Fused Deposition Modeling 3D Printing of Continuous Natural and Regeneration Fibers Reinforced Polymer Composites and Its Mechanical Properties under Extreme Environmental Conditions—A Critical Review

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Continuous fiber 3D printing, as a new technology, has attracted attention in an increasing number of applications. Research on continuous fiber 3D printing is currently still in its infancy, after less than 10 years since its discovery in 2016. Many technical and fundamental questions still need to be addressed. Most literature has dealt with 3D printing employing continuous synthetic fibers (*e.g.*, carbon and aramid fibers). This critical review summarizes the progress on the 3D printing of continuous natural plant fibers and their regenerated fibers and their use in reinforced thermoplastic composites. Their mechanical performance under extreme environmental conditions is further reviewed. These high-performance continuous fiber reinforced polymer composites have potential for high value applications such as aerospace technologies.

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INTRODUCTION

Fiber-reinforced polymer composites have attracted attention recently due to their merits in terms of their light weight, dimensional stability, and robust mechanical properties (Yao *et al.* 2008; Khayat *et al.* 2015). They have potential for aerospace, military, and automotive applications (Langhorst *et al.* 2018). Most fiber-reinforced composites with simple and regular geometry are primarily manufactured *via* conventional manufacturing processes such as extrusion, injection molding, and thermoforming, *etc.*, while additive manufacturing or three-dimensional (3D) printing can achieve the fabrication of complex structures and shapes. Fused deposition modeling (FDM) 3D printing has been applied to polymer composites reinforced with discontinuous short fibers because this type of polymer can achieve substantially higher mechanical properties (Lee *et al.* 2021; Yang *et al.* 2021). Additionally, the alignment of fibers during the extrusion process further enhances the composite mechanical properties and microstructures (Dong *et al.* 2019). In most studies, however, these fibers are usually mixed into thermoplastic filaments before printing, and the resulting short fibers reinforced composites have

limitations in enhanced mechanical properties due to the limits of stress-transfer between the polymer matrix and reinforcement fibers. The process of additively manufactured continuous fiber-based composites, which was invented a few years ago as a robust novel technique to design high-performance fiber reinforced composites, makes it possible to achieve printed parts with complex topology (Matsuzaki *et al.* 2016). Most importantly, this invention of new technique pushes additive manufacturing toward a new stage. With a relatively short history and many technical and fundamental research questions un-addressed, continuous fiber 3D printing using polymer composites reinforced with continuous fibers *via* additively manufacturing has not yet reached industrial viability (Fijul Kabir *et al.* 2020).

Most available reviews on 3D printing continuous synthetic fibers reinforced composites have dealt with non-natural and non-renewable materials, *e.g.*, carbon, glass, and aramid fibers. In contrast, natural fibers offer many advantages compared to synthetic fibers, such as being lighter, easier to recycle and degrade, and renewable. However, FDM 3D printing of reinforcing thermoplastic polymer with natural fibers has certain challenges. The lignocellulose in natural fibers undergoes degradation during the process of constant high temperature (>200 °C), so thermal stability of natural fibers is one important factor limiting their fabrication in reinforced polymer composites *via* FDM 3D printing for advanced applications. Additionally, without appropriate drying protocols of natural fibers, water vapors released from natural fibers easily cause the hydrolysis of polymer matrices during FDM 3D printing, thereby producing voids in natural fibers reinforcing polymer composites. According to a Web of Science database search with the keyword "continuous natural fiber 3D printing", the current publications with this theme are fewer than 50. There are two reviews on continuous natural fiber 3D printing that focused primarily on the diverse FDM printing methods of continuous natural fibers reinforcing polymer composites (Tao *et al.* 2023; Cheng *et al.* 2024). Additionally, most studies of 3D printing continuous fibers (*e.g.*, carbon and Kelvar fibers) reinforced polymer composites have concentrated on mechanical properties under the room-temperature conditions, and the resulting composite mechanical performance under extreme environmental conditions (*e.g.*, cryogenic and high temperature) has been rare. It is worth noting that these fiber reinforced polymer composite material applications in aerospace and military [*e.g.*, poly(ether sulfone) (PPS), polyether-ether-ketone (PEEK), polyimide (PI), and polyurethane (PU) or nitrile butadiene rubber (NBR)] are inevitably exposed to extreme environmental conditions, such as aerospace. Therefore, this critical review focuses on the continuous natural and regeneration fiber 3D printing of reinforced polymer composites with extreme environmental mechanics. The polymer matrix focuses on thermoplastic and engineering plastics instead of thermosets. These high-performance continuous fiber reinforced polymer composites have potential for aerospace applications.

Continuous Synthetic Fibers

Because of the unique properties and broad applications in traditional laminated composites of synthetic fibers, most studies presently are primarily about continuous synthetic fiber 3D printing of reinforced polymer composites. Printing approaches can be classified into two categories, namely, nozzle impregnation (during printing) and prepreg filaments (prior to printing), where the fiber is embedded and then coated with the thermoplastic polymer in either melting fluid or solution status (Chen *et al.* 2021). Glass fiber, Kevlar fiber, and carbon fiber as the most common synthetic fibers have been primarily studied in the continuous fiber 3D printing. Tables 1 and 2 summarize the typical

