ADDITIVE MANUFACTURING OF WOOD-BASED MATERIALS FOR COMPOSITE APPLICATIONS

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Abstract

Wood-based material components including sawdust, wood flour, lignin, and cellulose nanofibers are being explored as functional additives and reinforcements in thermoplastic and thermosetting matrices used in additive manufacturing (AM) or 3D printing. The 3D printing processes most typically reported using wood-based materials include the extrusion-based fused filament fabrication, fused deposition modeling, large scale pellet-fed system, granular material bonding (selective laser sintering), and liquid deposition modeling. The rationale for using wood-based components in 3D printing include enhancing the material properties of the resulting printed parts such as increased mechanical properties, reduced dimensional instability, i.e. part warpage, improved aesthetics, providing a green alternative to carbon or glass filled polymer matrices as well as reducing material costs. This paper will provide an overview of wood-based material applications in 3D printing for composites with a state of the art review of current research activities around the world.

Background and Requirements

Additive manufacturing (AM) or 3D printing is among the most exciting advances in materials development over the past several decades [1]. The manufacturing space for 3D printing is quite broad with the wide variety of material types and forms that can be utilized including plastics, metals, cementitious components, gels, foams, etc. Common AM manufacturing processes include extrusion-based processing, liquid deposition modeling, granular material binding (selective laser sintering), bioprinting, and sheet lamination. Some of the advantages of 3D printing include it being a green manufacturing technique having a wide range of applications, requiring minimal supervision, can provide high component customizability and can be a lower cost production process for certain applications. The scale of 3D printing processes has historically focused on the bench scale where part build volumes are typically around 1 to 3 cubic feet, although more recently large scale AM processes have explored build volumes of 300 cubic feet and larger that covers the product space of automobile manufacture and construction applications [2, 3].

Consumers, industry, and governments are increasingly demanding products made from renewable and sustainable resources that are biodegradable, non-petroleum based, carbon neutral, and have low environmental, human health and safety risks. Utilization of renewable wood-based components in polymer composites for 3-D printing or AM addresses this challenge. The use of wood-based materials for 3D printing polymer composite formulations can provide improved material properties for a diverse variety of product applications [4-6].

Scope and Objectives

It is the purpose of this paper to review the current research activity in the use of wood-based materials in additive manufacturing with an emphasis on AM processes, wood component types and attributes, manufacturing challenges and processing issues, material properties, and product types.

Additive Manufacturing (3D Printing Process)

The generic 3D printing process consists of three process steps (Figure 1) including: 1) preparing a model of the part to be made using computer aided design (CAD), 2) separating the CAD model into “slices” using slicing software that allows the printer to process the model in three dimensional space, and 3) printing the slices sequentially using the software commands from the slicing software resulting in the production of the part. In this paper we will focus on the materials used to manufacture 3D objects.

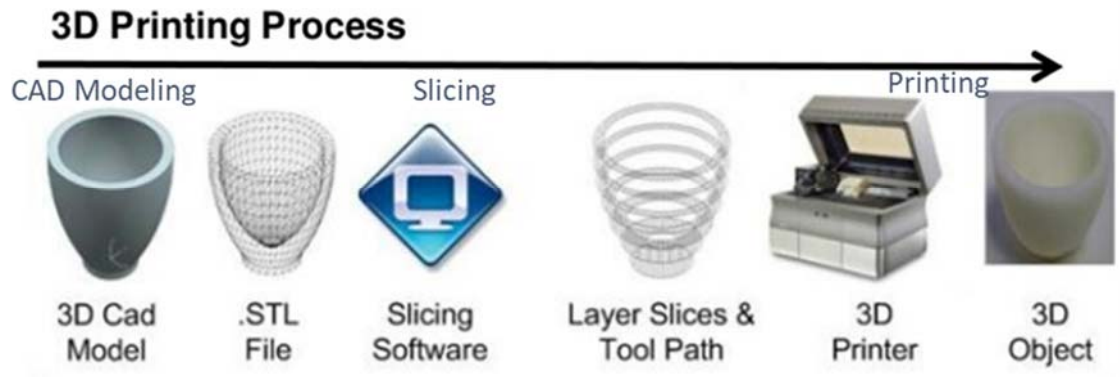

Figure 1. Process steps for 3D printing process https://www.slideshare.net/HorizonWatching/s12-3-d-printing-2014-horizonwatching-trend-summary-report-17apr2014.

