3D printing of biofiber-reinforced composites and their mechanical properties: a review

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Abstract

Purpose – This paper aims to summarize the up-to-date research performed on combinations of various biofibers and resin systems used in different three-dimensional (3D) printing technologies, including powder-based, material extrusion, solid-sheet and liquid-based systems. Detailed information about each process, including materials used and process design, are described, with the resultant products' mechanical properties compared with those of 3D-printed parts produced from pure resin or different material combinations. In most processes introduced in this paper, biofibers are beneficial in improving the mechanical properties of 3D-printed parts and the biodegradability of the parts made using these green materials is also greatly improved. However, research on 3D printing of biofiber-reinforced composites is still far from complete, and there are still many further studies and research areas that could be explored in the future.

Design/methodology/approach – The paper starts with an overview of the current scenario of the composite manufacturing industry and then the problems of advanced composite materials are pointed out, followed by an introduction of biocomposites. The main body of the paper covers literature reviews of recently emerged 3D printing technologies that were applied to biofiber-reinforced composite materials. This part is classified into subsections based on the form of the starting materials used in the 3D printing process. A comprehensive conclusion is drawn at the end of the paper summarizing the findings by the authors.

Findings – Most of the biofiber-reinforced 3D-printed products exhibited improved mechanical properties than products printed using pure resin, indicating that biofibers are good replacements for synthetic ones. However, synthetic fibers are far from being completely replaced by biofibers due to several of their disadvantages including higher moisture absorbance, lower thermal stability and mechanical properties. Many studies are being performed to solve these problems, yet there are still some 3D printing technologies in which research concerning biofiber-reinforced composite parts is quite limited. This paper unveils potential research directions that would further develop 3D printing in a sustainable manner.

Originality/value — This paper is a summary of attempts to use biofibers as reinforcements together with different resin systems as the starting material for 3D printing processes, and most of the currently available 3D printing techniques are included herein. All of these attempts are solutions to some principal problems with current 3D printing processes such as the limit in the variety of materials and the poor mechanical performance of 3D printed parts. Various types of biofibers are involved in these studies. This paper unveils potential research directions that would further widen the use of biofibers in 3D printing in a sustainable manner.

Keywords Mechanical properties, Polymer matrix composites (PMCs), 3D printing, Biofiber polymer composites (BFPCs)

Paper type Literature review

1. Introduction

Advanced composites are prevalent engineering materials due to their desirable mechanical properties, including light weight coupled with high stiffness, longitudinal strength and flexural performance. They also have broad, proven applications in aerospace, transportation vehicles, constructions, sports equipment sectors and medical applications (Brylawski and Lovins, 1998). However, waste produced from non-biodegradable polymer matrices during the disposal of these composites has caused many environmental problems around the globe such as water and soil pollution, agricultural land

occupation, live animal and plant toxication. As these polymers are derived from nonrenewable resources such as crude oil, mass production of these non-environmental friendly materials is leading to the declining reserves of fossil fuels and increasing of carbon emissions (Thompson et al., 2009). Moreover, waste polymer matrix composites are extremely difficult to recycle due to their complex structures and the limited time (2-3 times) they can be recycled due to quality downgrading (National Geographic, 2019). The only economical end-of-life options for this waste are burning for energy or disposal in landfills and both options incur a detrimental cost to the environment. The reinforcing fibers bound by the polymer matrix are also very difficult to reclaim or recycle due to difficulties in matrix

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removal. As much as one million pounds of cured and uncured carbon fiber (CF) pre-preg waste is generated by both Boeing and Airbus individually each year from 787 and A350 XWB production, respectively (Milberg, 2017). If the entire supply chain for these planes is included, the total amount of waste is closer to four million pounds per year; furthermore, the automotive industry is poised to consume (and dispose of) increasingly more CF (Sloan, 2016). The increase in global awareness regarding environmental issues, the enforcement of new environmental regulations and the attempts in avoiding unsustainable consumption of petroleum have created interest in the use of environmentally friendly materials (Mohammed et al., 2015). Biofibers are considered one type of such material that has good properties compared to synthetic fibers (May-Pat et al., 2013). The interest in biocomposites in terms of industrial applications (e.g. transportation vehicles, aerospace applications, military applications, civil construction, sporting goods and product packaging, etc.) and fundamental research because of their great benefits (e.g. renewable and recyclable, low-cost and biodegradable, etc.) is rapidly growing (Mohammed et al., 2015). Biocomposites are a type of novel material formed by a matrix (resin) and a biofiber reinforcement (Wikipedia, 2019). The matrix phase can be polymers derived from renewable and/or nonrenewable resources (Wei and McDonald, 2016).

A recent study by Mohammed et al. (2015) found out that the worldwide biofiber-reinforced composites industry reached \$2.1bn in 2010, and as indicated by evaluations, the biofiber polymer composites (BFPCs) industry is estimated to grow 10.93% worldwide in the next 5 years (2019-2024), with North America being the largest market and the Asia Pacific the fastest growing market (Mordor Intelligence, 2019). According to Faruk et al. (2012), the most common and commercially available biofibers in the world are bamboo, sugarcane bagasse, jute, kenaf, flax, grass, sisal, hemp, coir, ramie and abaca. The mechanical properties of BFPCs are affected by the fibers' orientations (Shalwan and Yousif, 2013), strength (Shinoj et al., 2011), physical properties (Benezet et al., 2012) and the interfacial bonding properties (Kakroodi et al., 2014). However, as long as there are strong base structures of BFPCs, the matrix can be easily improved and strengthened (Srinivasan et al., 2014). Applications of BFPCs are growing extremely fast in various engineering fields such as electrical and electronic applications, aerospace industries, sports and recreational equipment, transportation vehicles, machinery and office supplies (Mohammed et al., 2015). Because of their low specific weight and production cost, good strength and surface finish, biodegradability and acceptable mechanical properties, with abundant and renewable sources, the application of biofibers in polymer composites is being widespread (Shalwan and Yousif, 2013; Shinoj et al., 2010). On the other hand, the physical disadvantages of BFPCs include moisture absorption, restricted processing temperature and varying product quality, and these disadvantages significantly limit their performance (Gallo et al., 2013). Traditional techniques used to manufacture plastics or polymer composite materials are mainly implemented in the production of biocomposites (Wikipedia, 2019). Techniques used to deal with continuous biofibers include machine press, filament winding, pultrusion, compression molding, resin transfer molding or sheet molding

