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Recycled, Bio-Based, and Blended Composite Materials for 3D Printing Filament: Pros and Cons—A Review

Khanh Q. Nguyen[1], Pascal Y. Vuillaume[2], Lei Hu[2], Jorge López-Beceiro[3], Patrice Cousin[1], Saïd Elkoun[4], Mathieu Robert[1]

[1]Department of Civil & Building Engineering, University of Sherbrooke, Sherbrooke, Canada
[2]COALIA, Thetford Mines, Canada
[3]Centre for Research in Naval and Industrial Technologies (CITENI), Ferrol Industrial Campus, University of A Coruña, Ferrol, Spain
[4]Department of Mechanical Engineering, University of Sherbrooke, Sherbrooke, Canada

Email: Quoc.Khanh.Nguyen@USherbrooke.ca, pvuillaume@coalia.ca, lhu@coalia.ca, jorge.lopez.beceiro@udc.es, Patrice.Cousin@USherbrooke.ca, Said.Elkoun@USherbrooke.ca, Mathieu.Robert2@USherbrooke.ca

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Abstract

In recent years, additive manufacturing (AM), known as "3D printing", has experienced exceptional growth thanks to the development of mechatronics and materials science. Fused filament deposition (FDM) manufacturing is the most widely used technique in the field of AM, due to low operating and material costs. However, the materials commonly used for this technology are virgin thermoplastics. It is worth noting a considerable amount of waste exists due to failed print and disposable prototypes. In this regard, using green and sustainable materials is essential to limit the impact on the environment. The recycled, bio-based, and blended recycled materials are therefore a potential approach for 3D printing. In contrast, the lack of understanding of the mechanism of interlayer adhesion and the degradation of materials for FDM printing has posed a major challenge for these green materials. This paper provides an overview of the FDM technique and material requirements for 3D printing filaments. The main objective is to highlight the advantages and disadvantages of using recycled, bio-based, and blended materials based on thermoplastics for 3D printing filaments. In this work, solutions to improve the mechanical properties of 3D printing parts before, during, and after the printing process are pointed out. This paper provides an overview on choosing which materials and solutions depend on the specific application purposes. Moreover, research gaps and opportunities are mentioned in the discussion and conclusions sections of this study.

Keywords

Additive Manufacturing, 3D Printing, Fused Filament Deposition (FDM) Manufacturing, Recycled, Bio-Based, Blended Materials, Interlayer Adhesion, Surface Modification

1. Introduction

3D printing is a form of additive manufacturing (AM) technique, that has gained popularity in the last few years due to its simplicity, inexpensive cost, and customizability [1] [2] [3]. This technology allows for quick and cheap productions with specific shapes without requiring a die or mold compared to the traditional manufacturing process [4] [5]. Although the invention of the inkjet printer was the beginning of 3D printing in the 1970s, it was not until the 1980s that people started printing materials instead of ink. The first 3D printer was created by Charles Hull (1986) with the patent for stereo-lithography (SLA) to create objects by building layers of materials from computer-aided design (CAD) software [6]. However, this technology was applied to limited areas such as medical and engineering. Nowadays, 3D printing has become more popular in many industries, including food [7], construction [8], automotive, aerospace, and military [9] [10]. Despite the numerous advantages, a considerable amount of waste still exists due to failed print and disposable prototypes. In this context, the use of recycled, bio-composite materials, and polymer blends for 3D printing is the optimal solution to limit the impact on the environment. Furthermore, the reuse of recycled waste after 3D printing is a trend in recent years. A recycling code model has been developed by Hunt *et al.* [11] to identify resin after 3D printing. Codes, as recycling symbols, were printed on the surface of products to recognize the plastic blend after printing. Polymer-based products play an important role nowadays [12] [13]. However, they are one of the main issues affecting the environment, including land [14], water [15], and air pollution [16]. Using recycled polymeric materials, therefore, is an efficient way to reduce plastic waste and the dependency on natural resources [17]-[22]. Unfortunately, the reuse of polymeric materials causes the loss of their properties after several recycling times [23]. The presence of contaminants during the recycling process is the main challenge for recycled polymeric materials [24] [25]. For recycled polymeric materials used in 3D printing filament, the limited category of materials, a lack of standardization, testing procedures, and technologies in product quality control still exists and therefore need to be solved [26] [27] [28] [29]. In contrast, bio-composite (polymers matrix with natural fiber) is well-known to be a material with superior properties compared to pure material and potential material for 3D printing filament [30] [31] [32] [33] [34]. Composites based on natural fibers exhibit various advantageous properties such as lightweight, high strength, good stiffness, increased biodegradability, and eco-friendly materials [30] [35]

[36] [37]. However, the high-temperature extrusion during printing process could decompose natural fibers. Poor adhesion between layers and porosity are found, and therefore interfacial bonding between the fibers and matrix problems arise [31] [38] [39]. Although it holds tremendous potential, further development and testing are needed to better improve the properties of recycled and bio-composite materials suitable for 3D printing filament.

As concern to green and sustainable materials, this article presents an overview of recycled, bio-based, and blended materials suitable for 3D printing filament. The main objective is to identify the advantages and disadvantages of using recycled, bio-based, and blended materials. The challenges, solutions as well as research opportunities of these materials are mentioned. This study is laid out as follows. Section 2 presents an overview of the fused deposition modelling (FDM) technique of 3D printing and material requirements for 3D printing filament. Recycled, bio-based, and blended materials suitable for 3D printing filaments are presented in Section 3. This is followed by recommended solutions to improve the properties of recycled, bio-based, and blended filaments in section 4. Sections 5 and 6 consist of the discussion, research opportunities, and the paper's main conclusions.

2. FDM and Material Requirements for 3D Printing Filament

A variety of 3D printing technologies have been developed for specific purposes such as rapid prototyping, reduced manufacturing time and cost, controlled microstructure, the use of a vast range of materials, and excellent mechanical properties. The main techniques of polymer 3D printing are SLA [6] [40] [41], Digital Light Processing (DLP), powder bed fusion (SLS—selective laser sintering, MJF—multi jet fusion) [42] [43] [44], 3D plotting [45] [46], FDM/Fused Filament Fabrication (FFF) [47] [48], PolyJet/MultiJet modeling (PJM/MJM) [49], Laminated Object Manufacturing (LOM) [2] [50]. Depending on the type of materials used for 3D printing, the specific methods are used respectively in additive processing: liquid resin (SLA, DLP, PJM/MJM), polymer powder (SLS, MJF), and polymer films, pellets (LOM) [2]. For 3D printing filament, however, FDM is the most widely used technique to produce thermoplastic polymers and their composites. The FDM technique, also known as FFF technique, was developed by Stratasys Company in the 1990s [51]. However, in recent years, it has received attention in various segments, including biomedical engineering [52] [53], tissue engineering [54], electronics [55] [56], pharmaceutical [57], automotive, and aircraft [58] [59]. In the FDM technique, a continuous filament of materials is used to produce 3D print materials layer by layer (**Figure 1**). First, the filament is rolled into a spool. It is then pushed toward the extrusion head by drive wheels. The extrusion head controls the feeding and retracting of filaments in precise amounts. The filament is then heated and extruded on the platform through the nozzle. At the extrusion stage, the material changes from solid to a semi-liquid state to create layers upon layers. Finally, the layers stack on top of