works on continuous carbon and Kevlar fibers reinforced polymer composites, where the matrices involve thermoplastics and engineering plastics. For instance, comparative studies indicated that continuous carbon fiber reinforced nylon composites *via* additively manufacturing had the best mechanical performance (elastic modulus and tensile strength) among three types of continuous fibers, glass, Kevlar, and carbon fiber, which has the potential for complex aircraft components (Mei *et al.* 2019). Additionally, carbon fibers are always used in light-weight composites. Carbon fiber volume fraction has played a critical role in governing composite mechanical properties. Fiber orientation as another important parameter can govern composite mechanical performance. Carbon fibers can be printed with two approaches: concentric and isotropic. In the isotropic form, fibers can be printed at varied angles. Besides, optimized printing layer thickness and printing parameters in terms of printing speed, nozzle structure, printing temperature, infill pattern and density, printing path, feed rate of filament, *etc*., were found to contribute to enhanced composite mechanical properties. Furthermore, post-treatments could further enhance the mechanical performance of continuous carbon fiber 3D printing of reinforced composite (Handwerker *et al.* 2021; Wang *et al.* 2021), *e.g.*, hot press and thermal annealing. However, limitations have persisted in the continuous carbon fiber 3D printing due to its intrinsic in less flexibility and high cost (Zhang *et al.* 2020). In addition to technical issues, synthetic carbon fiber is far from meeting sustainability requirements. Natural fibers have merits in terms of their light weight and high strength and modules. Continuous natural fiber 3D printing also contributed to the reduction of greenhouse gas emissions, carbon peak reduction, and carbon neutralization. According to the life cycle analysis (LCA) regarding what competitive advantages quantitatively of natural fibers- *vs.* synthetic fibers-reinforced polymer composites, the natural fiber reinforced composites resulted in lower environmental impacts compared to the glass fiber reinforcing composites, and lower weight of natural fiber composites also improved fuel efficiency and reduced emissions during the use stage (Joshi *et al.* 2004). Additionally, easily accessible hydroxyl groups on the surface of natural fibers are feasible to take surface treatments for enhancing interfacial adhesion with their reinforced polymer composites along with their relatively cheap price. According to techno-economic analysis (TEA), natural fibers (*e.g.*, hemp or sisal fiber) have economically competitive advantages compared to synthetic fibers (*e.g.*, carbon or Kevlar fiber) in manufacturing fibers reinforced polymer composites. Therefore, additively manufactured continuous natural fibers reinforced polymer composites are promising for diverse applications.

Table 1. 3D Printing of Continuous Kevlar Fiber Reinforced Polymer Composites

Matrix and Fiber	Printing Type	TS/CS/FS (MPa)	FV (%)	YM/FM (GPa)	Literature
PLA/Kevlar Fiber	In nozzle impregnation	742.6[a]	50.0	41.3[a]	Hou *et al.* 2020a
PLA/Kevlar Fiber	In nozzle impregnation	150.0-200.0[b]	40.0	NA	Hou *et al.* 2023
PLA/Kevlar Fiber	In nozzle impregnation	207.0[c]	15.0	9.0[c]	Hou *et al.* 2020b
PLA/Kevlar Fiber	In nozzle impregnation	19.4[d]	3.5	0.9[d]	Dong *et al.* 2020
PLA/Kevlar Fiber	In nozzle impregnation	84.1[e]	20.5	3.6[e]	Cersoli *et al.* 2021
PLA/Kevlar Fiber	In nozzle impregnation	17.1[f]	11.5	NA	Hou *et al.* 2018

Notes: TS: Tensile Strength; CS: Compression Strength; FS: Flexural Strength; FV: Fiber Volume; YM: Young's Modulus; FM: Flexural Modulus; a: Longitudinal Tensile Strength and Modulus; b-c: Flexural Strength; d-e: Tensile Strength and Modulus; f: Compression Strength

Table 2. 3D Printing of Continuous Carbon Fiber Reinforced Polymer Composites

Matrices and Fiber	Printing Type	TS/FS (MPa)	FV (%)	YM/FM (GPa)	Literature
PA6/Carbon Fiber	Prepreg filament	591.1[a]	48.5	79.0[a]	Parandoush *et al.* 2019
PLA/Carbon Fiber	In nozzle impregnation	335.0[b]	27.0	30.0[b]	Tian *et al.* 2016
PP/Carbon Fiber	In nozzle impregnation	1930.0[c]	50.0	159.0[c]	Parker *et al.* 2022
Nylon/Carbon Fiber	Prepreg filament	200.0-250.0[d]	76.0	80.0-100.0[d]	Zhuang *et al.* 2023
Epoxy/Carbon Fiber	Prepreg filament	952.8[e]	58.0	74.0[e]	Ming *et al.* 2020
PLA/Carbon Fiber	Prepreg filament	9.6[f]	NA	0.12[f]	Liang *et al.* 2022

Notes: TS: Tensile Strength; FV: Fiber Volume; YM: Young's modulus; FM: Flexural Modulus; a-b and e: Flexural Strength and Modulus; c-d: Tensile Strength and Modulus; f: Compression Strength and Modulus

Continuous Natural Fibers

Natural fibers primarily involve two categories, organic and inorganic fibers. Plant fibers, as one type of organic fibers, are the most studied currently (Pal and Lucia 2019; Aruchamy *et al.* 2025; Manickaraj *et al.* 2025). Sisal, flax, hemp, ramie, jute, and cotton fibers are typical plant fibers, and their mechanical and physical properties are shown in Fig. 1 (Tuli *et al.* 2024). Short plant fibers reinforced polymer composites have been extensively investigated. For instance, the mechanical and fire retarding performance of wood and bamboo fibers reinforced polymer composites was systemically studied in terms of interfacial bonding, fiber volume fraction, polymer matrix type, *etc*. (Lei *et al.* 2007; Kim *et al.* 2011; Zhang *et al.* 2018). However, utilization of additively manufactured plant fiber reinforced polymer composites, with use of continuous fibers, is still rare (Long *et al.* 2021).