The AM processes and materials utilizing wood-based materials are summarized in Table I. The AM processes typically used with wood-based materials include extrusion-based, granular material bonding, and liquid deposition modeling (Figure 2).

Figure 2. Extrusion-based, granular material bonding, and liquid deposition modeling of wood-based materials Reprinted from [18, 21] with permission from Springer.

The extrusion-based methods including fused deposition modeling, fused layer modeling, and fused filament fabrication utilize thermoplastic polymers in filament or pellet form as the primary feedstock and wood flour, spray or freeze-dried cellulose nanofibers, lignin and bamboo as the wood component [7-13]. Bioprinting which is also an extrusion-based 3D printing process uses aqueous suspensions of nanocellulose and other natural-derived biopolymers to produce hydrogel materials for wound dressing, drug delivery and scaffolding for regenerative medicine [14-16]. Granular material bonding processes include selective laser sintering (melt bonding) using combinations of thermoplastic and wood-based feed stocks [17], and water activation processes using combinations of wood feed stocks and inorganic binders (gypsum, sodium silicate and Portland cement) [18]. Liquid deposition modeling that is a hybrid extrusion-based manufacturing process uses combinations of wood feed stocks (saw dust or wood flour) combined with methyl cellulose and water or wood adhesive as binder [19-22]. A similar process utilizing a hand held stick dispenser uses “chop stick” wood material coated with wood adhesive to make large architectural structures [23]. Extrusion-based large scale additive manufacturing using wood flour and cellulose nanofiber feedstocks with thermoplastic polymers has been recently demonstrated to produce marine tooling molds [24] (Figure 3).

Table I. Additive Manufacturing processes utilizing wood-based materials.

Additive Manufacturing Process	Primary Feedstock	Wood Component(s)	Other Additives
Extrusion-Based Fused Deposition Modeling Fused Layer Modeling Fused Filament Fabrication Bioprinting	Thermoplastics (PLA, ABS, PP, nylon, polyurethane) Hydrogel Material (Collagen, carrageen) or nanocellulose in aqueous suspension	Wood flour, spray or freeze-dried cellulose nanofibers, microcrystalline cellulose, lignin, bamboo	Pigments, coupling agents, lubricants, glass, carbon
Granular Material Binding Selective Laser Sintering Water Activation	Thermoplastics, Cementitious materials (Gypsum, Sodium silicate, Portland Cement)	Wood flour, Wood chips, lignin	Water
Liquid Deposition Modeling Stick Dispenser	Thermoplastic emulsion Resin	Sawdust, Wood Flour, methyl cellulose Chopsticks	Water

Wood species, particle sizes, polymer types, and loading levels commonly being explored in 3D printing composite formulations are listed in Table II. Wood species being examined include both softwoods and hardwoods as well as recycled wood material from wood composites. Depending on the particular AM process, wood particle size varies from 14 microns up to 2000 microns. Typical screen sizes to segregate wood particles are around 60 to 100 mesh. Common polymer types examined include poly-lactic acid (PLA), polypropylene (PP), styrene maleic anhydride (SMA) copolymer, polyvinyl acetate (PVAc), urea-formaldehyde (UF), and carbohydrates (starch and methyl cellulose). As mentioned earlier, cementitious inorganics like gypsum and Portland cement have also been examined. Some researchers have reported using commercially available wood-filled filaments with brand names such as ColorFabb Woodfill (blend

of PLA and PHA and 15 wt.% wood fiber), Laywood 40% wood fiber and EasyWood Coconut Form Futura with 40 wt.% coconut fiber [26]. Unfortunately, in some instances researchers do not specify wood species or particle size in their publications making it challenging to compare the effects of these factors on the resulting material properties among studies.

Table II. Wood species, particle sizes, polymer types, and loading levels commonly used in 3D printing composite formulations.