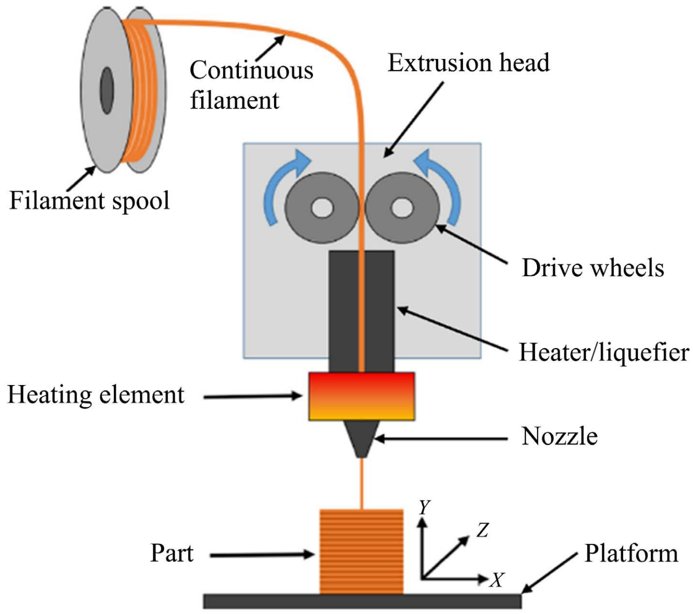

Figure 1. Fused deposition modelling (FDM) process (adapted from Stansbury and Idacavage [2]).

each other, and they are fused as the material hardens almost immediately. In order to create a 3D production, the extruder moves on the x-y axis while the platform moves on the z-axis. Furthermore, raster angle is one of the manufacturing parameters that play a key role in the FDM process. It is defined as the angle of the raster tool path of the nozzle with respect to the x-axis of the printing platform [51] [60] (**Figure 2(a)**). **Figure 2(b)** shows the typical raster angles (0˚, 15˚, 30˚, 45˚, and 90˚) for the FDM printing filament [61]. However, several limitations still exist regarding the quality of printing materials using the FDM method, including poor interlayer adhesion, high porosity, inferior mechanical properties, dimensional inaccuracy, defects and void formation, and undesirable residual stresses [62] [63] [64] [65]. Many previous studies have optimized process parameters to improve the quality of 3D printed products, including layer thickness [66] [67], nozzle size and temperature, raster angle (build orientation) [68], raster width, thermal processing conditions [69], and printing speed [70]-[77]. However, this review paper provides an overview of suitable materials that can be used for 3D printing filaments, specifically recycled and bio-composite materials. This review looks into the origin of the poor quality of the 3D printed material regarding the original properties of the material (contaminants, homogeneity and dispersion, viscosity, pore formation, melting temperature, fiber orientation) rather than the 3D manufacturing parameters. Solutions to improve 3D printed parts, including polymers modification, surface modification, minimizing void formation, etc., will be mentioned in this review article.

As mentioned earlier, recycled and bio-composites are potential materials for 3D printing filaments. In general, the following material requirements need to be met in order to produce the desired printed parts.

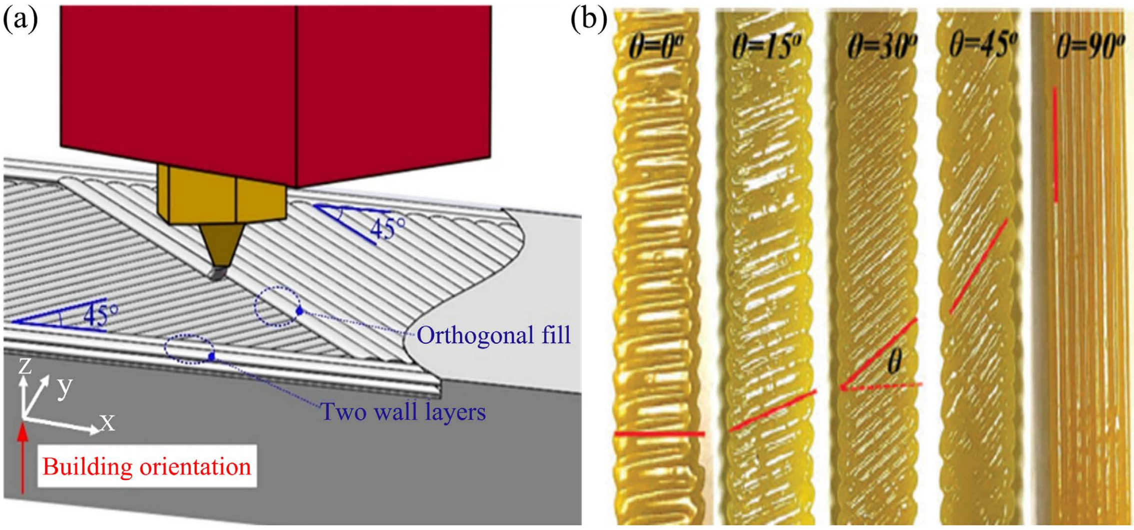

Figure 2. (a) FDM printing path (adapted from Ding *et al.* [60]) and (b) raster angles for FDM printing filament (adapted from He *et al.* [61]).

- ***Low melting point and viscosity.*** Under low pressure applied with FDM, low melting point and viscosity allow materials to be easily extruded from the nozzle [78] [79].
- ***Low glass transition temperature* (50˚C - 95˚C) *and slight material shrinkage.*** The low glass transition temperature (T_g) improves the extrusion process. As the material hardens almost immediately after printing, low material shrinkage could improve adhesion between layers [80] [81]. In other words, a low coefficient of thermal expansion (CTE) reduces internal stresses during the cooling process [82].
- ***Possibility of deformation under high temperature*** [81]. The decomposition of natural fibers in bio-composites could be occurred under high temperature.
- ***Enough stiffness.*** Material is fed as a filament for printing, the high stiffness of the material optimizes the feeding process [83].
- ***High thermal conductivity* (2 - 12 W/m·K).** Materials with high thermal conductivity raise heat distribution, resulting in high bonding between the filaments, and therefore mechanical properties are improved. High thermal conductivity comes along the sintering and melting process [79] [84] [85].
- ***Low contamination* (*for recycled materials*), *homogeneous filament*, *high dispersion*, *and high orientation fiber* (*for bio-based and blended materials*)** [68] [78].
- ***Non-toxic*** [81].

3. Recycled, Bio-Based, and Blended Materials Suitable for 3D Printing Filaments

3.1. Recycled Polymeric Materials

Thermoplastic polymers are the most common materials used in FDM 3D printing filament. Currently, common plastics are considered potential recyclable materials for 3D printing filament, including polylactic acid (PLA), acrylonitrile butadiene styrene (ABS), polyethylene terephthalate (PET), high-density polyethylene (HDPE), polypropylene (PP), polystyrene (PS) and high impact polystyrene (HIPS).