compounds (all of which either involve many manual operations or deal with a single or a few fiber strands, leading to excessive lead time). For instance, Misri et al. (2016) measured split-disk properties of kenaf yarn fiber-reinforced unsaturated polyester composites using filament winding method; Jiang et al. (2019), Jiang et al. (2017) and Jiang et al. (2020) reported using jute, flax and cellulose textiles as part facial reinforcements in the manufacturing of fungal mycelium-based sandwich biocomposites with vacuum-assisted resin transfer molding. Techniques used to address discontinuous biofibers include both extrusion and injection molding. To name a few, Migneault et al. (2008) made wood-plastic composites (WPC) with three length distributions through extrusion processes and concluded that mechanical properties of such biocomposites improved with increasing fiber length, yet their performance decreased in water immersion tests; Sanschagrin et al. (1988) studied strength and rigidity of injection-molded WPC made with three chemithermomechanical pulps aspen fibers having length to diameter (L/D) ratios ranging from 5 to 25 and found both of these two parameters increased with increasing L/Dratio. Besides these examples mentioned above, 3D printing of composite materials has been used in industrial and sociocultural sectors such as manufacturing, medication and military, facilitating 3D printing to become a successful commercial technology (Taufik and Jain, 2016) on a rapidly increasing basis. To address recent research and progresses in biocomposite 3D printing technologies, a comprehensive review is performed in this paper, together with potential research directions and areas in the near future identified.

2. Three-dimensional printing technologies for biofiber polymer composites

Current three-dimensional (3D) printing technologies can be classified in various ways. The classification system used in this paper is based on the form of the starting material in the process:

- powder-based;
- material extrusion;
- · liquid-based; and
- solid-sheet systems (Groover, 2016).

2.1 Powder-based systems

A common feature of powder-based 3D printing systems is that they start with materials that are in the form of powder. Powder-based technologies are generally not ideal for creating fiber-reinforced composites, as making a smooth layer of the powder-fiber mixture is not an easy task (Guo and Leu, 2013). It is the goal of many studies to mix the composite powder homogeneously (Goh et al., 2018) to improve the 3D printed parts' resolution and surface finish. Methods used for conventional advance reinforcing fibers in achieving this goal include using either mechanical (Chung and Das, 2006; Hon and Gill, 2003) and melt mixing (Goodridge et al., 2011) or by coating the fibers using dissolution-precipitation (Yan et al., 2008, 2009) and surfactant-facilitated latex (Yuan et al., 2016), while few of them have been tested with biofibers. Therefore, the types of biofibers used in such systems are very limited at present, with all of them adopting the selective laser sintering (SLS) process. SLS was developed by Dr Carl Deckard at the

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University of Texas at Austin in the mid-1980s. It uses a moving laser beam to fuse the starting material in powder form in areas corresponding to a computer-aided design model one layer at a time to build a solid part (Groover, 2016). SLS involves numerous cycles of rapid melting, cooling, followed by solidification (Song and Koenig, 1997; Das et al., 1998) and uses a 25-50 W YAG or CO₂ laser to sinter the fine powder material on a powder bed (Bourell et al., 1992; Schueren and Kruth, 1994). The loose powder material is preheated to right below their melting point to facilitate bonding and reduce distortion of the finished product, while the preheating also reduces the power required by the laser (Groover, 2016). In areas not sintered by the laser beam, the powder remains loose but supports the solidified regions of the part (Groover, 2016). These powders can be separated and removed easily from the completed part after the fabrication is finished. SLS typically has part accuracy and layer thickness of 0.1 mm (Watson, 2014). One advantage of SLS is its ability to reduce the manufacturing time and cost of production for metal components (Wohlers, 2015), together with its flexibility and potential to produce complex geometries (Kruth et al., 2007).

One of the most commonly used loose powders in biocomposite SLS is wood powder, which is also known as wood flour. Xin et al. (2009) reported using 80-100 μ m wood powder from aspen trees with a 120-180 μ m hot-melt adhesive powder and 100-180 µm polypropylene (PP) powder for 3D printing biocomposite parts via SLS. A mixture of PP and hotmelt adhesive with a ratio of 1:2.5 was used as the base material with no combining agent (Xin et al., 2009). The contents of wood powder varied from 10% to 40% with increments of 10%. The effect of varying the wood powder content on the mechanical properties of the 3D-printed biocomposite was investigated. The authors found that both the tensile and flexural strengths were negatively affected by increasing the wood powder content, which is due to the poor interfacial bonding between the wood and the plastic. However, both the tensile and flexural moduli of the biocomposites increased gradually as the wood powder increased (Table 1), indicating the reinforcing effects of the biofiber.

Guo et al. (2011) used alkalized wood flour to reduce the hydrophilic nature of wood fiber and improve its wettability. Copolyester (Co-PES) was used as the hot-melt adhesive powder in the experiment, and someviscosity reducer (graphite, calcium carbonate, white carbon black, talcum or glass powder) was also added into the powder mix to make it easier for the levering roller to spread the powder evenly on the printing bed (Guo et al., 2011). The wood plastic composite (WPC) was formed through the SLS process using a powder mixture of alkalized wood flour and Co-PES hot-melt adhesive

Table 1 Mechanical properties of composites with different contents of wood powder

Wood powder content (%)	0	10	20	30	40
Tensile strength (MPa)	7.93	5.39	5.25	5.08	4.86
Tensile modulus (GPa)	0.602	0.674	0.742	0.817	0.883
Flexible strength (MPa)	15.43	12.44	11.05	10.82	9.95
Flexible modulus (GPa)	0.550	0.584	0.613	0.632	0.777
Source : Xin <i>et al.</i> (2009)					

powder with a ratio of 10:8-9 in volume, while the viscosity reducer and the light stabilizer accounted for 5% to 20% and 0.2% to 6% of the total mass of the WPCs, respectively (Guo et al., 2011). When the applied laser had an energy density of 283 W/mm², the SLS-printed composite parts obtained sufficient strength and relatively high dimensional accuracy. Wax infiltration was introduced as a post-processing to improve properties of 3D-printed parts, after which the void fraction decreased significantly from 51% to 7%, with mechanical properties also improved greatly, namely, average values of tensile strength, bending strength and impact strength increased to 1.214 MPa, 2.730 MPa and 1.413 kJ/m², respectively (Figure 1) (Guo et al., 2011). The surface quality of the final parts was also improved after post-processing.