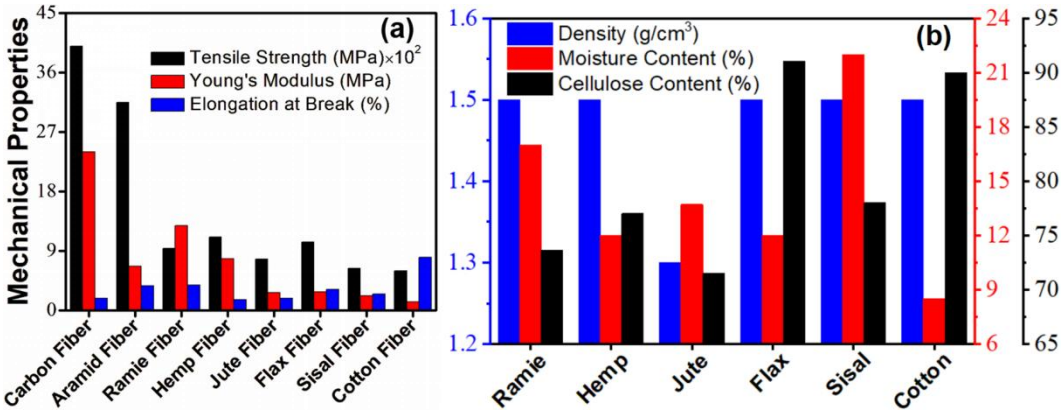

Fig. 1. Mechanical and physical properties of synthetic and natural fibers (a: Mechanical properties of synthetic and natural fibers; b: Physical properties of natural fibers)

For the continuous plant fiber 3D printing of reinforced composites, the composite matrix can be divided into four categories, petro- and bio-based non-biodegradable and petro- and bio-based biodegradable polymers. At the early stage during discovering this

new technique of continuous fiber 3D printing, non-biodegradable and synthetic polymers were explored as matrix polymers (acrylonitrile butadiene styrene ABS, polypropylene PP, and polyamide PA). For instance, the interfacial properties on the dependence of printing parameters on continuous ramie fiber 3D printed PP composites fabricated by in nozzle impregnation printing was studied (Cai *et al.* 2022). The extrusion flow rate governed both inter- and intra- strength of the resulting composites, and the printing speed parameter almost had no influence on mechanical performance of resulting ramie fiber reinforced composites. For another example, fiber orientations at three different patterns of 0, 90, and ±45 ^{o} on continuous flax fiber 3D printing of reinforced PA6 composites through in nozzle impregnation were studied (Terekhina *et al.* 2022), and the best tensile properties of resulting composites were achieved for printing at a raster angle of 0 ^{o}. Conversely, the tensile properties of the printing composites at 90 ^{o} were found to be the worst. Compared to the in-nozzle impregnation, prepreg impregnation was also investigated in continuous plant fiber 3D printing, *e.g.*, continuous jute fiber 3D printing of reinforced ABS composites (Razavi-Nouri *et al.* 2024). Fiber type is another important factor that also influences mechanical properties of fibers reinforced composites. Comparative studies about jute fiber, flax fiber, pineapple leaf fiber and ramie fiber indicated that the composite tensile strengths were found to depend on fiber types (Le Duigou *et al.* 2019; Suteja *et al.* 2022; Cheng *et al.* 2023).

Table 3. 3D Printing of Continuous Natural Fiber Reinforced Polymer Composites

Matrix and Fiber	Printing Type	TS/CS (MPa)	FV (%)	YM/FM (GPa)	Literature
PLA/Flax Fiber	Prepreg filament	183.0[a]	26.4	17.2[a]	Le Duigou *et al.* 2020
PLA/Jute Fiber	Prepreg filament	57.1[b]	6.1	5.1[b]	Matsuzaki *et al.* 2016
PLA/Jute Fiber	In nozzle impregnation	56.3[c]	60.0	3.4[c]	Hinchcliffe *et al.* 2016
PLA/Ramie Fiber	In nozzle impregnation	60.0-70.0[d]	15.0	3.2-4.0[d]	Wang *et al.* 2024
HIPS/Basalt Fiber	In nozzle impregnation	122.2[e]	55.2	5.6[e]	Zhang *et al.* 2023
R-PET/Banana Fiber	In nozzle impregnation	47.5[f]	15.8	1.5[f]	Ror *et al.* 2024

Notes: TS: Tensile Strength; CS: Compression Strength; FV: Fiber Volume; YM: Young's Modulus; FM: Flexural Modulus; a-e: Tensile Strength and Modulus; f: Flexural Strength; R-PET: Recycling PET; HIPS: High-impact Polystyrene

With the continuous fiber 3D printing technique development, critical questions in terms of low fiber volume fractions at less than 70% and relatively high voids have received attention, as the continuous fiber-reinforced composites *via* the traditional molding processes made it possible to achieve fiber volume fractions (> 60 to 70%) and void contents (< 1%) (Terekhina *et al.* 2022). The short impregnation time and small contact pressure during additively manufacturing are the primary reasons. Additionally, the natural yarns twisted by short fibers and with uneven surface make it difficult for the polymer matrix to flow over the fiber surface during the 3D printing (Kajbič *et al.* 2023; Long *et al.* 2023). As the mechanical performance of fibers reinforced polymer composites is generally positively correlated with fiber volume fractions, diverse methods have been

studied to further enhance fiber volume fractions and reduce voids in continuous fiber 3D printing (Zhang *et al.* 2023). However, most synthetic polymers in additively manufactured continuous fiber reinforced polymer composites have issues in biodegradability (Hubbe *et al.* 2021).