3.1.1. Recycled Polylactic Acid (PLA)

Polylactic acid (PLA) is a biodegradable and recyclable thermoplastic produced from renewable resources such as plant starch [78]. It is a semi-crystalline thermoplastic with an extrusion temperature of 160˚C - 220˚C. PLA is well-known as an easy material to deal with during 3D printing. It is considered as an environmentally friendly material, biodegradable and biocompatible, with good processability and low-cost production. It can be printed at high speeds and faces fewer shrinkage issues as compared to other materials. With the FDM technique, a hot printing bed is not required for PLA [86]. However, PLA is susceptible to degradation during its use, melting, and the recycling processes [87] [88] [89]. The thermal, mechanical, and fracture behaviors of PLA processed by extrusion were investigated by Nascimento *et al.* [90]. They concluded that a single recycling process resulted in only a few structural changes rather than altering significantly material performance. In contrast, repeated processing cycles resulted in the degradation of PLA. The chain scissions occurred after seven processing cycles. Reduced molecular weight led to a decrease in stress and strain at break, and Young's modulus of PLA [91] [92]. The number of extrusion cycles has a significant effect on the tensile, impact strength, and the cold crystallization temperature (Tcc) of PLA. However, there is no effect on the T_g [93] [94]. For 3D printing PLA filament, the recycled materials present similar properties to the virgin materials. As a result, tensile strength, modulus, and hardness decreased, but shear strength of recycled materials increased [95].

3.1.2. Recycled Acrylonitrile Butadiene Styrene (ABS)

Acrylonitrile butadiene styrene (ABS) is well-known as an amorphous copolymer with good mechanical properties, such as heat resistance, high rigidity, toughness, and impact strength. A hot printing bed is required for ABS with an extrusion temperature of 215˚C - 250˚C. Furthermore, ABS faces shrinkage and warping problems during printing [86]. It is a non-biodegradable and material capable of emitting toxic smoke during 3D printing. It can be pointed out that repeated extrusion cycles (up to five) significantly affect the impact strength of ABS rather than its tensile properties [18] [96]. Based on the results of Mohammed *et al.* [97], the flow rate of recycled ABS filaments was relatively unchanged with increasing extrusion temperature. The print quality of recycled ABS filaments was similar to the commercial ones. However, a 13% - 49% decrease in ultimate strength was found in samples printed from recycled ABS filaments. Sharing the same point of view, Charles *et al.* [98] concluded that the tensile and impact strengths of recycled ABS filament were similar to virgin ABS after two recycled cycles. A slight change in polymer viscosity was observed, resulting in a small improvement in print quality. Furthermore, T_g was mostly constant during the extrusion process.

3.1.3. Recycled Polyethylene Terephthalate (PET)

Polyethylene terephthalate (PET) is a semi-crystalline thermoplastic with an extrusion temperature of 212˚C - 235˚C. For 3D printing PET filament, a heated bed does not require. PET is fairly hard, recyclable, and odorless when printing. This material has good properties such as good tensile, impact strength, and thermal stability. However, PET is not widely used for FDM printing, because of its high melting temperature, water absorption, and low crystallinity. Currently, available PET filaments are recycled PET and glycol-modified PET (PET-G). PET-G is an amorphous plastic, where the ethylene glycol chain is replaced by cyclohexanedimethanol, leading to reduce its brittleness [99]. PET-G is durable, biocompatible, flexible, recyclable [100], and easy to deal with during 3D printing filament compared to PET [101]. For 3D printing filament, PET-G is considered a material possessing good properties of ABS (durability, high strength) and PLA (biodegradable, high flexibility) [100]. Therefore, PET-G and recycled PET are considered an alternative to virgin PET for 3D printing. Zander *et al.* [102] concluded that the molecular weight and the viscosity of recycled PET filaments were unchanged after two extrusion cycles. However, it is difficult to achieve uniform diameters for filaments, due to the low viscosity of recycled PET. Furthermore, Schneevogt *et al.* [99] compared the potential of using recycled PET and PET-G filaments for 3D printing. They pointed out that there was a small variation in the linear region of stress-strain curve, but a significant difference in the non-linear region between PET-G and recycled PET. Specifically, ductility failure was found in PET-G, whereas recycled PET showed a tendency for brittle failure in the non-linear region. They, therefore, recommended using these materials only for engineering designs and avoiding non-linear deformation.

3.1.4. Recycled High-Density Polyethylene (HDPE)

High-density polyethylene (HDPE) is a semi-crystalline thermoplastic with excellent properties such as high tensile strength, stiffness, a rather low melting point (~120˚C), and highly crystalline. It is a lightweight material, flexible, easy to dye and mold, non-water absorbent and chemical resistant [20]. Recycled HDPE is available for filament extrusion because many packaging products (e.g., detergent bottles and milk jugs) are made from it. Moreover, an extrusion temperature is controlled at 180˚C - 190˚C for recycled HDPE. Unlike PLA and ABS, HDPE is, however, rarely used in 3D printing filaments. High-temperature nozzle and heating bed are required for HDPE. Furthermore, poor adhesion, shrinkage/warping, as well as stress-induced deformation during printing are found in recycled HDPE [103]. However, several changes in FDM process parameters or polymer modification could improve the quality of the printed parts.

A base layer or adhesion tools (raft, brim) will improve the adhesion of the printed part to the printing surface. Recycled HDPE was successfully printed for the first time for the functional boat by Washington Open Object Fabricators (WOOF) using the FDM technique. They built a unique build plate using a fused-HDPE surface. The extruder was equipped with a heater to heat the previous layer. The CAD model was then adjusted to print a sacrificial flange with the boat to avoid warping [103] [104]. Moreover, Baechler *et al.* [105] concluded that recycled HDPE filament could be fed consistently into a 3D printer with a constant extrusion rate.

3.1.5. Recycled Polypropylene (PP)

Polypropylene (PP) is another semicrystalline thermoplastic widely used in industry and in the manufacturing of everyday objects along with HDPE, and PET. PP has good properties such as good chemical, abrasion, fatigue, and environmental stress crack resistance, shock absorbing, relative rigidity, and flexibility [106]. However, PP has a low-temperature resistance and is sensitive to UV rays, resulting in its susceptibility to thermal expansion. For 3D printing filament, recycled PP has recently gained interest due to its availability and recyclability. PP can be recycled up to four times by thermal processing without much alteration of its properties. Vidakis *et al.* [107] concluded that the tensile properties of PP were affected by thermal stress after six recycling cycles. In contrast, flexural behavior could be improved; impact strength and microhardness could be maintained under repeated thermal reprocessing. Recycled PP is, however, still not widely used for making 3D printing filaments due to its warping and poor interlayer adhesion issues. The significant diameter variation and elliptical shape are found in recycled PP filament. Furthermore, recycled PP presents a considerably high flow as compared to ABS and PLA [106]. Recently, Kumar *et al.* [108] pointed out that the ideal temperature was 210˚C - 230˚C for printing PP using the FDM technique. Atsani and Mastrisiswadi [109] optimized the extrusion process conditions such as spooler and extrusion speeds to obtain recycled PP filaments. The results showed that the rough and curved surface of filaments still exists. As mentioned earlier, the mechanical properties of recycled PP are relatively unchanged during recycling cycles. Therefore, recycled PP is a potential material for 3D printing in the future.