Zeng et al. (2012) used the same material and process as Guo et al. did; the only difference is that the wood powder was replaced by rice husk powder in their study. The volume ratio of the reinforcement and matrix in the powder mixture remained the same, with the same amount of viscosity reducer and light stabilizer. The tensile, impact and bending strengths of rice husk plastic composites (RPCs) were 10.7 times, 78% and 2.6 times those of the WPCs, respectively [Figure 2(a)]. After the wax post-processing, mechanical properties were improved further, and the average tensile, bending and impact strengths were 1.47 MPa, 3.86 MPa and 3.74 kJ/m², respectively [Figure 2(b)] (Zeng et al., 2012). Therefore, the RPC parts exhibited tensile, bending and impact strengths that are 21%, 41% and 165% higher than those of the WPCs, respectively (Zeng et al., 2012).

In the paper presented by Zhang et al. (2016), wood flour made from various plants (rice husk, cornstalk, pine, bamboo, eucalyptus and microcrystalline cellulose) were mixed with two types of thermal plastic materials (PP and Co-PES). Flory-Huggins Theory was implemented to estimate the miscibility of the cellulose and Co-PES used in the WPCs, and the optimized mass ratio of these two contents was found to be 25:75. They also introduced an alternative post-process option that uses epoxy resin to infiltrate the 3D-printed parts. Both efforts significantly improved the mechanical properties of the WPCs, and Table 2 shows the detailed mechanical property improvements.

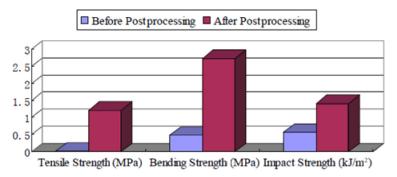
To sum up, very few types of biofibers have been implemented in powder-based 3D printing systems so far, with all of them being short fibers. The key idea of using a powder-based 3D printing system is to hot-melt some adhesive polymer powder and bond mixed short biofibers together to form the part. There are also inspirations of post-processing methods such as wax or epoxy infiltration, which can significantly reduce the 3D printed parts' porosity but improve their mechanical properties. However, the overall properties of 3D printed bioparts using SLS are still much lower than those of parts made from pure plastic polymers due to the poor interfacial bonding qualities between the biofiber and the polymer. The highest mechanical properties reported by all literature mentioned above are summarized in Table 3.

2.2 Material extrusion systems

A common 3D printing technology used to produce biocomposites based on fibers infused with molten polymers is material extrusion, which can be applied to both discontinuous

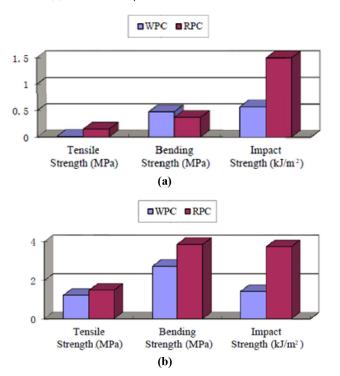
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Figure 1 Mechanical property comparisons of SLS-printed WPCs before and after post-processing



Source: Guo *et al.* (2011)

Figure 2 Average mechanical property comparisons of (a) WPCs and RPCs and (b) wax-infiltrated parts



Source: Zeng et al. (2011)

(short) and continuous (long) fibers. This technology is by far the most widely used AM process today (Turner and Gold, 2015; Galantucci *et al.*, 2009; Bellini *et al.*, 2004), and probably the most developed technology in 3D printing of

biofiber-reinforced polymers. In traditional material extrusion systems, the parts are constructed through deposition of molten filament materials made from polymers (HashemiSanatgar et al., 2017) such as polylactic acid (PLA) (Ang et al., 2007), acrylonitrile butadiene styrene (ABS) (Nikzad et al., 2011; Hernandez, 2015) and ABS plus (the ultimate ABS replacement that prints strong and beautiful parts on any fused deposition modeling 3D printer without common problems associated with regular ABS such as warping and horrible bed adhesion) (Nuñez et al., 2015; Gibson and Stucker, 2015), although metals (Kazmer, 2017) and ceramics (MEMSnet, 2019; Accuratus, 2013) can also be used. The solid filament is heated to a temperature between 210°C to 250°C (Kaveh et al., 2015; Rahman et al., 2018), which is above its melting temperature before being extruded. The extrudate is then coldwelded to the part surface of a much lower temperature in approximately 0.1s (Groover, 2016). A separately extruded support material is used to create support structures for overhangs (Buj-Corral et al., 2018), flat base supports (Villalpando et al., 2014), and to stabilize the part during fabrication (Song and Telenko, 2017; Wohlers, 2013).

For biofiber-reinforced biocomposites, polymer pellets and cut short fibers are first mixed in a blender and then sent to an extruder to be made into filaments. A second extrusion process could be conducted to ensure the homogenous distribution of fibers (Wang et al., 2017). Alternatively, long continuous biofiber can also be infused or coated with polymer paste to create pre-preg composite filaments and then directly extruded for 3D printing parts. Both these processes are typically referred to as fused filament fabrication (FFF) (Goh et al., 2018). Traditional advanced short fibers, including glass fibers (Zhong et al., 2001) and CFs (Tekinalp et al., 2014; Ning et al., 2015; Love, 2014; Ning et al., 2016; Griffini et al., 2016), are commonly used as reinforcements to strengthen mechanical

Table 2 Comparisons of mechanical properties before and after content mass ratio optimization

		Before		After	
Mechanical properties	Green parts	Wax-infiltrated parts	Green parts	Wax-infiltrated parts	Epoxy resin-infiltrated parts
Tensile strength (MPa)	0.014-0.54	1.24-2.40	2.30-4.85	5.80-6.34	10.42-11.03
Bending strength (MPa)	0.22-0.48	0.76-2.73	8.22-10.69	10.43-11.70	15.13-19.02
Impact strength (kJ/m²)	0.36-0.57	1.41-1.55	1.14-1.25	1.43-2.97	4.45-5.51

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Table 3 Maximum mechanical properties of bio composites reported using SLS

Mechanical properties	Tensile strength (MPa)	Flexural strength (MPa)	Impact strength (kJ/m²)
40% wood powder + PP and hot-melt adhesive (Xin et al., 2009)	7.93	15.43	N/A
~50 v% wood flour + Co-PES after post-processing (Guo et al., 2011)	1.214	2.73	1.413
~50 v% rice husk powder + Co-PES after post-processing (Zeng et al., 2012)	1.47	3.86	3.74
75% cellulose powder with Co-PES after epoxy infiltration (Zhang et al., 2016)	10.42-11.03	15.13-19.02	4.45–5.51

properties of polymer composites in 3D printing areas. The fiber orientation and void fraction of composites are two important factors in determining the properties of the final composite parts. Currently, the added content of fibers can be as high as 40 Wt.%, and composites with higher fiber contents are unable to be printed due to print head nozzle clogging issues. In addition, composites with higher fiber loading are difficult to make into continuous filaments for FFF due to the loss of toughness (Wang et al., 2017). Therefore, the properties of the resulting composites are limited by low fiber content. Another challenge for FFF is the difficulty of adding continuous fibers. To date, most studies have presented only the addition of short fibers in the polymer matrix (Wang et al., 2017).

