of energy, or for bio-fuel production. Dynamic testing and scalability investigation was also identified as an important research gap. Most studies tested the static response through conventional mechanical testing (e.g., tensile) but limited research was conducted on the assessment of the thermomechanical response, which is vital for numerous industrial or engineering applications, or the response in shock or dynamic loads. Impact, frequency, and thermal-dependent sweeps stand as an important research gap and an intermediate step to possible upcycling. Dynamic testing not only enhances the knowledge around the material properties but also provides a more complete overview of the possible applications, helping in more comprehensive and accurate Life Cycle Assessment analyses.

6. Conclusions

In the present study, an investigation of the manufacturability of wood byproducts and bioproducts via additive manufacturing for the development of sustainable feedstock materials and manufacturing systems was conducted. The literature analysis showed that the incorporation of wood fillers can improve certain properties, such as thermal stability and esthetics, but it is also accompanied by technical challenges related primarily to the homogeneity of the material and the cohesion between the printed layers. One of the key observations from the analysis was that when the filler was used without any modification, the performance of the composite materials was significantly reduced and, in many cases, inferior to that of the pure polymer or other composite materials. Specifically, poor dispersion of the filler in the matrix, weak adhesion between the phases, the appearance of pores, and poor mechanical strength were observed. Filler content within the polymer matrix was observed to greatly influence the final product, as percentages above 20% led to a further deterioration of homogeneity, increased brittleness, and the formation of agglomerates, which significantly reduced the mechanical performance of the material. It was also found that, in most cases, wood particles act as stress concentration points, intensifying failure phenomena and interlayer delamination. Conversely, in cases where appropriate chemical modifications were applied, an improvement in the overall performance of the composite materials was observed. Beyond chemical modification, the printing parameters were also observed to influence the final properties of the materials. The particle size of the filler, layer height, nozzle diameter, printing speed, layer orientation, and infill density significantly affect both printability and mechanical strength. Beech and pine exhibited high compatibility with the matrix material, as well as more stable thermal and mechanical behavior when their content was limited to 10–20%. Composites with pine fibers exhibited higher dynamic stiffness without negatively affecting the glass transition temperature (Tg), indicating good thermomechanical stability. The environmental assessment via LCA (Life Cycle Assessment) highlighted the superiority of wood-based composite materials over those made entirely from thermoplastics or other conventional materials (e.g., metal), particularly in terms of CO_2 emissions. Pine and oak byproducts combined good compatibility and technical performance at low content levels. Despite their limited mechanical properties, these composite materials can be used in a multitude of applications where high mechanical strength is not required, such as decorative objects, prototype models, and applications of an experimental or educational nature involving 3D printing. Further investigation of suitable compositions, modification techniques, and printing parameters is a necessary step

for the full utilization of these materials in functional additive manufacturing solutions that combine technological performance with environmental responsibility.

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