3.1.6. Recycled Polystyrene (PS) and High Impact Polystyrene (HIPS)

Polystyrene (PS) is an amorphous polymer of high clarity, hard, but rather brittle. PS can be found in the packaging and insulation applications, disposable cups, and bowls. PS is considered a difficult material to recycle due to the high cost of transport [110]. It is often not recycled locally but must be transferred to a recycling facility, leading to increased recycling costs and investment capital for companies [111]. In fact, PS foam is completely recyclable for FDM filaments production. However, the recycling rate of PS foam is still relatively low [112]. For 3D printing filament, high-impact polystyrene (HIPS) is commercially available. HIPS is similar to ABS with high impact resistance. However, it is a biodegradable material and easy to fabricate. For 3D printing filament, the extrusion temperature is 190˚C - 210˚C, and a heated printing bed is required for HIPS [78]. Recently, Ng *et al.* [110] compared the properties of recycled PS foam with those of HIPS for FDM filaments. As the result, 45% higher tensile strength and 52% greater stiffness were found in recycled PS compared to HIPS. Recycled PS exhibited a more brittle tendency than HIPS. Furthermore, recycled PS filament showed lower viscosity than HIPS at high temperatures. Based on the results of this study, recycled PS could be a potential material to produce 3D printing filaments. Another study on the properties of recycled and virgin PS filaments has been conducted [113]. The results showed that the fracture surface of recycled PS filaments was uniform and without defects after the tensile test. The Tensile strength and T_g of recycled PS were lower than those of virgin PS. The melt flow index (MFI) was, however, similar for both recycled and virgin PS filaments.

3.2. Bio-Based and Blended Composite Materials

Fiber-reinforced polymer composites and thermoplastic-based composite blends are a promising material to make 3D printing filament for the FDM technique [28]. These materials are formed of bio-based fillers and polymeric matrix, which can also be recycled. Bio-composite and blended materials show improved mechanical properties, such as higher modulus and tensile strength than neat thermoplastic materials [68].

3.2.1. PLA-Based Composites

The fillers used for PLA-based composites are typically cellulose and natural fibers. The use of wood fibers as reinforcement in PLA polymer was developed for 3D printing filament. Kain *et al.* [114] concluded that the mechanical properties of wood fiber/PLA composites were improved by adding wood fibers up to 25 wt% compared to pure PLA. However, the authors pointed out that the mechanical performance was dependent on the infill orientation of fibers. Furthermore, increasing the print width reduces the cohesion of the wood fiber/PLA, resulting in a decrease in tensile strength and an increase in water absorption [31]. The effect of lignin on the thermal and mechanical properties of lignin/PLA filament was also investigated. Gkartzou *et al.* [115] showed that adding 5 wt% lignin makes the composites more brittle and decreases the break elongation. In addition, a reduction in tensile strength and Young's modulus by 18% and 6%, respectively, were found compared to pure PLA. Recently, however, Long *et al.* [116] have successfully produced PLA composites with excellent properties for the FDM technique. Ethyl acetate treated lignin nanospheres (EALNSs) with a high specific surface and uniform size, were used to reinforce PLA. As a result, lignin nanoparticles can improve the melt flow and mechanical properties of 3D printing products. The flexural, tensile, and impact strength of EALNSs/PLA composites were increased by 130.8%, 56.1%, and 14.2%, respectively, by adding 0.5 wt% lignin nanoparticles. Recycled continuous carbon fibers from 3D printed parts were used to strengthen the material. The carbon fiber/PLA composite had 25% higher flexural strength as compared to the original printing composites [117]. However, there are still many challenges in achieving good quality of printed products through the FDM technique. Heidari-Rarani *et al.* [118] modified the processing conditions to obtain a reliable print such as using polyvinyl alcohol (PVA) solutions to increase the cohesion between carbon fiber/PLA, rapid cooling of printed composites, the simultaneous injection of carbon fiber and melted PLA. As a result, the tensile and bending strength of printing composites were increased by 35% and 108%, respectively, as compared to pure PLA. The result demonstrated that there was a good adhesion between the interface of carbon fiber and PLA. Contrariwise, the main failure mode of carbon fiber/PLA composite was the delamination. In addition, basalt fiber reinforced PLA filament was also studied by Yu *et al.* [119]. They have successfully printed basalt fiber/PLA composite via the FDM technique with a lighter weight and better mechanical properties than those of conventional molding materials. It should be noted that the voids (inter- and inner-filament voids) still exist during the 3D printing process. Fiber length and fiber orientation played a key role in the mechanical properties of composites.

3.2.2. ABS-Based Composites

For ABS-based composite, carbon fibers (CF) are considered typical filler. ABS reinforced with short carbon fibers of an average length of 150 μm was investigated for fabricating the 3D printed parts by the FDM technique [120]. The results showed that the tensile strength increased by 22.5% with 5 wt% carbon fiber as compared to the pure ABS. However, the porosity increased as the carbon fiber content was higher than 10 wt%. Yang *et al.* [121] also studied the continuous carbon fiber/ABS composite with carbon fiber content of 10%. The composite had greater tensile and flexural strength than neat ABS and similar properties to injection-molded ABS parts. The interlaminar shear strength, however, was only 2.8 MPa, a very small value as compared to the shear strength (24 MPa) of CF/ABS parts obtained by injection molding due to the poor interface. It should be pointed out that the use of short carbon fiber can reduce the distortion and warping of ABS [122]. Tekninalp *et al.* [123] also successfully printed short carbon fiber (with a length of 0.2 - 0.4 mm) reinforced ABS by the FDM technique. Sharing the same point of view, the authors reported that the tensile strength and modulus of FDM parts increased by 115% and 700%, respectively. Contrariwise, 20% of void formation was found in the composite sample due to the gaps of deposition lines and poor adhesion between carbon fibers and ABS matrix. However, the porosity of the CF/ABS composite parts can be reduced from the auxiliary heating process, by mounting an auxiliary heating plate on the printing head of the 3D printer [124]. It should be noted that this auxiliary heating should not exceed the degradation temperature of ABS. Furthermore, glass fibers (GF) were also used to reinforce ABS. Billah *et al.* [125] reported that the stiffness of GF/ABS composites increased by 84% compared to the neat ABS. In contrast, the composites had similar thermal stability to neat ABS. Moreover, natural fibers such as Kevlar, palm, bamboo, pine cone, and rice straw fibers are also potential fillers for ABS-based composites. The addition of Kevlar and carbon fibers to the ABS matrix improves its rigidity and ductility [126]. Filaments produced from ABS containing 15 wt% of palm fibers show 42% higher hydrogen bonding and similar T_g compared to the neat ABS [127]. The ABS matrix reinforced with chemically modified bamboo fibers shows reduced density, but the mechanical properties of the composites remain unchanged [128]. In addition, after chemical treatments by alkaline and bleaching, adding 2 - 5 wt% pine cone fibers into ABS matrix does not change the filaments diameter and density [129]. The rice straw fiber content (5 - 15 wt%) reinforced ABS was investigated by Osman *et al.* [130]. The results showed that the tensile and flexural strength of rice straw/ABS composites decreased as the rice straw fiber content increased, resulting in an increase in water absorption. Although natural fibers can reinforce ABS, their concentrations in composites are still low in current studies. Natural fiber/ABS composites generally present good properties with 5 wt% fiber content.