To implement FFF processes in the manufacturing of biocomposites with short biofibers, Correa et al. (2015) developed a method that allows for greater control and intensified wood transformations through the precise design of multi-material prints composed of both wood fibers and polymers. The so-called "Four-dimensional printing" consists of 3D-printed multi-material polymer structures, which are able to change shape and physical properties over time. The "wood" FFF filaments were developed by combining a suitable 3D printing polymer with micro-wood fibers such as a Co-PES composite with high cellulose content ($\sim 40\%$), that is, commercially available under the name "laywood" (Correa et al., 2015). By taking advantage of the FFF technology, they managed to deposit material in precisely defined grain patterns, controlling the anisotropic behavior of the material by designing a custom "wood grain" specifically to enhance shape change (Correa et al., 2015). It is their plan to analyze the physical and mechanical properties of the 3D printed wood composites.

Montalvo and Hidalgo (Montalvo and Hidalgo, 2019) studied the FFF process with compound filaments using different plastic matrices and sugarcane bagasse as the filler. A plastic extruder was modified to obtain a 1.75 mm compound filament by using a 3×4 design of experiments with the following factors, namely, fiber content (10%, 20% and 30%) and matrix type (polyethylene (PE), PP, ABS and PLA). Table 4 shows the extrusion results of all fiber-matrix combinations they tested. They found that the mixture of PP with 20% sugarcane bagasse had the best behavior at the time of extrusion, yet the fibers could be considered only as filler but not as reinforcing agents, as pure PP has better properties than most compounds. This combination was used in a test that determined the best nozzle diameter, and the nozzle test results are listed in Table 5. Based on these results, the extruder nozzle diameter was selected to be 0.6 mm, which is 50% larger than the regular nozzle used in polymer 3D printing.

Table 4 Extrusion results of different fiber-matrix combinations, where "Y" = "yes," "N" = "no," "H" = "high," "M" = "medium" and "L" = "low"

		Fiber content				
Matrix	Property	0%	10%	20%	30%	
PE	Extrusion	Υ	Υ	Υ	Υ	
	Speed	M	L	N	N	
	Fragility	L	L	_	_	
PP	Extrusion	Υ	Υ	Υ	Υ	
	Speed	Н	Н	M	L	
	Fragility	L	L	L	M	
PLA	Extrusion	Υ	Υ	Υ	Υ	
	Speed	M	L	N	N	
	Fragility	Н	Н	_	_	
ABS	Extrusion	Υ	N	N	N	
	Speed	M	_	_	_	
	Fragility	Н	-	-	_	

Source: Montalvo and Hidalgo (2019)

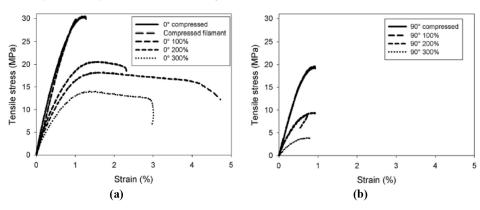
Duigou et al. (2016) reported using the "Woodfill fine" filament from a company named ColorFabb as a blend of PLA and polyhydroxyalkanoate (PHA) matrix reinforced with recycled wood fibers for FFF 3D printing. The fiber content equaled $15.2 \pm 0.9\%$ weight fraction. A Prusa i3 Rework 3D printer with a 0.4 mm nozzle was used for printing samples for tensile tests, water uptake and swelling tests and free curvature measurements. Some samples were subsequently compressed on a thermal press (15 bars for 2 min at 210°C). Measured mechanical behavior and properties are shown in Figure 3. Properties obtained from 3D printed/compressed samples and compressed filaments are in the range of extruded or injection molded PP/30% wood and high-density polyethylene/40% wood combinations, but are lower than PHA/20% wood and PLA/20% wood combinations. Because the FFF process generates neither a high shear rate nor a high molding pressure, biocomposite parts made by FFF are typically worse in mechanical properties than those manufactured from extrusion, compression or injection molding (Duigou et al., 2016), however, it allows parts to be manufactured with more complex shapes. The hydroscopic behavior of FFF-printed biocomposites influences their mechanical response, making the parts more fragile than native biocomposites, which could be considered a drawback (Duigou et al., 2016). For hydromorphic dynamics, the authors found that a higher porosity could promote water transport, thereby enhancing the actuation speed of the biocomposite part in water. In contrast to compression molding, the FFF process leads to material

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 Table 5
 LDM printing test results using different nozzle diameters

Nozzle diameter (mm)	Continuous flow	Time without clogs (s)	Printing results
0.40	N	3	Very thin with some blank spaces
0.45	N	4	
0.50	N	5	Some clogs and separated lines
0.55	Υ	6	
0.60	Υ	10	Well-formed lines and separations
0.65	Υ	10	
0.70	Υ	10	Constant flow and overlapping lines
0.75	Υ	10	Overlapping lines and the material flows with the motor off
0.80	Υ	10	

Figure 3 Tensile behavior of wood biocomposite made by FFF as a function of printing width (100%, 200% and 300%) for (a): longitudinal printing direction (0°) compared to compressed samples and (b) 90° to the longitudinal direction



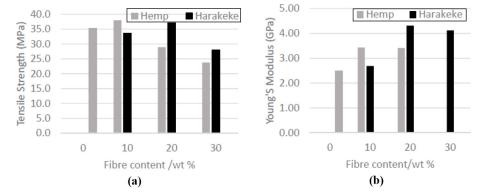
Source: Duigou et al. (2011)

characteristics that are suitable for a range of moisture-induced biocomposite actuation functionalities associated with improved mechanical properties (Duigou *et al.*, 2016).