Wood Species	Particle Mesh Size	Particle size microns	Polymer	Wood Loading Level wt.%	Reference
Osage Orange (*Maclura pomifera*)					
Paulownia (*Paulownia tomentosa*)	230	63	PLA	25	[7] Tisserat et al. 2015
Pine (*Pinus* sp.)	100	150	PP, SMA, PLA	0 to 30	[24] Gardner et al. 2018
Beech (*Fagus* sp.)	120 and 400	40 and 120	Starch	66	[22] Gardan et al. 2016
Beech (*Fagus sylvatica*)	~60	237	PLA	0 to 50	[25] Kariz et al. 2018
Spruce (*Picea* sp.)	~ 10 and 20	800 to 2000	Gypsum, Sodium silicate, cement	33	[18] Henke and Treml 2013
Beech (*Fagus sylvatica*)	~60	237	PVAc and UF	75 to 87.5	[19] Kariz et al. 2016
Aspen (*Populus* sp)	~635	14	PLA	5	[9] Tao et al. 2017
Beech(*Fagus sylvatica*)	~ 40 and 60	250 to 400	Methyl cellulose	84.5 to 89	[21] Rosenthal et al. 2018
MDF Furniture Waste	~200	80	PLA	10 to 40 (vol. fraction)	[26] Pringle et al. 2018
European Softwood	~200	75	UF	13	[20] Pitt et al. 2017

Figure 3. Large scale 3D printed boat roof tooling mold made from 20 wt.% wood flour and 1 wt.% CNF in a PLA matrix [24].

Wood components (cellulose and lignin) used in 3D printing applications are listed in Table III. Cellulose has been investigated at both the microscale including microcrystalline, microfibrillated cellulose, and microfibrillated wood and the nanoscale including cellulose nanofibrils and cellulose nanocrystals. Cellulose is typically melt mixed or extrusion compounded with thermoplastic polymers like PLA and PP or added as the main component in aqueous suspension for bioprinting. Loading levels for polymer addition are typically below 10 wt.% and in some instances surface modification by silane or titanate treatment has been done. More details on the use of cellulosics in bioprinting can be found in recent review articles [14-16]. Lignin preparations including organosolv and Kraft have been explored as additives for melt compounded thermoplastics including ABS, HIPS, Nylon 12 and PLA. Various loading levels of lignin addition from 5 to 60 wt.% have been studied.

Table III. Wood components (cellulose and lignin), delivery media, polymer types, additives and loading levels for 3D printing.

Wood Component	Delivery Process/ Medium	Particle Size	Polymer Type	Loading Level (wt.%)	Treatment	Reference
Microfibrillated cellulose	Extrusion	2 mm	PLA	1		[29]
Microfibrillated wood	Extrusion	2 mm	PLA	1		
Microcrystalline cellulose	Melt mix	not specified	PP	10	Silane Treatment	[28]
Microcrystalline cellulose	Melt extrusion	not specified	PLA	1, 3, 5	Titanate Treatment	[8]
Cellulose Nanofibrils	Water	Nanoscale	Neat or hydrogels	Varied	Crosslinkers	[14][16]
Cellulose Nanocrystals	Water	Nanoscale	Neat or hydrogels	Varied	Crosslinkers	
Cellulose Nanocrystals	Water	Nanoscale	None	10, 20	NaCl	[30]
Organosolv Lignin	Melt mix	not specified	ABS	20, 30 40	Carbon fiber	[12]
Organosolv Lignin Softwood Kraft	Melt mix		ABS, HIPS, Nylon 12	40, 50, 60		[13]
Pine Kraft Lignin	Melt mix		PLA	5, 10, 15, 20		[11]

Compounding and Processing

Many of the processing steps used to compound wood and wood components into plastics for 3D printing are analogous to the processing steps commonly used in wood plastic composites for profile extrusion, injection and/or compression molding [31, 32]. The wood or wood component furnish should be dried below 1 % moisture content to facilitate proper mixing with the plastic. The wood components are typically mixed with the plastics and/or other additives via batch mixing in thermokinetic mixers or continuously using twin screw compounding extruders. Depending on component loading levels required in the thermoplastic, the creation of a masterbatch at higher wood component loading level followed by dilution to lower filler loading levels can be beneficial to improved distribution and dispersion of the filler in the polymer matrix [34]. The plastics examined thus far for 3D printing typically have melt temperatures below 225 degrees C which is below the thermal decomposition temperature of wood and wood components. For bioprinting, the cellulose is processed in water with or without hydrogel polymers and other additives as appropriate for the particular application [14-16].

Filament versus Pellet Fed 3D Printers

Most bench top extrusion-based 3D printers typically use filament fed extruders to produce parts. More recently pellet fed extruders are becoming common for bench top 3D printers and pellet fed extruders are standard for large scale extrusion-based 3D printing. Filament fed extruders have specific requirements for filament diameter and dimensional tolerances such that improper filament size, presence of voids or shape (non-circularity) can be problematic to properly feed the printer and produce adequate parts. Adding wood-based components to plastics followed by filament manufacture can also be challenging in terms of maintaining dimensional tolerances and filament quality. An example of filament production problems is shown in Figure 4. Filament voids can be produced during the extrusion process and can be inconsistent throughout the filament. Non-circularity creates filaments with an aspect ratio that negatively effects the quality of part production [33].