3.2.3. PET-Based Composites

Carbon fibers are commonly used to reinforce PET for parts obtained by the FDM technique. With a carbon fiber content of 15 wt%, the elastic modulus, tensile, and shear strength of CF/PET composites increase by 180%, 230%, and 40%, respectively, compared to neat ABS [131]. As mentioned earlier, commercially available PET filaments are made from recycled PET and glycol-modified PET (PET-G). Kichloo *et al.* [132] revealed that adding 20 wt% of carbon fiber into PET-G matrix resulted in a maximum of 43.7% and 25% in tensile and flexural strength, respectively, for honeycomb pattern. However, carbon fiber reinforced PET-G shows an increase in melt viscosity and a weaker interlayer bonding, resulting in a reduction of its mechanical properties compared to neat PET-G printed parts [133]. The optimization of printing parameters, therefore, was conducted. Post-processing was performed when the annealing temperature was higher than the T_g of PET-G [133]. The printed temperature of 250˚C, 0.1 mm of layer height, and 0.6 mm of nozzle diameter were reported to optimize CF/PET-G printed parts [134] [135]. As a result, an increase in mechanical properties, and a void content of 3% were found. In addition, the filament properties remained unchanged when virgin PET-G was replaced with recycled PET-G with 25 wt% of carbon fiber [136] [137]. Recently, Carrete *et al.* [138] revealed that post-consumer textile could be used to reinforce recycled PET (rPET) matrix. Using the surface modification technique (acid hydrolysis and silane functionalization), the cotton fibers made the melt flow index (MFI) of composite higher than neat rPET. Also, a ductile fracture of cotton fiber/rPET composites was found.

3.2.4. HDPE-Based Composites

As mentioned earlier, there are many problems with printing HDPE filaments such as poor adhesion, shrinkage/warping, and stress-induced deformation [139]. Very little information has been found in the literature about the 3D printing of HDPE composites by FDM. Recently, however, natural fibers (birch and wood fibers) reinforced HDPE have been discovered for fabricating 3D printed parts by the FDM technique [140] [141]. Koffi *et al.* [140] successfully printed birch fiber/HDPE composite by the FDM for the first time without significant warping, shrinkage, and other geometric deformation problems. The authors revealed that the composite was composed of HDPE matrix with 10 - 30 wt% of yellow birch fibers as filler and 3 wt% of maleic anhydride as a coupling agent. The results showed that shrinkage, warping, and geometric deformation of the composite were overcome. The deformation was reduced up to 80%, and Young's modulus increased up to 35% as fiber content increased compared to neat HDPE. In addition, Migneault *et al.* [141] also created a potential HDPE composite for 3D printing. 40 wt% of wood fibers were used to reinforce HDPE and 3 wt% of maleated polyethylene (MAPE) was used as a coupling agent. Observing surface chemical characteristics, the results showed that the strength of wood fiber/HDPE increased as the level of carbohydrates on the fiber surface increased. Moreover, Gregor-Svetec *et al.* [142] studied cardboard dust/HDPE composite materials for 3D printing filament. The authors pointed out that the high porosity, structure nonuniformity, decreased crystallinity, and lowered T_g, were found in composite filaments as cardboard dust content increased. As a result, a decrease in mechanical properties, tenacity, and elastic modulus was observed when 20 wt% cardboard dust was added. Nevertheless, cardboard dust/HDPE filament could be printed when the cardboard content was up to 50 wt%.

3.2.5. PP-Based Composites

Along with HDPE, PP is also not a typical material used for 3D printing by the FDM technique because of its warping and shrinking during the printing process. However, adding coupling agents made PP composite easily printable. Stoof and Pickering [143] pointed out that the maleated polypropylene coupling agent improved the mechanical properties of harakeke fiber (New Zealand flax)/PP composites. As a result, the tensile strength and Young's modulus of PP composites increased by 74% and 214%, respectively, compared to neat PP. After the printing process, however, these properties tended to decrease because of the stress relaxation of polymers. In contrast, with 30 wt% harakeke fiber content, the shrinkage of composite was reduced by 84%. Sharing the same point of view, Wang *et al.* [144] revealed that the incorporation of maleic anhydride polypropylene (MAPP) into cellulose nanofibril (CNF)/PP composite improved its mechanical properties. The results showed that the flexural strength and modulus of compatibilized PP composite containing of 10% wt CNF were improved by 5.9% and 26.8%, respectively, compared to neat PP. Also, Spoerk *et al.* [145] successfully printed carbon fiber/PP composite containing MAPP coupling agent. With 10 wt% carbon fiber content, a uniform filler dispersion, a good interface adhesion, and an increase in mechanical properties were found for its composites. However, the authors revealed that the printing orientation affected the mechanical properties of the composite. In contrast, Sodeifian *et al.* [146] found that adding maleic anhydride polyolefin (POE-g-MA) to glass fiber/PP composites resulted in a decrease in modulus and strength and an increase in the flexibility of the composite. In addition, surface modification is also a way to improve the properties of microcrystalline cellulose (MCC)/PP composites [147]. The use of n-octyltriethoxysilane to modify the MCC improved the dispersibility of the MCC into the PP matrix. The results showed that the filaments had a good surface finish, good mechanical properties, and easy printing. Furthermore, recycled PP (rPP) has also recently been interested in composite 3D printing [148] [149]. Rice husk/rPP filament had been successfully printed with a fiber content of 5 - 10 wt% [148]. The results showed that the composite density decreased, and its crystallinity increased as rice husk fiber content increased. The 3D printed part presented lower warping compared to printed neat rPP. The water absorption of composites increased because of the hydrophilic behavior of natural fibers. However, the study found that the fracture took place at the interface between the natural fibers and the rPP matrix [149].

3.2.6. PS-Based Composites

As mentioned earlier, PS foam is completely recyclable to produce FDM filaments. Recently, recycled PS (rPS) from post-used expanded polystyrene foam (EPS) has been investigated to produce FDM filaments [150]. 2.5 - 10 wt% of corn husk fiber was used to reinforce rPS for 3D printing. In addition, a layer of glue was added onto the surface of the print bed to improve the first layer adhesion. The results showed that the composite filament containing 10 wt% of corn husk fiber failed to be printed. This cause was explained by the premature thermal degradation and high melt viscosity of corn husk fiber/rPS composites. In contrast, rPS containing 2.5 - 7.5 wt% of fiber could be printable. However, the tensile strength and modulus of the composite decreased as corn husk fiber content increased. The dull and rough surfaces as well as a slight decrease in thermal stability were found for corn husk fiber/rPS composites. Moreover, cellulose nanocrystal/PS composite was also considered a potential material for 3D printing. Lin and Dufresne [151] performed the surface modification of cellulose nanocrystal using polyethylene glycol/polyoxyethylene (PEG/PEO) to reinforce the PS matrix. As a result, the mechanical and barrier properties of composites were improved. Therefore, cellulose nanocrystal/PS composite could be used for 3D printing.

3.2.7. Polymer Blend Materials

The difficulties encountered in printing polymers by FDM are not only explained by the high temperatures required for their transformation; the weakness of inter-layer adhesion can also lead to a significant drop in mechanical properties. A viable solution to this problem consists in reinforcing the polymer matrices by adding synthetic or natural fibers. However, while reinforcing fibers are less adhesive than thermoplastic polymers, interlayer adhesion is weakened by their presence. The mechanisms of polymer-fiber inter-layer adhesion for FDM printing are complex and the theoretical and experimental bibliography is still limited [152]. Considering that the adhesion mechanisms of polymer-polymer interfaces are now relatively well understood, polymer blends have recently been studied and successfully printed through the FDM technique.