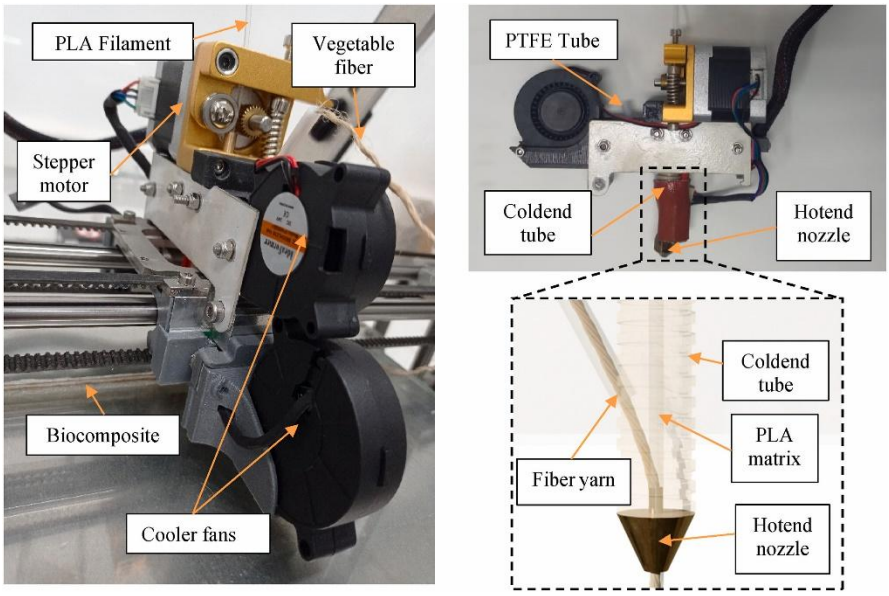

Fig. 2. Printing head images in 3D printing of continuous vegetable fiber (Santos and Cardoso 2023). Reprinted with permission from Elsevier Publishing Co.

To reduce carbon emissions and achieve a sustainable society, biodegradable polymer matrices such as polyhydroxyalkanoate (PHA) and polylactic acid (PLA) provide promising options for the development of continuous plant fiber 3D printing of reinforced bio-composites (Kovalcik *et al.* 2019; Qin *et al.* 2022; Tourang *et al.* 2023). PLA, as an example of bio-based polymer, was most studied in continuous natural plant fibers 3D printing of reinforced bio-composites in Fig. 2 (Santos and Cardoso 2023). Post-treatment as one simple and straightforward method is possible for tailoring fiber interfacial adhesion with PLA matrix. For instance, the post-thermal treatment contributed to the enhanced interfacial bonding in continuous vegetable fiber 3D printing of reinforced PLA composites, thereby enhancing mechanical performance of resulting vegetable fiber reinforcing bio-composites (Cheng *et al.* 2021; Wang *et al.* 2021; Jahangir *et al.* 2019). Due to less compatibility between the hydrophobic PLA matrix and hydrophilic plant fibers, interfacial adhesion as an important factor governs mechanical performance of additively manufactured fiber reinforced PLA bio-composites. For instance, additively manufactured PLA reinforced with continuous flax fiber was accompanied by plenty of flax fiber pull-outs inside the yarn during mechanical test. Additionally, surface treatment of natural fibers *via* silane coupling agents is another approach to further enhance interfacial adhesion. For example, silane coupling agents have been studied for tailoring interfacial adhesion of flax fiber and PLA matrix regarding 3D printing of continuous flax fiber reinforced PLA composites *via* an in-nozzle impregnation approach (Long *et al.* 2021; Chen *et al.* 2024). By tailoring printing parameters, there is also an efficient approach in regulating bonding adhesion between the plant fibers and PLA matrix. For instance, the interlayer strength of the PLA/ramie fiber composites improved with the increase in printing temperature and reduction in printing speed and layer thickness (Cheng *et al.* 2021). In addition to the

interfacial adhesion issues, inherently variable structure of natural plant fibers caused their reinforced composites with pits or nodes. Regeneration of natural and polyester fiber has merits compared to natural fibers as the regeneration fibers have regular cross-section and significantly less variability than natural fibers. Additionally, almost no obvious defects were observed. Considering the intrinsic limitations of natural plant fibers, development of regeneration of cellulose, lignin or polyester fibers attracted attention for the continuous fiber 3D printing.

Regeneration of Natural and Polyester Fibers

Regeneration of cellulose fibers

The preparation of regeneration cellulose fibers generally involves two stages, cellulose dissolution and then its regeneration. Cellulose dissolution is completely different from those of small molecular compounds. The polymer is swollen with the "good solvent" first, and individual chains then separate from each other if the dissolving power is great enough, leading to a dissolved state. To be able to serve as a "good solvent," it is necessary that the solvent dissolution parameters of the target polymer and solvent are similar. Unfortunately, many common organic solvents are good swelling agents for cellulose, but none of them can dissolve cellulose due to its strong inter- and intra- hydrogen bonds and intrinsic crystalline nature (French 2022). However, the strong hydrogen bonding interactions do not automatically explain their difficult solubility, as most hydrogen bonded substances dissolve in water because hydrogen bonding between water and the compound is stronger than between the compound molecules themselves. Additionally, crystallinity by itself does not explain its low solubility, as many crystalline systems dissolve in water. Therefore, cellulose dissolution is a constantly challenging theme.