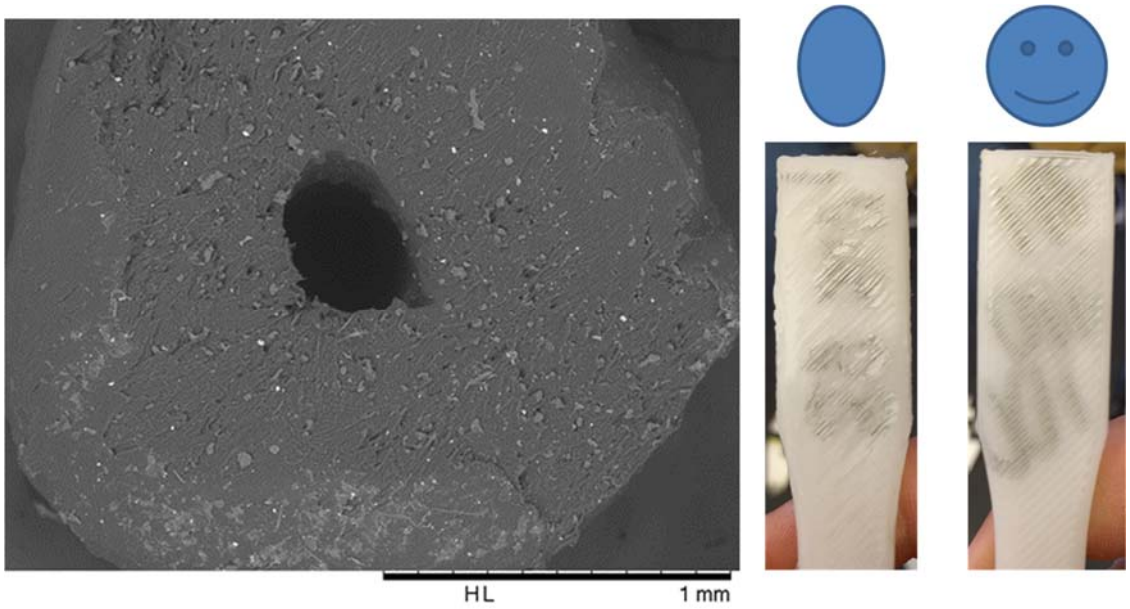

Figure 4. Challenges with filament production including voids and non-circularity [33].

Higher loading levels of wood-based components can result in problems with filament brittleness and breakage as well as issues with even printing the wood-filled filament [7]. The particle size and loading level of the wood component being added to the plastic can also affect the quality of the filament in terms of poor printing quality and printer nozzle plugging [26]. Issues with 3D printer nozzle plugging can be partly overcome by using nozzles with the proper size opening. Pellet fed extruders circumvent some of the issues related to producing quality filament with wood-based thermoplastics. However, wood-filled pellets should be dried prior to printing to reduce possible problems with absorbed moisture during the printing process [26]. Making test prints with wood-filled thermoplastics is a good way to screen various formulations for printing quality in terms of interlayer bonding and print quality based on rheological behavior [24].

Material Properties

The material properties of wood component containing 3D printed parts are quite varied based on the different AM processes and material combinations being examined. A synopsis of mechanical and physical property values for 3D printed parts appearing in the literature are listed in Table IV.

Table IV. Mechanical and physical properties of 3D printer wood-filled parts.

3D Printing Process	Tensile MOR (MPa)	Tensile MOE (GPa)	Bending MOR (MPa)	Bending MOE (GPa)	Density (g/cm3)	Reference
FDM Wood (%) in PLA						
0	55	3.27			0.63	[25]
10	57	3.63			0.52	
20	49	3.94			0.52	
30	48	3.84			0.48	
40	42	3.86			0.48	
50	30	3			0.48	
LSAM						[24]
Wood (19%) CNF 1% in PLA						
Longitudinal	54.5	5.14				
Transverse	32.9	4.04				
FDM						[29]
1% Fiber in PLA						
PLA-MFLC			91	3.3		
PLA-MFW			103	3.6		
Liquid Deposition Modeling (LDM)						
Beech and Methyl Cellulose			2.3	0.285	0.33	[21]
			5.1	0.547	0.44	
			7.4	0.733	0.48	
LDM (Wood and Adhesive)						
17.5% PVAc			3	0.013		[19]
20% PVAc			5	0.045		
15% UF			19	1.93		
17.5% UF			18	2		
LDM (Wood and Adhesive)						[20]
87% UF			57	6.8	1.22	
LDM (wood flour starch filament)	5.45	0.6				[22]
Granular Material Binding						
0.15 Wood/Cement			0.5		0.7	[18]
			0.95		0.8	