Harris *et al.* [153] successfully produced PLA/PP blends with compatibilizer PE-g-MAH (polyethylene graft maleic anhydride) by the FDM technique. With 7.5 wt% PP content, the materials presented good thermal stability. Ausejo *et al.* [154] also found that the thermal stability of PLA/PHBV poly(3-hydroxybutyrate-co-3-hydroxyvalerate) blends was improved by self-compatibilization during the degradation of materials. The thermal stability of PLA/PHBV increased as PHBV content increased. In addition, PLA can be blended with other (co)polymers for 3D printing filament. PLA/S-*co*-MMA poly(styrene-*co*-methyl methacrylate) blends were recently studied [155]. The result showed that the thermal decomposition temperature of PLA/S-*co*-MMA blends was lower than that of amorphous PLA and poly(S-*co*-MMA). In contrast, the PLA-based blends had a higher Young's modulus than amorphous PLA. Another study also showed that PLA/PHBV/PBAT poly(butylene adipate-co-terephthalate) blends produced by FDM printing had a three-time higher tensile strength than those of injection specimens [156]. However, poor interphase adhesion still exists for polymers blend materials. The mechanical properties of polymer blends were dependent on the applied infill orientation [157]. FDM Filament based on PLA containing up to 20 wt% of natural rubber (NR) was produced [157]. The result showed that NR improved the elongation at break and impact strength using a linear infill parallel to the length of specimens. PLA/BioPBS (poly(butylene Succinate)) biobased filament with BioPBS content higher than 50 wt% was unprintable due to the high viscosity and low thermal stability of composites [158]. The results showed that the coefficient of linear thermal expansion (CLTE) decreased as BioBPS content increased in blend filaments. However, the ductility and crystallinity of PLA were improved by adding BioPBS. The 3D printed PLA/BioPBS with 10 wt% of BioPBS presented higher tensile and impact strength than neat PLA.

ABS blends are also considered a suitable material for 3D printing filament. Monofilaments were prepared from ABS/UHMWPE (ultrahigh molecular weight polyethylene) with SEBS (styrene ethylene butadiene styrene) as a compatibilizer [159]. As a result, monofilaments were successfully printed when the content of UHMWPE was less than 25wt% in the ABS/UHMWPE blend. The smoothest surface was found in 75:25:10 ABS/UHMWPE/SEBS. Furthermore, blending ABS with TPU (thermoplastic polyurethane) was also investigated [160]. The results showed that blends containing 10 - 20 wt% of TPU had improved inter-layer adhesion without loss in yield strength. In contrast, the blends maintained a good adhesion with 30 wt% TPU, while the yield strength was closed to that of neat TPU rather than ABS.

On the other hand, thermoplastics with poor 3D printability such as HDPE and PP were also successfully printed via blending with highly printable thermoplastics such as PLA and ABS [161]. For the first time, PLA/HDPE and PLA/PP blends without additives were successfully printed by Choe *et al.* [161] with optimized FDM printing parameters. Moreover, microfibrillar composites (MFCs) of PP/PET blends and PP/PS blends were successfully processed by the FDM technique [162] [163]. PET and PS could be stretched into fiber form and oriented along the deposition direction during the FDM process. Both PET and PS fibers enhanced the crystalline structure of PP, resulting in the superior mechanical properties of PP/PET blends and PP/PS blends, respectively, compared with neat PP.

4. Solutions to Improve the Properties of 3D Printing Filaments

Although recycled, bio-based, and blended materials are suitable for 3D printing filaments, the mechanical properties of FDM 3D printed parts are still low. Poor inter-layer adhesion, and many other technical challenges still exist compared to those made from the conventional methods. One of the limitations of FDM printing is the anisotropic properties of 3D printed parts. Many previous studies have optimized process parameters to improve the quality of 3D printed products such as nozzle size and temperature, raster angle, raster width, thermal processing conditions, and printing speed [66]-[77]. Contrariwise, the current overview investigates the effects of external approaches (surface modification, chemical crosslinking, and plasma treatment, etc) on the original properties of 3D printing filaments (viscosity, pore formation). In general, solutions to improve the properties of 3D printed parts can be performed before (pre-process), during (in-process), and after (post-process) the printing process.

4.1. Pre-Process Treatment

For pre-process treatment, plasticizers, compatibilizers, additives, and a variety of surface modification methods such as surface coating, low-temperature plasma treatment, and surface chemical reactions were used to improve the performance of 3D printed parts. Recycled materials used for 3D printing typically go through mechanical recycling processes. In other words, their polymer chains are subjected to thermomechanical degradation by high shear force and temperature during crushing and extrusion process [82]. As a result, chain scission occurs leading to a decrease in molecular weight and viscosity. Additives or plasticizers are therefore often used to control the viscosity, thermal stability, molecular weight, and crystallinity of recycled materials. Pan *et al.* [164] revealed that the addition of 1% Fe(iron)-Si(silicon)-Cr(chromium) or Fe(iron)-Si(silicon)-Al(aluminum) nano-crystalline powder into recycled PP/HDPE filament resulted in an increase by up to 37% and 17% for tensile strength and Young's modulus, respectively, compared to the original recycled filament. Wasti *et al.*

[165] added two plasticizers polyethylene glycol 2000 (PEG) and Struktol® TR451 into filaments made from lignin (20% wt) and PLA. The results showed that PEG and Struktol® TR451 improved the tensile stress and elongation at maximum load of composites up to 19% and 35%, respectively. Poly(styrene-maleic anhydride) (SMA) compatibilizer was added into PA(polyamide)/ABS to enhance interlayer adhesion [166]. The isotropy ratio for modulus, strength, and elongation at break of PA/ABS composites were improved by 62%, 77%, and 56%, respectively. Recently, polydopamine coating was used to enhance the adhesion behavior of filaments used for 3D printing [167] [168]. The results indicated that the mechanical properties of recycled PLA after coating with polydopamine were improved [167]. Moreover, a barrel atmospheric plasma system was used for the treatment of ABS and PLA polymer particles for FDM filament [169]. The treatment was performed under a helium discharge with oxygen or nitrogen addition. The results showed that the tensile strength of treated parts increased by 22% compared to those of untreated parts. Furthermore, creating self-healing filaments during 3D printing utilizing solvent-filled microcapsules is a potential future solution. Recently, Shinde *et al.* [170] have successfully created self-healing high impact polystyrene (HIPS) filaments for 3D printing. Double-walled self-healing microcapsules filled with ethyl phenylacetate solvent were synthesized and coated onto HIPS filaments for 3D printing.