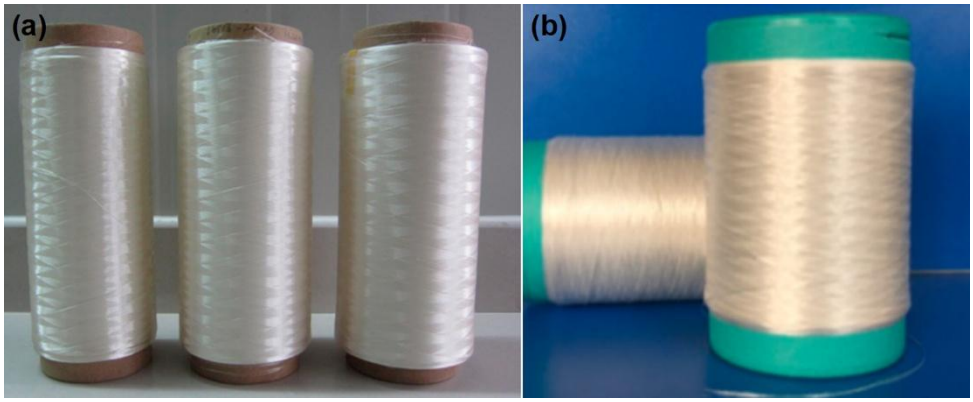

Fig. 3. Regenerated cellulose filaments from cellulose (a) and its hybrids with chitosan or cellulose nanofiber (b) (Fu *et al.* 2014; Zhu *et al.* 2019). Figure republished with permission from American Chemical Society.

Cellulose dissolution solvents are primarily classified as either derivatizing solvents or non-derivatizing solvents. The non-derivatizing solvents are a primary theme, as they separate the individual cellulose chains from each other without chemical modifications. Such solvent systems include sodium hydroxide/water, phosphoric acid/water, and transition metal complexes as the conventional solvent systems, and deep eutectic solvents and ionic liquids as novel solvent systems (Stefanescu *et al*. 2012; Petrovan *et al*. 2000). For instance, regeneration cellulose filaments have been prepared in mass scale *via* the environmentally friendly sodium hydroxide and water mixed solvents

with concentration at 5 to 35%, as shown in Fig. 3 (Fu *et al.* 2014; Yue *et al.* 2016; Zhu *et al.* 2019). As another example, to further tailoring cellulose fiber strength, cellulose nanofibers were co-assembled with cellulose to produce cellulose filaments on a mass scale (Qiu *et al.* 2018). These cellulose filaments have the potential for continuous cellulose fiber 3D printing as a potential alternative to Kevlar and carbon fibers.

Although cellulose fiber has unique merits in continuous fiber 3D printing, few studies have been reported on the continuous cellulose fiber 3D printing of reinforced composites. No commercial cellulose filaments are now available to manufacture these composites. Only one study has been reported dealing with 3D printing of composites *via* prepreg filament from continuous regeneration cellulose fiber and Pebax, a polyether block amide thermoplastic elastomer, and the composites were shown to have robust mechanical properties compared to other manufacturing processes (Touchard *et al.* 2023). Although the intrinsic hydrophilic nature of cellulose fibers poses a challenge in interfacial adhesion with polymer matrix, this unique property makes possible the development of PLA fibers reinforced with cellulose or PHA bio-composites with water-responsive shape memory functions, named as 4D printing of continuous cellulose fibers reinforced bio-composites. For instance, cellulose fiber hydrogen bonding interactions were found to vary upon moisture absorption, leading to development of a shape memory property of the resulting printing composites (Mulakkal *et al.* 2018; Irvin *et al.* 2021). For another route, semi-crystalline PLA and PHA have shape memory functions, giving them potential for 4D printing of continuous cellulose fibers reinforced PLA or PHBV bio-composites with thermal-responsive shape memory behaviors (Mahdavi and Zolfaghari 2024; Kim *et al.* 2005). For instance, different types of continuous fibers are incorporated into biodegradable PLA matrix (Dezaki and Bodaghi 2024), and 3D-printing bio-composites have been found to display an outstanding increase in recovery forces, up to nine times.

Regeneration of lignin carbon fibers

From the biorefinery perspective, three components in natural plant fibers can be separated into individual cellulose, lignin, and hemicellulose *via* microwave liquefaction (Xie *et al*. 2016). In addition to cellulose fibers, another primary component of lignin with aromatic structures has potential to develop high-temperature resistance lignin carbon fiber as a potential alternative to polyacrylonitrile (PAN)-based carbon fiber (Song *et al*. 2015). Lignin as a renewable resource has been heavily studied for the development of lignin-based carbon fibers (Mohanty *et al.* 2018). Lignin-based carbon fibers *via* melting and wet-spinning methods are considered as a promising alternative to PAN-based carbon fibers in the development of reinforced sustainable composites. For instance, the lignin carbon fibers on a mass scale were reported, as shown in Fig. 4b (Baker *et al.* 2012). In addition to the preparation approaches, regeneration solvents are critical in the development of lignin-based carbon fibers. For instance, ionic liquid and deep eutectic solvents produced lignin carbon fibers, as shown in Fig. 4a (Zahra *et al.* 2022). These lignin carbon fibers are ideal alternatives to PAN-based carbon fiber for continuous fiber 3D printing, and the resulting carbon fiber reinforced engineering plastic composites have potential for use as high-strength materials, *e.g.*, lignin carbon fiber reinforced PPS, PI, and PEEK composites. However, lignin-based carbon fiber process and its production in a pilot scale have some bottlenecks. For producing lignin-based carbon fibers, lignin raw materials need to be in high purity, but the normal kraft lignin does not generally meet quality requirements. Additionally, carbonization is a crucial step to producing carbon fibers. The glass transition temperature (T_g) of the lignin is far below the temperature required for carbonization, so