Most researchers either report flexural bending strength (modulus of rupture (MOR)) and stiffness (modulus of elasticity (MOE)) or tensile MOR and MOE. In some instances, densities are reported. Higher strength and stiffness values have been reported for the extrusion-based 3D printing processes like FDM and LSAM using wood-filled PLA where tensile MOR ranges from 30 to 57 MPa and MOE ranges from 3 to 5.1 GPa. The liquid deposition modeling and granular material binding mechanical properties are an order of magnitude lower in MOR and MOE compared to the extrusion-based processes. Reported densities range from about 0.33 g/cm^3 to 0.8 g/cm^3. The 3D printing process part with the highest reported properties was for an 87% urea-formaldehyde (UF) wood-filled part with a flexural MOR of 57 MPa and MOE of 6.8 as well as a density of 1.22 g/cm^3.

Challenges and Opportunities in 3D printing with wood-filled plastics

Three thermoplastics that are used extensively in the wood plastic composites (WPCs) industry are polypropylene, polyethylene and polyvinyl chloride [32]. However, these three plastics are not commercially available for 3D printing applications. The primary reason for the unavailability of such materials in melt extrusion-based 3D printing is their dimensional instability, usually warping caused by rapid crystallization during the printing process as shown in Figure 5 [34]. A simple way to address the warping issue in 3D printing is to avoid using high-crystallinity plastics [36]. Polypropylene copolymers, including PP block copolymer [35, 37], PP random copolymer [38] have been successfully printed with less shrinkage and warping compared to isotactic PP (iPP). An example is shown in Figure 5. Likewise, it is envisioned that syndiotactic or atactic PP could also be explored in extrusion-based 3D printing, though no results have yet been reported in the literature. Low density polyethylene (LDPE) with a lower crystallinity (28%) was added to high density polyethylene (HDPE) (62%) to improve the dimensional accuracy of 3D printed parts [40].

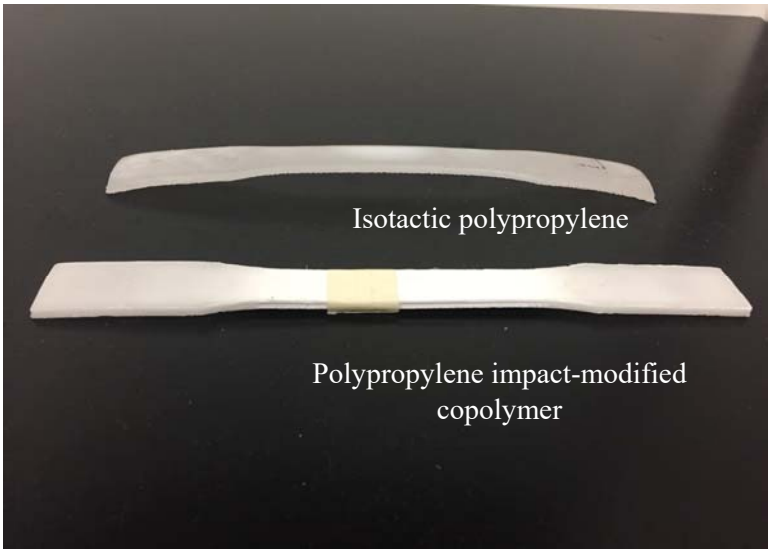

Figure 5. Comparison of the dimensional stability of 3D printed PP and PP copolymer.

It is noteworthy that most of the thermoplastic products in the wider commercial plastics marketplace are made of materials with high crystallinity which often results in better mechanical properties [39]. To improve the processing of such materials and retain or enhance mechanical properties, nanofibers have been investigated for 3D printing using iPP [34]. Addition of fillers in

a semi-crystalline polymer can affect its crystallization behavior in different ways at different stages of processing. During the nucleation process, most fillers can act as nucleation agents that provide additional sites for spherultic growth. During the crystallization growth stage, if the gaps among fibers are extremely narrow, crystal growth can be limited within the small spaces as shown in Figure 6 [34]. Overall, nanofibers can act as crystallization retardants, instead of nucleating agents for semi-crystalline polymers [41]. The fiber loading level of nanofibers should be carefully controlled to regulate the crystallization behavior of the polymers during processing.