4.2. In-Process Treatment

For in-process treatment, heating the deposited surface before adding the next layer is used to enhance the bonding strength between layers [171] [172]. Han *et al.* [173] used local pre-deposition heating to improve the interlayer adhesion of filament during 3D printing. Firstly, specimens were printed layer by layer on a raft. The laser spot with a 10.6 μm wavelength located 4 mm ahead of the nozzle was turned on at a 0% energy level during raft print. Once specimens begin to be printed, the energy level of the laser was increased to the pre-set value. The results showed that the isotropic behavior was found at interlayer interphase. Therefore, the tensile strength increased by 178%, and an isotropy value of 82% was achieved. Moreover, to avoid the anisotropy behavior of 3D printed parts, a chemical crosslinker between layers was also studied [174]. In addition, microwave heating was also used to improve 3D printed parts strength during printing [175]. It should be noted that the process of surface heating can lead to the warping and shape inaccuracy of 3D printed parts due to thermal stress. To address this issue, a non-heating-based solution, named cold plasma treatment (CPT) was investigated [176]. The authors revealed that the bonding strength of PLA filament improved by over 100% and 50% with a treatment duration of 30s and 300s, respectively.

4.3. Post-Process Treatment

Chemical vapor treatment is commonly used for the post-treatment of 3D filaments to minimize surface roughness. Lavecchia *et al.* [177] used ethyl acetate vapor treatment to improve the surface finish of 3D printed PLA parts. The authors revealed that almost 90% of roughness reduction was achieved. Mu *et al.* [178] used acetone, ethyl acetate, and their mixed vapor to post-treat ABS specimens fabricated by FDM. The results demonstrated that the surface finish of ABS specimens improved with all chemical vapors. However, treatment with acetone and mixed vapor caused a decrease in tensile strength as exposure time increased. In addition, the weight of specimens also increased with treatment time. Furthermore, heat treatment was also used to improve the bonding strength of finished 3D printed parts [179] [180]. However, the treatment with hot vapors may present challenges in controlling the damage to the part surfaces as well as all surfaces are not treated uniformly. Garg *et al.* [181] adapted the cold vapor treatment by acetone to achieve a good surface finish and dimensional accuracy of FDM parts. Moreover, ultrasonic vibration was also conducted to improve the mechanical properties of 3D printed parts without adjusting the printing parameters [182].

5. Discussions, Challenges, and Opportunities

As mentioned earlier, recycled, bio-based, and blended materials are available for 3D printing filaments. Therefore, engineers and researchers consider these materials depending on the specific requirements, such as the availability of recycled resources, rapid prototyping, manufacturing time and cost, availability of raw materials, excellent mechanical properties, printability, printing speeds, shrinkage and warping issues, need for heated print, thermal stability, and need for polymer modification. The findings in the selected papers on the pros and cons of recycled, bio-based, and blended materials are detailed in **Table 1** and **Table 2**. It was found that recycled and bio-based PLA, ABS, PET, HDPE, PP, and PS are available for the fabrication of 3D FDM filaments. Among commercially available filaments, PLA, ABS, and their composites are the most widely used due to their low cost and widespread availability. In contrast, materials known for their poor 3D printability such as PET, HDPE, PP, PS, recycled and their composites have also a great potential by integrating suitable processing treatments (pre-, in-, and post-process) such as the use of plasticizers, compatibilizers, additives, surface modification by coating, low-temperature plasma treatment, surface heating before depositing the next layer, or chemical vapor treatment. However, some issues regarding 3D printed parts still exist such as poor mechanical properties, poor interlayer bonding, the low fiber content in composite, voids formation, and high-water absorption. Thus, further studies should be conducted to overcome these limitations. Further surface characterization technique (atomic force microscopy (AFM)) needs to be carried out to understand the molecular orientation, thereby providing chemical cross-linking solutions to improve the mechanical properties of 3D printed parts. Creating self-healing materials during printing is also a potential solution that needs further investigation. For recycled materials, the material categories, as well as standards,

Table 1. Pros and cons of recycled filament after 3D printing by FDM.

Recycled Materials	Printing Temperature (˚C)	Pros	Cons	Ref
Recycled PLA	160 - 220	Biodegradable, non-toxic; easy to deal with 3D printing filament; high printing speeds; fewer shrinkage issues; no heated print bed necessary; no effect on T_g after number of extrusion cycles.	Slow cooling down; susceptible to degradation during its use, the melting, and recycling process; not recycled on a large scale.	[86] [87] [88] [89] [91] [92] [184]
Recycled ABS	215 - 250	Heat resistance; slightly flexible; relatively unchanged of flow rate with increasing extrusion temperature; ideal for mechanical parts; easy to recycle.	Non-biodegradable, toxic material; heated print bed necessary; shrinkage and warping issues during printing; recycled with specific program.	[86] [96] [97]
Recycled PET	212 - 235	Odorless, fairly hard, lightweight; no heated print bed necessary; availability of PET-G filaments (biocompatible, flexible, recyclable); availability of recycled resources.	High melting temperature, water absorption, lower crystallinity; non-uniform diameter for filaments; using only for engineering designs; more brittle failure than PLA.	[99] [100] [101] [102]
Recycled HDPE	180 - 190	Lightweight, flexible, non-water absorption, chemical resistance; easy to dye and mold; availability of recycled resources; easy to recycle; feed consistently into 3D printer with a constant rate of extrusion.	Heated print bed necessary; high-temperature nozzle; poor adhesion; shrinkage/warping issues; stress-induced during printing; polymer-modified necessary.	[20] [103] [185]
Recycled PP	210 - 230	Good chemical, fatigue, environment stress crack resistance; flexible; unchanged of flexural and impact strength after repeated thermal reprocessing.	Low-temperature resistance; sensitive to UV rays; warping and poor layer adhesion; significant diameter variation, elliptical shape for filaments.	[106] [107] [108] [109]
Recycled PS or HIPS	190 - 210	Availability of PS foam for filaments; availability of HIPS for filaments (similar to ABS, high impact strength, biodegradable, easy to fabricate); low viscosity for recycled PS rather than HIPS; same viscosity for recycled and virgin PS filaments.	Difficult to recycle; not recycle locally; low recycling rate; heated print bed necessary for HIPS; a more brittle tendency for recycled PS rather than HIPS.	[78] [110] [111] [112] [113]

Table 2. Pros and cons of bio-based and blended filament after 3D printing by FDM.

Matrix	Reinforcing agent	Compatibilization	Pros	Cons	Ref
PLA	Wood fiber (25 wt%)	-	Mechanical properties improved	Mechanical properties are dependent on the infill orientation of fiber; cohesion decreased, tensile strength decreased, and water absorption increased as print width increased.	[31]
PLA	Lignin (5 wt%)	-	Uniform dispersion of lignin	More brittle; break elongation decreased; tensile strength and Young's modulus decreased by 18% and 6%, respectively, compared to pure PLA.	[115]