the lignin must be pretreated to prevent its softening by oxidation (*e.g.*, air oxidation) to increase its T_g. However, it is tricky to control the rate of heating. Inadequate control of the heating rate causes lignin fibers to directly enter a liquid-rubbery state, such that they then fuse together, and they then lose their fibrous characteristics. Besides, the lignin-based carbon fibers are relatively weak in mechanical properties (tensile strength and modulus) compared to PAN-based carbon fibers as the tensile moduli of PAN-based carbon fibers increase with temperature during graphitization, while lignin-derived carbon fiber moduli stay the same or decrease (Sagues *et al.* 2019). Therefore, no continuous lignin carbon fiber 3D printing of reinforced polymer composites has been reported, while it is a promising topic for future studies.

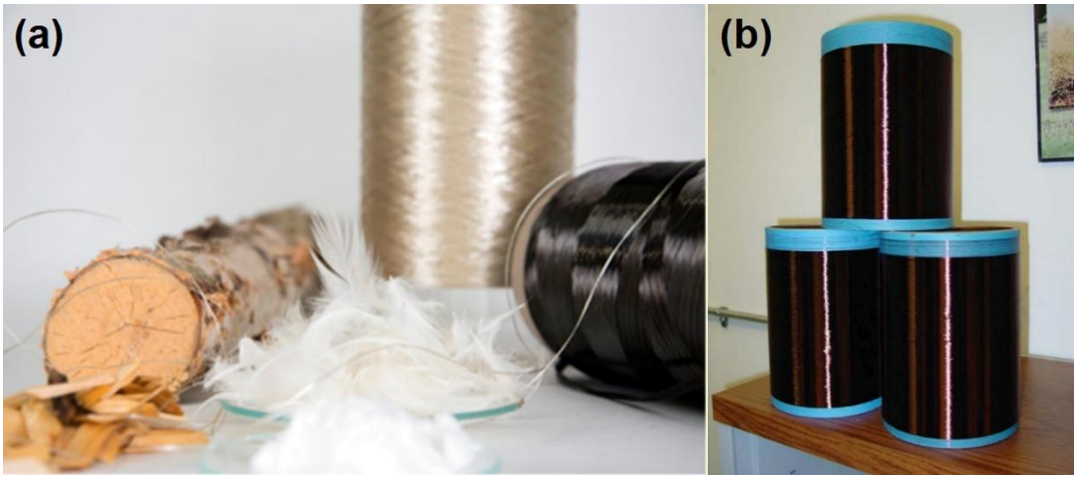

Fig. 4. Potential 3D printing continuous lignin carbon fiber from wood derived hybrid keratin and cellulose (a) and lignin (b) (Zahra *et al.* 2022; Baker *et al.* 2012). Figure republished with permission from Wiley Publishing Co. and American Chemical Society.

Regeneration of polyester fibers

As commercial polyester fabrics and waste polyethylene terephthalate (PET) bottles have the same chemical formula, a natural question is whether these waste PET bottles as raw materials can be converted into polyester fibers. As large amounts of waste PET bottles are generated each year (Lei *et al.* 2009), these plastic packaging wastes cause serious environmental pollution issues. Therefore, it is a good idea to develop continuous regeneration polyester fiber 3D printing of reinforced polymer composites if these waste bottles could be converted into polyester fibers *via* melting spinning. Different grades of PET waste (*e.g.*, water and soda bottles) were recycled for polyester fibers according to recent literature. These recycling polyester filaments have been successfully used for continuous regeneration fiber 3D printing (Bex *et al.* 2021; Sadeghi *et al.* 2021; Ror *et al.* 2023). Additionally, 3D printing self-reinforced fiber composites, also named single-polymer composites, have potential as alternative approaches to address plastic pollution and recycling issues. For instance, 3D printing polyphenylene sulfide PPS fiber self-reinforced PPS composites have been investigated, and the resulting composites maintained excellent mechanical performance after recycling a few times (Zhang *et al.* 2023a,b). In another case, the recyclability of additively manufactured continuous fiber reinforced composites has been demonstrated with ultra-high-molecular-weight polyethylene fibers reinforced high-density polyethylene (Tian *et al.* 2017; Zhang *et al.* 2021). As an alternative perspective, the polymer matrix filaments are also possibly

produced from waste plastics. The general protocols to produce regenerated waste plastic filaments are by a single- or twin- screw extruder with fixed die diameters (*e.g.*, 1.75 mm) and controlled temperatures along with post-cooling steps (*e.g.*, air or cold water). The resulting regeneration filaments from waste plastics work for FDM 3D printing then. For instance, post-industrial waste thermoplastics are ideal sources in the development of 3D printing of continuous fibers reinforcing sustainable composites, *e.g.*, recycling polymer filaments from waste ABS, HIPS, PP, and polycarbonate PC. For one case study, recycling waste ground tire rubber modified PU filaments in FDM 3D printing has been demonstrated (Badini *et al.* 2024).