Stoof *et al.* (2017) explored the feasibility and factors involved in using FFF to produce natural fiber-reinforced composite components. Uniform 3 mm filaments of hemp and harakeke (phormiumtenax) in various weight percentages with

PLA matrix were produced and used to print tensile test samples (Stoof *et al.*, 2017). Their test results indicated that 20 Wt.% harakeke FFF samples had a 42.3% higher Young's modulus and a 5.4% higher tensile strength than plain PLA samples, respectively, as shown in Figure 4. The same research team also produced a range of composite filaments with differing fiber and gypsum weight contents using pre- or

Figure 4 (a) Tensile strength and (b) Young's modulus of 10, 20 and 30 w% hemp and harakeke mixed with PLA



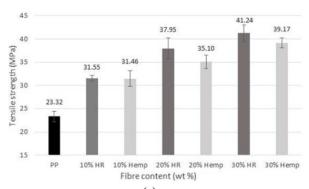
Source: Stoof et al. (2017)

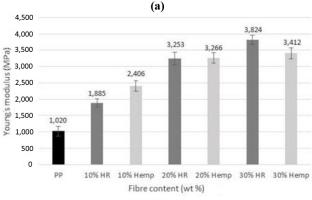
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postconsumer PP through the FFF process (Stoof and Pickering, 2019). They studied the influence of the tensile strength, Young's modulus and fiber content on the filament's surface finish. The most successful filaments in terms of tensile properties were made from 30 Wt.% harakeke in a postconsumer PP matrix that had a tensile strength and Young's modulus of 41 MPa [Figure 5(a)] and 3.8 GPa [Figure 5(b)], respectively, which were 77% and 275% higher than those of the plain PP samples, respectively (Stoof and Pickering, 2019). They also performed a mechanical property PP/glass comparison between preconsumer fiber biocomposites and postconsumer PP/natural fiber biocomposites and found out the latter exhibited substantially higher strength and Young's modulus.

Montalvo et al. (2018) investigated two different matrices of PP and PLA filled with four different wood flour contents (0%, 10%, 20% and 30%). The fiber content of 20% was found to have characteristics that are suitable for the FFF process. Biocomposite filaments were further used in FFF to obtain test samples for tensile and flexural property characterizations. These two mechanical properties of FFF samples were then compared to those of samples obtained by injection molding. The specimens produced by the FFF process exhibited lower tensile modulus and flexural modulus than the injection-molded ones. The injection-molded PLA and wood flour composite (PLA + WF) exhibited the highest tensile modulus of approximately 3.1 GPa and a flexural modulus of 3.4 GPa. This finding is in agreement with the stiffness of this composite,

Figure 5 (a) Tensile strength and (b) Young's modulus of postconsumer PP/harakeke and PP/hemp fiber composite filaments





Source: Stoof and Pickering (2019)

which is superior to that of the injected plain PLA. The PLA and PLA + WF specimens made through FFF, however, had much lower moduli than their injection-molded counterparts (Figure 6). They also performed thermogravimetric analyzes on the specimens to determine the optimum temperatures for processing biocomposites through FFF based on their thermal behavior. Based on their study, a suitable processing zone for FFF with formulations of wood flour based biocomposites is between 150°C and 195°C.

Matsuzaki et al. (2016) developed a method that uses the PLA filament and continuous fibers [CFs and twisted yarns of jute fibers (JF)] that are supplied separately to the FFF 3D printer. The reinforcement fiber was fully infused with a PLA within the heated nozzle right before printing takes place. Their study showed that the FFF-printed composites with continuous CF reinforcement exhibited significantly higher Young's modulus and tensile strength than those fabricated using commercially available 3D printers, regardless of whether the latter was produced via SLS, stereolithography (SLA) or FFF process. The strength of the CF composites from this study was doubled comparing to that of the composites made through conventional FFF, which expands the applicability of 3D printing to load-bearing components that are unable to be achieved by conventional FFF processes. However, the tensile strength of JF reinforced biocomposites is not improved significantly compared to that of CF ones, the tensile modulus and strength are 5.11 (± 0.41) GPa and 57.1 (± 5.33) MPa, respectively, which are corresponding to 157% and 134% of those of pure PLA specimens (Figure 7).

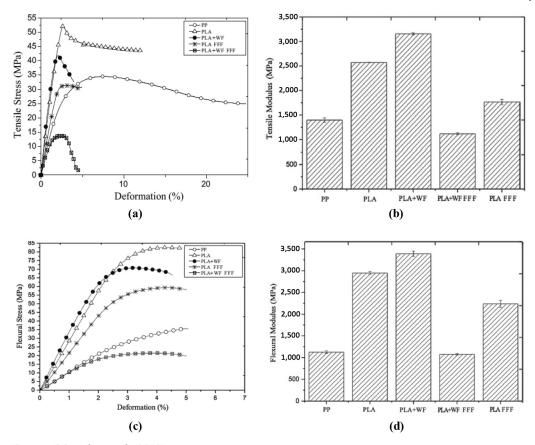
Another 3D printing technology based on material extrusion is known as liquid deposition modeling (LDM). In LDM, the composite feedstock is in the form of paste or fluid, the materials are selectively deposited from a syringe, that is, attached to the computerized numerical control machine (Goh et al., 2018), and only discontinuous fibers have been used in the composite feedstock so far.

Kariz et al. (2016) made the first attempt to print wood composites by means of LDM. They made mixtures of various ratios of wood powder and polyvinyl acetate and ureaformaldehyde adhesive for extrusion. A nozzle of 3 mm was used for extrusion and the force needed for extrusion was measured. It is found that the extrusion force increased with the adding amount of wood powder in the adhesive mixture, and the amount of wood powder in the 3D printed objects was restricted to 15% to 20%. The 3D printed blocks were left to cure on a hot plate with a temperature of 50°C for 2h and followed by one-week solidification period in a standard climate. They also found that the bending properties of the 3D printed blocks depended to a high degree on the type of adhesive used (Figure 8). Part shrinkage was also observed for both types of adhesives due to water removal. The largest shrinkage was found in the z-direction (thickness), which was also partly caused by the material flowing down due to gravity.

Rosenthal et al. (2018) reported using air-dried sawdust and methylcellulose (MC) as the binding agent for LDM 3D printing. In total, two sawdust particle sizes were used (0.25 mm and 0.4 mm) and they were mixed with MC and water at different ratios. An 8 mm nozzle with a length of 51 mm was used and the printing was performed at a traverse speed of 1 mm/s. Both the printed parts' dimensional stability

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Figure 6 (a) Tensile curves, (b) tensile moduli, (c) flexural curves and (d) flexural moduli of different material combinations in the study



Source: Montalvo et al. (2018)

and bending properties were measured and they found a reduction of 17.3% to 20.0% shrinkage of the parts' heights, with the MOR ranging from 2.3 to 7.4 MPa, the MOE ranging from 284.8 to 733.1 MPa, and density of 3D printed specimens ranging from 0.33 to 0.48 g/cm³.