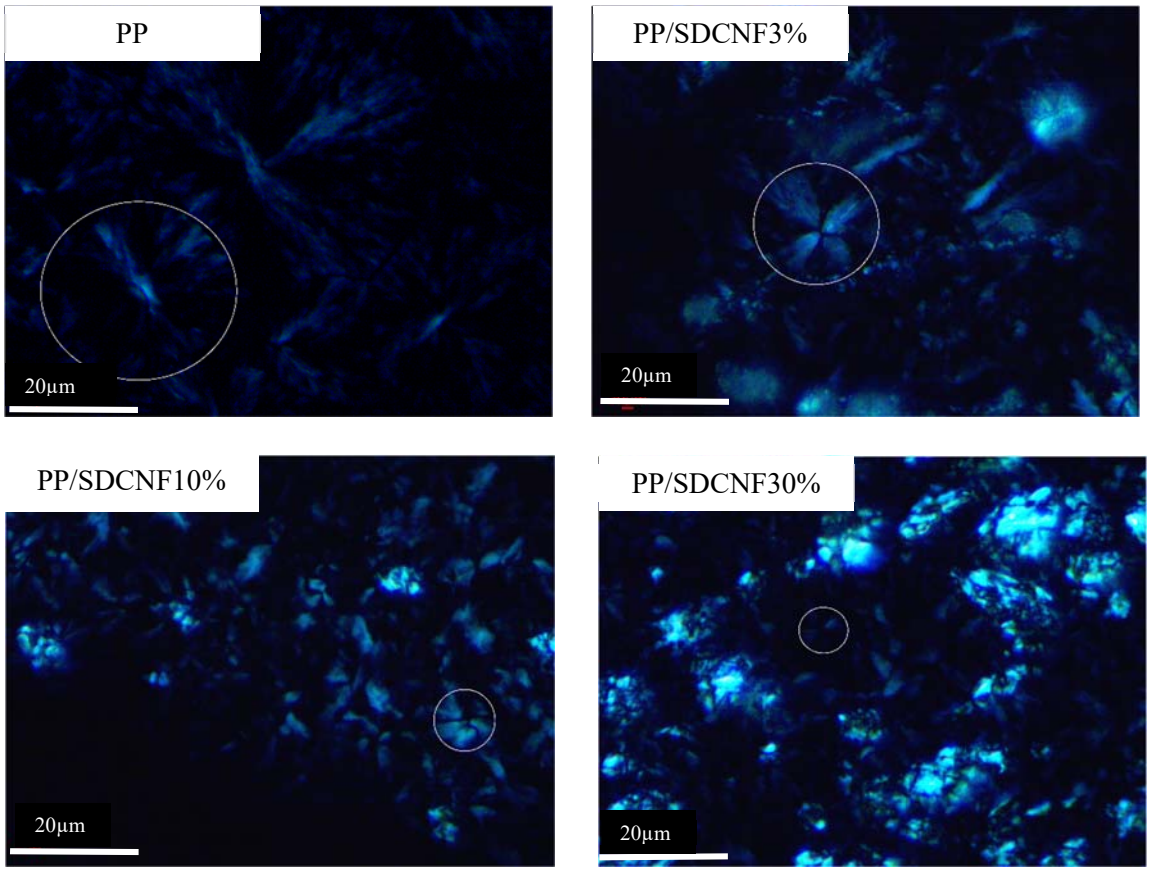

Figure 6. Effect of spray-dried cellulose nanofibrils loading level on the crystal size of iPP Reproduced with permission from [34].

There are differing opinions over whether the addition of fibers degrades or enhances the interlayer bond strength of 3D printed parts (Figure 7). On one hand, fibers remaining at the interface between two layers can reduce the area for polymer chains to diffuse from one side to the other [43]. Because the degree of polymer diffusion is proportional to interlayer bond strength, such strength can be reduced by the presence of fibers. Alternatively, if the interaction between the fibers and polymer matrix is strong, e.g. with the application of coupling agents, and stresses can be transferred from the polymer chains to the fibers, the interlayer bond strength can be increased. In 3D printing, most fibers will align in the printing direction attributable to the extrusion process. The strength properties of a printed part along the fiber's transverse direction is often weaker than in the longitudinal direction. To improve the interlayer bond strength, fibers will need enhanced transverse strength. Additionally, a large degree of crystallization with high crystallinity was found to degrade the interlayer bonding strength of semi-crystalline polymers [48]. When

each laid-down polymer strand cooled it experienced shrinkage, and it tended to pull itself away from adjacent strands, creating extra separating forces at the interface. The proper addition level of nanofibers with adequate transverse integrity can reduce the matrix crystallinity and provide reinforcement to polymer chains near the interfacial area, and therefore, potentially increase the interlayer bond strength.