Mechanical Properties Under Extreme Temperature Conditions of 3D Printing of Continuous Fiber Reinforced Composites

For a number of applications, continuous fiber 3D printing of reinforced composites has demonstrated notable performance under extreme temperature conditions, such as cryogenic conditions and temperatures higher than 500 ℃. It is highly desirable to investigate extreme mechanical performance of continuous fiber 3D printing of reinforced polymer composites and develop these composite materials for serving in advanced applications (Liu *et al.* 2022; Sumesh *et al.* 2024), *e.g.*, aircraft structure materials. However, almost all mechanical properties of continuous fiber 3D printing of reinforced polymer composites have been reported only for room temperature. Table 4 summarizes the typical studies about mechanical properties under the cryogenic temperature of 3D printing of continuous fiber reinforced polymer composites.

Table 4. 3D Printing of Continuous Carbon Fiber Reinforced Polymer Composites: Mechanical Properties under Cryogenic Environmental Conditions

Matrix and Fiber	Cryogenic Temperature Conditions	TS (MPa)	FV (%)	YM (GPa)	Literature
Nylon/Carbon Fiber	-173 to -195 °C	212.6	30.0	10.5	Siddiqui *et al.* 2023
Nylon/Carbon Fiber	-196 °C	45.8	NA	5.6	Huang *et al.* 2024

Tips: TS: Tensile Strength; FV: Fiber Volume; YM: Young's modulus

A typical setup of *in-situ* mechanical test under various temperatures ranging from -190 to 500 °C in an insulated environment chamber is shown on Fig. 5 (Meng *et al.* 2020), where the cryogenic temperature condition is achieved by using liquid nitrogen. For instance, the cryogenic mechanical property of continuous carbon fiber 3D printing of reinforced ABS composites was studied (Bartolome *et al.* 2017), and results indicated a brittle fracture mode from tensile testing at 77 K.

In addition to the conventional tensile or bending mechanical performance of 3D printing fiber reinforcing composites, its wear or tribological mechanical performance under extreme temperature conditions is important in aerospace applications (Palanisamy *et al.* 2024). A typical setup of *in-situ* tribology test under various temperature conditions from -190 to 1000 °C is shown in Fig. 6 (Lan *et al.* 2018). Some studies have reported on tribology performance of fiber reinforced composite materials by conventional hot-press lamination manufacturing or vacuum assisted resin transfer molding as non-additively manufacturing, but little literature is available on extreme temperature mechanical

properties of continuous fiber 3D printing of reinforced composites. Therefore, it is highly recommended to further investigate it in the future.

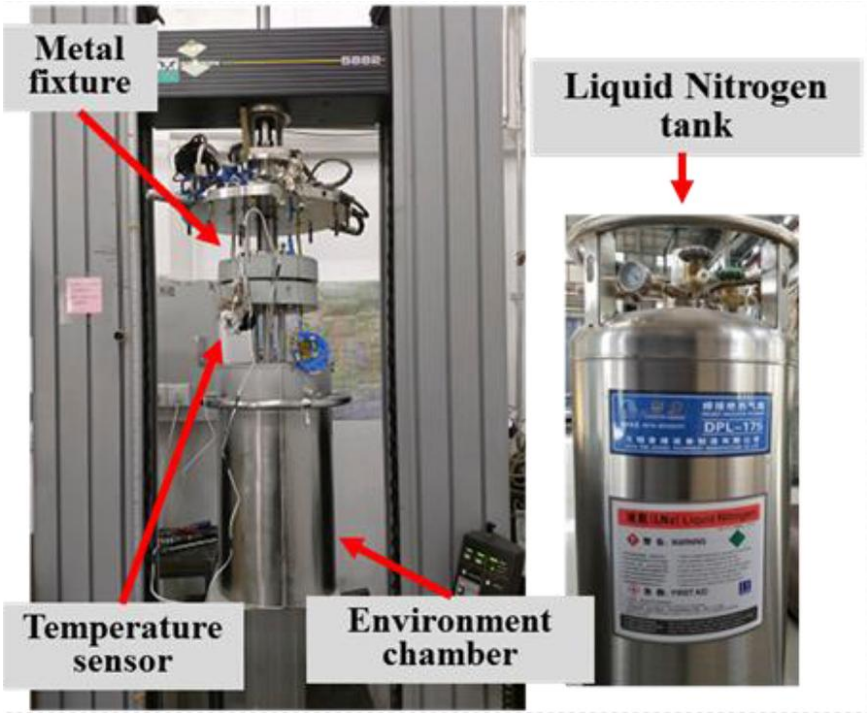

Fig. 5. Experimental setup of *in-situ* mechanical tests under various temperatures (Meng *et al.* 2020). Reprinted with permission from Elsevier Publishing Co.