For 3D printing of biofiber reinforced composites based on material extrusion, several studies have been explored for using the FFF technology, with both cut short fibers and continuous long fibers. For the short-fiber scenario, various biofibers are cut and mixed within different polymers typically used for FFF processes, but they are rarely helpful in improving mechanical properties for the printed parts. Most of them can only serve as fillers and may be helpful in increasing the complexity of the parts that can be 3D printed. On the contrary, continuous long fibers that are infused or coated by polymers right before being extruded from the printing nozzle seem to be very beneficial in improving the printed parts' mechanical properties. Therefore, FFF technology using continuous long biofiber should obtain more and closer attention in future research and investigations. For LDM based biocomposite 3D printing, the number of studies is much limited comparing to FFF, with all research performed so far regarding short fibers, especially wood powders. The measured mechanical properties are much lower comparing to those of parts made from FFF. Table 6 shows a summary of the mechanical properties reported by the studies covered in this section.

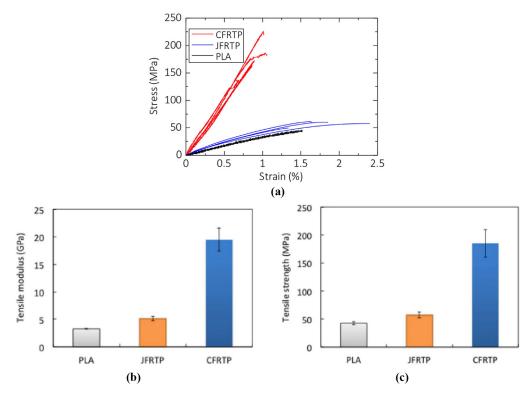
2.3 Liquid-based systems

Both SLA and digital light processing (DLP) are classified as liquid-based systems because they start with a liquid polymer or resin. DLP is a 3D printing process originally developed in 1987 by Larry Hornbeck of Texas Instruments (Wikipedia, 2019), in which a projector is used to cure photopolymer resin. The first DLP-based projector was introduced by Digital Projection Ltd. in 1997 (Wikipedia, 2019). A safelight (light bulb) is used instead of the ultraviolet (UV) laser to cure the photopolymer resin based on optical micro-electro-mechanical technology that uses a digital micromirror device (Wikipedia, 2019). High-resolution objects are 3D printed at a high speed either being pulled out of the resin, which creates space for the uncured resin at the bottom of the container and to form the next layer of the object or down into the tank with the next layer being cured on the top (Sculpteo, 2019).

Li et al. (2019) used DLP in producing cellulose nanocrystals (CNCs) reinforced polyethylene glycol diacrylate (PEGDA) glycerol composites. CNCs are highly crystalline cellulose nanoparticles that have diameters between 5 to 50 nm and lengths between 100 and 1,000 nm (Habibi et al., 2010; Moon et al., 2011; Faruk et al., 2012), with an elastic modulus higher than that of glass, Kevlar, and even some metal fibers. In their work, Freeze-dried CNC derived from wood pulp were introduced to improve PEGDA's physical strength, which provided a more efficient, green and scalable approach for generating CNC compatible polymer resin and enabled larger-

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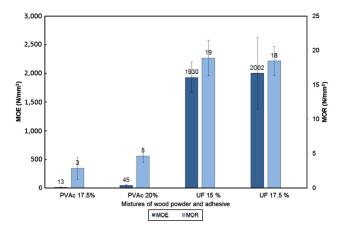
Figure 7 Mechanical property comparisons among carbon fiber reinforced thermoplastic (CFRTP), jute fiber reinforced thermoplastic (JFRTP) and pure PLA



Notes: (a) Stress-strain curves; (b) tensile moduli; (c) tensile strengths

Source: Matsuzaki et al. (2016)

Figure 8 Average modulus of elasticity (MOE, left columns) and flexural strength (MOR, right columns) of the 3D printed blocks made from different mixtures



Source: Kariz et al. (2016)

scale use in future industrial applications. Performed tensile tests on dog bone-shaped composites printed with different CNC concentrations showed a positive effect on the average Young's modulus as the concentration of CNCs increased. The average strain at break maintained roughly constant around 10% until the CNC concentration went above 1 Wt.%. Finally, the average ultimate strength reached its peak value of

 $7.6\pm0.3\,\mathrm{MPa}$ at 1 Wt.% CNC concentration and then decreased slightly as CNC concentration further increased (Li et al., 2019) [Figure 9(a)]. The authors also studied the relationship between the 3D printing layer thickness and Young's modulus of the printed part. The tensile testing results indicated that the Young modulus ranged from $80.7\pm3.7\,\mathrm{MPa}$ to $54.4\pm7.5\,\mathrm{MPa}$ as the curing layer thickness increased from $25\,\mu\mathrm{m}$ to $300\,\mu\mathrm{m}$ (Li et al., 2019) [Figure 9(b)].

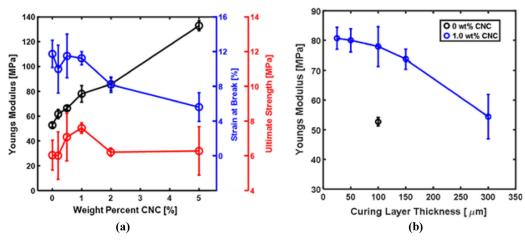
SLA was the first material addition rapid prototyping technology, dating back to around 1988 when it was made available by 3D Systems Inc. based on the work of Charles Hull (Groover, 2016). The original version of SLA is a process for fabricating a solid plastic part out of a photosensitive liquid polymer using a directed laser beam for part solidification, in which every single layer consists of its own two-dimensional shape so that the continuous addition of layers creates the solid part shape (Groover, 2016). SLA can produce fine features using a variety of polymers, metals and ceramics with good accuracy (Choi et al., 2009), and is generally less expensive than photosensitive resins (Groover, 2016). It has traditionally used photo-sensitive materials (or resins) (Taft et al., 2011; Chang et al., 2016; Dusel et al., 1995), however, other photosensitive powders have been used within suspensions for this process (Sun and Zhang, 2002; Lian et al., 2017; Liu et al., 2018; Wu et al., 2017; Bartolo and Gaspar, 2008; Sano et al., 2018), as well as composites such as discontinuous (e.g. nano, micro and milli scales) and continuous fibers with polymers (Goh et al., 2018; Sano et al., 2018), etc.