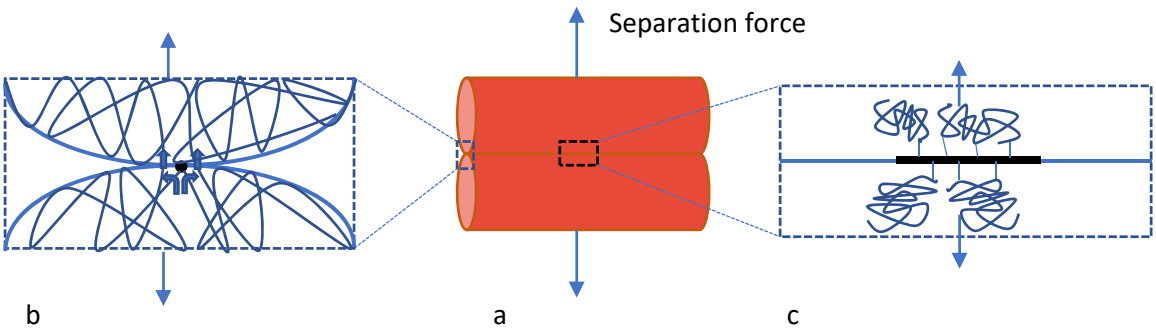

Figure 7. Schemes of a) two adjacent laid-down strands under separation force, b) a fiber (black dot) at interface acting as a hindrance to molecular diffusion between polymer chains (random strings) at both sides and c) a fiber (black string) with good transverse strength and treated with coupling agent (short straight strings) acting as a strengthening agent.

There are several disadvantages in adding fibers to polymers in 3D printing. For filament-based printers, the filament can remain flexible enough for spooling only to a certain fiber content for a given polymer. For example, a 20 wt.% fiber content was found to prevent a wood-iPP filament from properly spooling because of filament brittleness [33]. Another issue related to fiber addition is the formation of porous structures inside printed beads/parts [35]. Possible causes contributing to the creation of pores include fibers incompatible with matrix polymers and nozzle clogging attributable to insufficient extrusion force [35]. Moreover, the viscosity of the fiber-filled polymer composites are increased compared to neat polymers, thus composite filaments require a larger processing force. Filament-based melt-extrusion 3D printers typically do not utilize screws in their heating barrels. The flow of melts primarily relies on the gravity of materials and the force from feeding gears trying to push the filament into the barrel [44]. If the viscosity of a polymer (i.e. PP and PE) is not sensitive to temperature change, the addition of fibers in the polymer would experience flow difficulties and deposition issues. A pellet fed extruder may be a potential solution to all the issues raised above when printing a thermoplastic composite with high fiber content. A pellet fed extruder possesses a screw, creating shear flow which can lower the viscosity of the composites to the level of pure polymers at high shear rates [10, 45, 46]. In this instance, any problems occurring with high fiber loading can be aesthetic because high fiber contents in a polymer also contribute to rough surface quality during printing. For instance, the surface of a 30 wt.% harakeke-filled PP print was coarse and hairy [47] and may require an extra surface finishing process for part production.