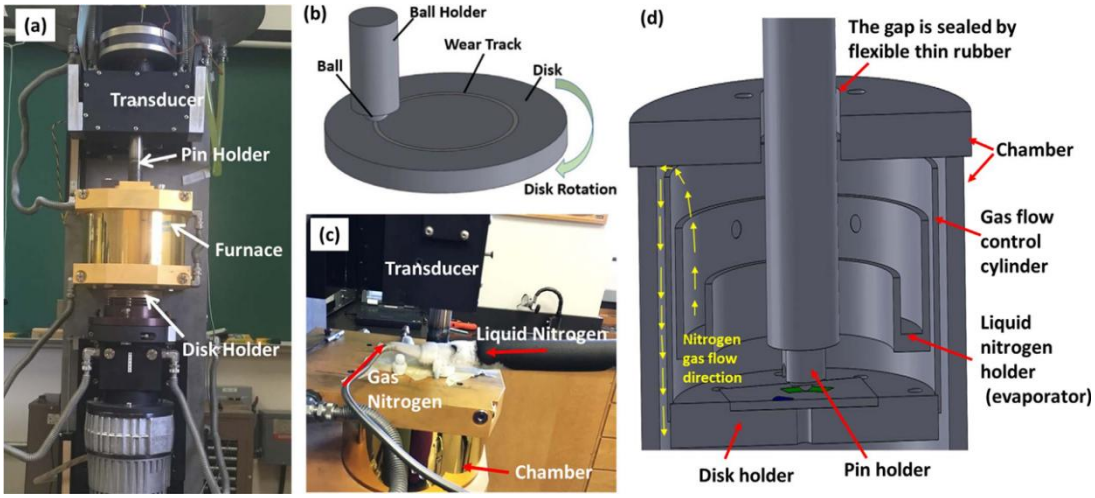

Fig. 6. Tribology test instrument setup for various temperatures (Lan *et al.* 2018). Reprinted with permission from Elsevier Publishing Co.

CONCLUSIONS AND FUTURE PROSPECTS

This critical review has focused on the continuous natural and regeneration fibers 3D printing of reinforced polymer composites and their mechanical performance under

extreme temperature conditions. Future work is needed to fill this gap of mechanical properties of continuous fibers 3D printing of reinforced polymer composites under extreme temperature conditions. Additionally, most of the published studies have been performed on thermoplastics, while engineering plastics have unique merits such as high temperature resistance performance. Thus, it is recommended to carry out future studies with poly(ether sulfone) (PPS), poly(ether-ether ketone) (PEEK), and polyimide (PI). Besides, more mechanical studies of continuous fibers reinforced engineering composites need to be further conducted, especially under high temperature, vacuum, or pressure conditions. Furthermore, for the current commercial engineering resins, they still have limitations to meet the special application requirements. There is a large room for further development of high temperature resistant PI, PPS, or PEEK engineering resins for continuous Kevlar, carbon, or basalt fiber 3D printing of reinforced composites. Moreover, follow-up study is recommended regarding assessments of 3D printing of continuous fibers reinforced polymer composites from multi-aspects, *e.g.*, mechanical performance (conventional and extreme environmental conditions) and their LCA and TEA sustainable analysis quantitatively (Bianchi *et al.* 2022; Andreozzia *et al.* 2024). Combined experimental and machine learning simulation characterizations of additively manufactured continuous natural fiber reinforced polymer are highly desirable (Palanisamy *et al.* 2025). For instance, the synchrotron X-ray tomography and *ex situ* or *in situ* synchrotron wide-angle and small-angle X-ray scattering in the Center for Advanced Microstructures and Devices at Louisiana State University, Baton Rouge, LA, USA along with small-angle neutron scattering at Oak Ridge National Laboratory, Oak Ridge, TN, USA are ideal tools in elucidating printing-structure-property relationships of continuous fiber 3D printing of reinforced composites.

Almost all case studies in terms of continuous fibers reinforced bio-composites have merely focused on the PLA matrix. PHA, poly(3-hydroxybutyrate-co-3-hydroxyvalerate) PHBV, and polyhydroxybutyrate PHB are promising alternative biopolymers in continuous fiber 3D printing of reinforced bio-composites, as shown in Fig. 7 (Ehman and Area 2021). PHBV, as a novel shape memory polymer, has potential for the continuous natural fiber 4D printing of reinforced bio-composites.

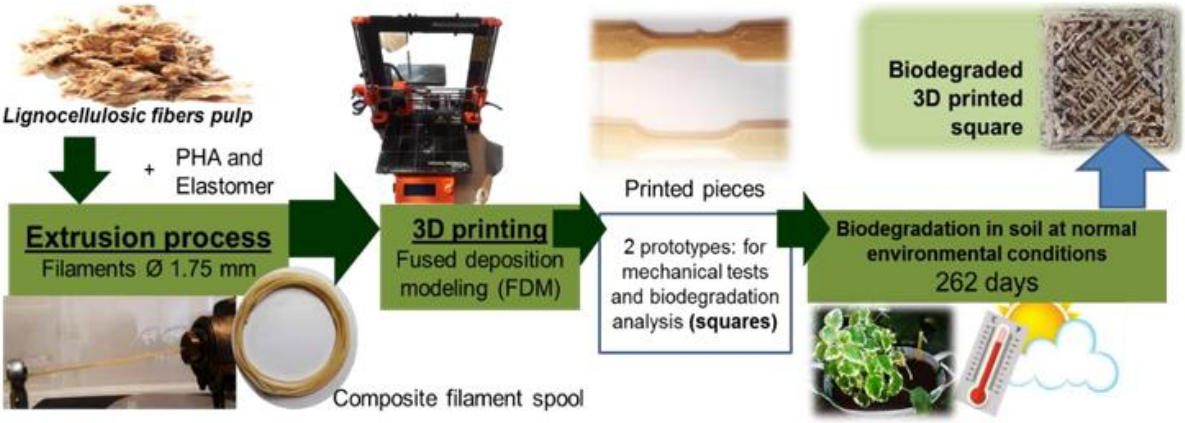

Fig. 7. FDM 3D printing of cellulose fiber reinforced PHA bio-composites (Ehman and Area 2021). Reprinted with permission from the authors.

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