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 Table 6
 Maximum mechanical properties of bio composites reported using material extrusion

Mechanical properties	Tensile strength (MPa)	Tensile modulus (GPa)	Flexural strength (MPa)	Flexural modulus (MPa)
15.2 \pm 0.9 w% woodfill file filament $+$ PLA and PHA, 0° compressed				
(Duigou et al., 2016)	\approx 30	≈4	N/A	N/A
10 w% hemp + PLA (Stoof <i>et al.</i> , 2017)	\approx 38	≈ 3.5	N/A	N/A
20 w% harakeke + PLA (Stoof et al., 2017)	\approx 37	pprox 4.2	N/A	N/A
30% Harakeke + postconsumer PP (Stoof and Pickering, 2019)	41.24	3.824	N/A	N/A
20% wood fiber + PLA (Montalvo et al., 2018)	\approx 17	\approx 1.1	pprox 20	≈ 1.05
6.1 v% jute + PLA (Matsuzaki <i>et al.</i> , 2016)	57.1	5.11	N/A	N/A
15% wood powder + UF (Kariz et al., 2016)	N/A	1.93	19	N/A
17.5% wood powder + UF (Kariz <i>et al.</i> , 2016)	N/A	2.002	18	N/A
85.5 w% wood sawdust + MC (Rosenthal <i>et al.</i> , 2018)	N/A	0.7331	7.4	N/A

Figure 9 (a) DMA tensile mechanical properties of dog bone composites DLP printed with various CNC concentrations. Black: Young's modulus, Blue: Strain at break, and Red: Ultimate strength; (b) Young's moduli of dog bone composites DLP printed with different curing layer thicknesses



Source: Li et al. (2019)

The low strength of the cured photosensitive resin has always been a major problem in the development of photocuring SLA, yet there are few studies regarding biofiber-reinforced SLA polymer composites so far. Quan et al. (2018) reported using a three-dimensional braided fabric structure to compensate for this shortcoming. They built a novel SLA printing platform that integrated different fiber yarns [carbon, glass or high-strength PE (HSPE)] in the x, y and z directions. In total, three UV lighting projectors were used to deliver overlapping UV light at the same time with the same light strength. The fiber yarns were fixed in UV-sensitive resin while curing. The schematic layout of the platform is shown in Figure 10. Different fiber yarns were used with the same resin to make tensile test samples. Their test results indicated that both the elastic modulus and the tensile fracture strength of the SLA-based 3D woven parts were significantly improved than those of the parts made from pure resin. The reinforcing effect of the HSPE yarn was the best, followed by glass fiber and CF (Figure 11) (Quan et al., 2018).

To date, 3D printing with biofibers using SLA technology is still under initial development, and therefore, calls for further research and investigation. The study carried out by Quan *et al.*

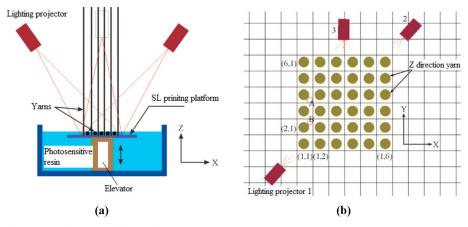
does indicate an improvement of part mechanical properties than those of its plain resin counterparts. Although only advanced fibers were used in their study, their attempt inspires future investigation with the implementation of biofibers with traditional and/or biodegradable photosensitive resins in making biocomposite parts via SLA technology.

2.4 Solid-sheet systems

Solid-sheet 3D printing systems use solid sheets as their starting material. One typical example of the solid-sheet system would be laminated object manufacturing (LOM). LOM uses laser, ultrasonic or knife cutting, lamination and bonding of two-dimensional cross-sections to build parts layer by layer (Ahn et al., 2012; Park et al., 2000) with a typical layer thickness between 0.05 and 1.0 mm depending on the material and application (Feygin et al., 1998; Kechagias, 2007; Kunwoo, 1999; Liou, 2008). Any excess material in each layer remains in place after it is cut to support the part, that is, being built (Groover, 2016). The sheet material usually comes with an adhesive backing as rolls that are spooled between two reels (Groover, 2016); otherwise, an additional adhesive coating step

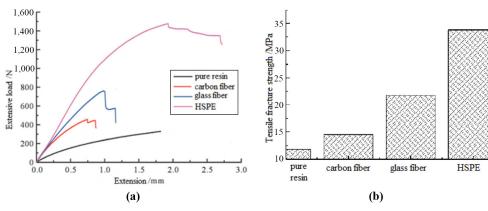
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Figure 10 Schematic layout of the SLA 3D printing platform



Notes: (a) Sde view; (b) top view **Source:** Quan *et al.* (2018)

Figure 11 (a) Sample tensile test curves and (b) tensile fracture strengths of different yarns



Source: Quan et al. (2018)

process must be included for each layer. Helisys, Inc. was the original company that offers LOM systems but was later acquired by Cubic Technologies. Their machines process paper sheet stock backed with adhesive and use a sequence in which the most recently added sheet is bonded to the existing structure before cutting the outline in that layer. A heated roller is then used to melt the thermoplastic adhesive in the bonding operation (Groover, 2016). Paper, cardboard, and plastic in sheet stock form are traditional starting materials for LOM 3D printers (Groover, 2016). If paper or cardboard is used, the finished products are biodegradable but with relatively low mechanical properties. Subsequent updates in LOM technology introduced by other researchers and companies to date include:

- use of a blade or ultrasonic cutter rather than laser to do the cutting;
- polymeric sheet stock or metal foils (Prechtl *et al.*, 2005) rather than paper as the starting material; and
- changing the process sequence so cutting of the layer outline goes before bonding of the new layer to the previous one (Groover, 2016).

The highlight of this last change is that it facilitates the fabrication of objects that possess internal features (Mohammed et al., 2015).

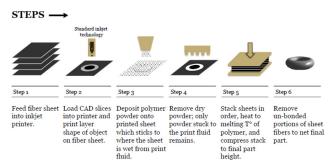
For the second change mentioned above, a few studies have been performed in strengthening the mechanical properties of the sheet stocks. A good example here would be the use of woodderived biomorphous ceramics. Weisensel et al. (2004) reported a LOM product developed from binding a pyrolyzed filter paper composed of cellulose fibers with an adhesive tape made from slurry containing phenolic resin, polyvinyl butyral, benzyl butyl phthalate and ethanol, in which the reaction bonding technique was used. In this process, the molten silicon infiltrates a porous SiC-rich preform and reacts with in situ carbon to fill up pores (Klosterman et al., 1999). After binding, this product was pyrolyzed again at 800°C in a nitrogen atmosphere to convert the phenolic resin into carbon. The porous carbonized samples were then post-infiltrated with liquid Si at 1,500°C under vacuum for 1 h or 7 h to produce a BFPC. The composite parts obtained had an average bending strength of 130 ± 10 MPa if infiltrated for 1 h or 123 ± 8 MPa if infiltrated for 7 h, both in the same range as other SiC-materials made through 3D printing.