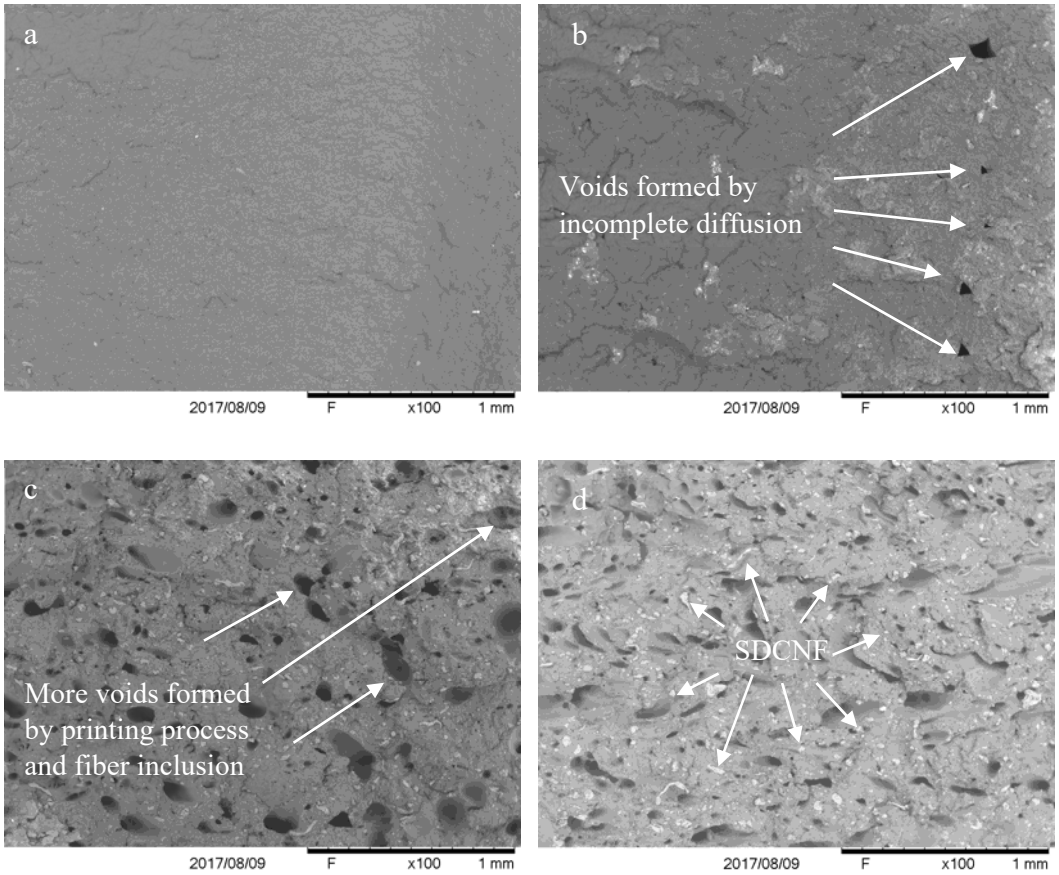

Figure 8. SEM graphs of injection molded PP copolymer (a), 3D printed PP copolymer (b), 3D printed PP copolymer/7.5wt.% spray-dried cellulose nanofibrils composites (c) and 3D printed PP copolymer/15 wt.% spray-dried cellulose nanofibrils composites (d). White dots in the micrographs are spray-dried cellulose nanofibrils Reproduced with permission from [35].

Pores typically exist at the junctions of laid-down strands because of incomplete interfacial diffusion in melt-extrusion 3D printing (Figure 8). In most cases, 3D printed parts possess lower density values than injection molded parts, which produces lower mechanical properties in 3D printer parts [48]. However, when the weight of a part is an equally important attribute to its mechanical properties (e.g. in light-weight applications), 3D printed parts can exceed injection molded parts in performance [48]. Because the addition of natural fibers generates extra pores in 3D printed parts, densities of 3D printed parts are lower than their injection molded counterparts [33]. Again, the decrease in density may not be a disadvantage for 3D printed fiber filled polymer composites when appropriate applications are identified. For example, a heat deflection temperature of 104 °C was found in a 3D printed 15 wt.% spray-dried CNF filled PP impact-modified copolymer part compared to 84 °C for the injection molded PP copolymer [35].

Summary and Conclusions

Additive manufacturing using wood-based components has been explored across the manufacturing space of extrusion-based processing, granular bonding and liquid deposition molding. Various combinations of wood and polymer and/or inorganic feedstocks are being

examined for various applications. For wood feedstocks, various species, particle sizes from micron to millimeter length scale and loading levels within a matrix are being evaluated. Polymer components from wood being explored in additive manufacturing include cellulose and lignin especially on the nanoscale. The mechanical and physical properties of printed parts can, in certain instances, approach the material property space of conventional wood composites such as particle board, fiber board and wood thermoplastic composites. Challenges in additive manufacturing with wood-based materials include processing issues during extrusion and part production, especially regarding part dimensional stability and material brittleness depending on wood component loading level as well as impacts on polymer crystallization behavior during processing. Opportunities exist for producing lighter weight and lower cost composite parts for applications in mold tooling and automobile parts.

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