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Another recently developed solid-sheet based 3D printing technology is known as Composite-Based Additive Manufacturing (CBAM), which follows the process as shown in Figure 12. Inkjet technique was used to first deposit water-based glue onto a single layer of advanced fiber sheet based on the cross-sectional shape of the part to be built. Once the glue deposition is completed, polymer powder is deposited to cover the entire solid-sheet layer and sticks to the sheet at locations where the glue was printed. Excess powder is then removed and leaving locations, where the glue was applied, covered with the polymer powder. This process is repeated for all layers until all sheets are stacked. The entire part is then heated to the melting temperature of the polymer and compressed for consolidation. Finally, mechanical (e.g. sandblast) or chemical process is applied to remove un-bonded portions of sheet fibers to reveal the desired part (Impossible Objects, 2018).

One company that implements the CBAM technology to date for its production is called Impossible Objects, located in Northbrook, IL. It uses nonwoven mats of carbon, glass and Kevlar fibers to 3D print strong, functional and complex parts at a faster speed than using conventional processes (Impossible Objects, 2016). The tensile strength of their CBAM carbon-nylon parts can reach 150 MPa, with the tensile modulus being as high as 11.0 GP, both of which are several times higher comparing to the strongest FFF printed nylon parts, as shown in Figure 13. The company is in the process of expanding possible fiber fabrics from

Figure 12 How CBAM works



Source: Kaplan (2017)

those mentioned above to others including biofibers such as silk and cotton (Kaplan, 2017), making the process more sustainable.

Generally, solid-sheet based 3D printing is inexpensive due to the readily available raw material, relatively high dimensional accuracy, high fiber contents and short building time, and it is the only technology that ensures improvements in material properties comparing to the same process using its original starting materials so far. However, the types of reinforcing fibers it can use are still very limited at present. There remains much more to be investigated for this system to be further developed and used in AM of BFPC parts consisting of biofiber reinforcements (e.g. natural woven textiles or mats) and/or bioresins, which should result in a considerable enhancement in material properties of built parts.

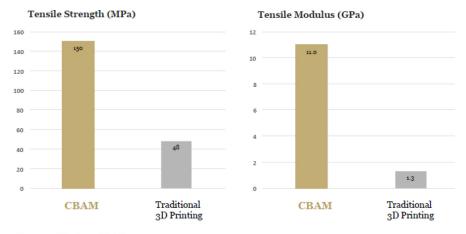
2.5 Summary of three-dimensional printing studies regarding biofiber reinforced composites

All the 3 D printing studies performed so far regarding biofiber reinforced composites introduced above are summarized in Table 7, with the technology they implemented, together with the biofiber, matrix and any necessary post-processing listed.

3. Conclusions

This review paper is a summary of current attempts to use biofibers as reinforcements together with various resin systems as the starting materials for 3D printing processes, with the focus on research and studies performed in a lab rather than commercially available 3D printers, and most of the currently available rapid prototyping techniques are included herein. All of the reviewed attempts are solutions to some principal problems with existing 3D printing processes such as the limit in the variety of materials and the poor mechanical performance of 3D printed parts. Different types of biofibers are involved in these studies, namely, wood flour, rice husk powder and cellulose in SLS processes; microwood fibers, sugarcane bagasse, hemp and harakeke short fibers, jute long yarns and wood flour in FFF processes; wood powder, sawdust and filter paper in LDM processes; CNC derived from wood pulp in DLP processes; cellulose sheet stocks in LOM processes; and finally silk and cotton fibers in CBAM processes. Additionally, these studies implemented both traditional (e.g. ABS, PP, PE and Co-PES) and biodegradable

Figure 13 Comparison of CBAM carbon-nylon to the strongest traditionally 3D printed nylon (FFF)



Source: Kaplan (2017)

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 Table 7 Summary of 3D printing of biofiber reinforced composites discussed in this paper

Technology	Biofiber	Matrix	Post-processing	Reference
SLS	Wood powder	Hot-melt adhesive powder and PP powder	N/A	Xin et al. (2009)
SLS	Alkalized wood flour	Co-PES with some viscosity reducer	Wax infiltration	Guo et al. (2011)
SLS	Rice husk powder	Co-PES	Wax infiltration	Zeng <i>et al.</i> (2012)
SLS	Wood power	PP and Co-PES	Epoxy infiltration	Zhang et al. (2016)
FFF	Microwood fibers	Co-PES	N/A	Correa et al. (2015)
FFF	Sugarcane bagasse	PE, PP, ABS and PLA	N/A	Montalvo and Hidalgo (2019)
FFF	"Woodfill fine" filament	PLA	N/A	Duigou <i>et al.</i> (2016)
	Recycled wood fibers	PHA		
FFF	Hemp and harakeke filaments	PLA	N/A	Stoof et al. (2017), Stoof
		Pre- or postconsumer PP		and Pickering (2019)
FFF	Wood flour	PP and PLA	N/A	Montalvo et al. (2018)
FFF	CFs and twisted yarns of jute fibers	PLA	N/A	Matsuzaki et al. (2016)
LDM	Wood powder	PVAc or UF	2 h cure on a 50°C hot plate followed by 1-week standard climate drying	Kariz <i>et al.</i> (2016)
LDM	Air-dried sawdust	MC	Drying (60°C, 5 days) and conditioning (20°C, 65% RH, 7 days)	Rosenthal et al. (2018)
DLP	CNC derived from wood pulp	PEGDA	N/A	Quan et al. (2018)
LOM	Pyrolyzed filter paper composed of cellulose fibers	Adhesive tape made from slurry containing phenolic resin, polyvinyl butyral, benzyl butyl phthalate and ethanol	Pyrolyze again at 800°C in a nitrogen atmosphere, and post-infiltrated with liquid Si at 1500°C under vacuum for 1 h and 7 h	Weisensel et al. (2004)
CBAM	Silk and cotton	Polymer powder	Mechanical (e.g. sandblast) or chemical process applied to remove un-bonded portions	Kaplan (2017)

(e.g. PLA and PHA) resin systems. Most of the biofiber-reinforced 3D-printed products exhibited improved mechanical properties than products printed using pure resin, indicating that biofibers are good replacements for synthetic ones. However, biofibers are far from successfully replacing synthetic fibers due to several of their disadvantages including higher moisture absorbance, lower thermal stability and mechanical properties. Significant numbers of studies are being performed to solve these problems, yet research concerning biofiber-reinforced composite parts is still quite limited regarding some 3D printing technologies (e.g. LOM and SLA). This paper unveils potential research directions and areas that would further widen the technology and the variety of materials that can be used in 3D printing in a sustainable manner for researchers working in relevant fields.

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