Continued

PLA	Lignin nanoparticles (0.5 wt%)	Ethyl acetate	Melt flow and mechanical properties improved; flexural, tensile, and impact strength increased by 130.8%, 56.1%, and 14.2%, respectively.	-	[116]
PLA	Carbon fiber (25 wt%)	-	25% higher flexural strength compared to original printing process; good adhesion CF/PLA.	-	[117]
PLA	Carbon fiber	PVA	Tensile and bending strength increased by 35% and 108%, respectively, compared to pure PLA; good adhesion.	Delamination failure	[118]
PLA	Basalt fiber (20 wt%)	-	Lighter and better than conventional mold-pressed composites.	Voids (inter- and inner-filament voids) still exist; mechanical properties of composites depend on fiber length and fiber orientation	[119]
ABS	Short carbon fiber (5 wt%, length of 150 μm)	-	Tensile strength increased by 25% compared to pure ABS.	Porosity increased as fiber content increased	[120]
ABS	Carbon fiber (10 wt%)	-	Greater tensile and flexural strength compared to neat ABS; distortion and warping decreased.	Low interlaminar shear strength compared to injection parts; poor interface.	[121] [122]
ABS	Short carbon fiber (length of 0.2 - 0.4 mm)	-	Tensile and modulus increased by 115% and 700%, respectively, compared to neat ABS.	20% void formation; poor adhesion.	[123]
ABS	Glass fiber	-	Stiffness increased by 84% compared to neat ABS; thermal stability unchanged.	-	[125]
ABS	Kevlar/carbon fibers	-	Rigidity and ductility increased.	-	[126]
ABS	Palm fiber (15 wt%)	-	Hydrogen bonding increased by 42%; T_g unchanged.	-	[127]
ABS	Bamboo fiber	Chemical treatment	Mechanical properties unchanged.	Density decreased.	[128]
ABS	Pine cone fiber (2 - 5 wt%)	Chemical treatment	Filament diameter and density unchanged.		[129]
ABS	Rice straw fiber (5 - 10 wt%)	-	-	Tensile and flexural strength decreased as rice straw fiber content increased; water absorption increased.	[130]
PET	Carbon fiber (15 wt%)	-	Elastic modulus, tensile, and shear strength increased by 180%, 230%, 40%, respectively, compared to neat PET.	-	[131]

Continued

PET-G	Carbon fiber (20 wt%)	-	Maximum 43.7% and 25% in tensile and flexural strength for honeycomb pattern; filament properties unchanged when replaced with recycled PET-G.	Viscosity increased; lower interlayer bonding; post-process treatment necessary.	[132] [133] [136] [137]
Recycled PET	Post-consumer textile (10 wt%)	Acid hydrolysis and silane functionali-zation	Impact resistance and the dampening characteristics improved; good adhesion; ductile failure.	High melt flow index (MFI).	[138]
HDPE	Birch fiber (10 - 30 wt%)	Maleic anhydride	Without significant warping, shrinkage, and other geometric deformation issues; deformation reduced up to 80%; Young's modulus increased by 35%.	-	[140]
HDPE	Wood fiber (40 wt%)	MAPE	Strength of composite increased.	-	[141]
HDPE	Cardboard dust	-	Filaments could be printed with the cardboard content was up to 50 wt%.	High porosity; non uniformity of structure; T_g decreased; mechanical property, tenacity, and elastic modulus decreased.	[142]
PP	Harakeke fiber (30 wt%)	Maleated PP	Tensile and Young's modulus increased by 74% and 214%, respectively, compared to neat PP; shrinkage decreased by 84%.	-	[143]
PP	Cellulose nanofibril (10 wt%)	MAPP	Flexural strength and modulus increased by 5.9% and 26.8%, respectively.	-	[144]
PP	Carbon fiber	MAPP	Uniform filler dispersion; good interface adhesion; mechanical properties improved.	Printing orientation affected the mechanical properties of composites.	[145]
PP	Glass fiber	POE-g-MA	Flexibility increased.	Modulus and strength decreased.	[146]
PP	Microcrystalline cellulose	n-octyltrieth-oxysilane	Good surface finish; easy printing; good mechanical properties.	-	[147]
Recycled PP	Rice husk fiber (5 - 10 wt%)		Crystallinity increased as rice husk fiber content increased; low warping.	Density decreased and water absorption increased as rice husk fiber content increased.	[148]
Recycled PS	Corn husk fiber (2.5 - 10 wt%)	Pre-process treatment with a layer of glue	Filament containing 2.5 - 7.5 wt% of fiber could be printable; slight decrease in thermal stability.	Filament containing 10 wt% of fiber failed to be printed; tensile strength and modulus decreased as fiber content increased; dull and rough surfaces.	[150]
PS	Cellulose nanocrystal	PEG/PEO	Mechanical properties improved.	-	[151]

should be established for 3D printing filaments. Unheated treatment such as cold plasma treatment can achieve a good surface finish and dimensional accuracy of FDM parts. Furthermore, nylon (often called polyamide) and thermoplastic elastomers (thermoplastic elastomer (TPE) and thermoplastic polyurethane (TPU)) are suitable for 3D printing. However, studies on the use of recycled or composite materials from nylon and thermoplastic elastomers have not yet been conducted. In addition, recycled high-performance polymers including polyetherimide (PEI) needs to be investigated for producing 3D filaments. It should be noted that thermosetting photopolymers occupy half of the 3D printing materials market. Future research on reprocessable thermosets is, therefore, essential [183].

6. Conclusion

In this paper, an overview of various recycled, bio-based, and blended materials for FDM 3D printing filaments was conducted. The advantages and disadvantages of thermoplastics and their composites were discussed. This review is intended as a reference resource for engineers and researchers to select suitable materials for 3D printing. When compared with injection molded materials, 3D printed parts have worse mechanical properties. The 3D printing technology, however, has a huge potential, comprising its simplicity, inexpensive cost, and customizability. It is evident that 3D printing technology has affirmed its position in the rapid supply of medical and technological products in the recent coronavirus period. Furthermore, the use of recycled and composite materials from natural fibers for 3D printing contributes to saving oil and energy as well as reducing the impacts on climate change and the environment. Thus, it should be taken to account the economic and social benefits of 3D printing nowadays. From the findings in the present paper, however, some limitations still exist for 3D printed parts from recycled and composite materials. In this regard, further solutions must be found to improve the quality and availability of recycled, bio-based, and blended materials for 3D printing. Using plasticizers, compatibilizers, additives, surface modification by coating, low-temperature plasma treatment, heating process before depositing the next layer, or chemical vapor treatment should be conducted to enhance the adhesion bonding and mechanical properties of 3D printed parts. Otherwise, a standard certification for 3D printing filament from recycled and composite materials needs to be established. Future studies on self-healing materials and reprocessable thermoset materials need to be investigated to expand the category of materials for 3D printing technology.

Conflicts of Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Nomenclature

AM	Additive manufacturing;
FDM/FFF	Fused deposition modelling/Fused Filament Fabrication;
SLA	Stereo-lithography;
DLP	Digital Light Processing;
CAD	Computer-aided design;
SLS	Selective laser sintering;
MJF	Multi jet fusion;
PJM/MJM	PolyJet/MultiJet modeling;
LOM	Laminated Object Manufacturing;
CTE	Coefficient of thermal expansion;
PLA	Polylactic acid;
ABS	Acrylonitrile butadiene styrene;
PET	Polyethylene terephthalate;
HDPE	High-density polyethylene;
PP	Polypropylene;
PS	Polystyrene;
HIPS	High impact polystyrene;
Tcc	Cold crystallization temperature;
T_g	Glass transition temperature;
PET-G	Glycol-modified polyethylene terephthalate;
WOOF	Washington Open Object Fabricators;
MFI	Melt flow index;
EALNSs	Ethyl acetate-treated lignin nanospheres;
PVA	Polyvinyl alcohol;
CF	Carbon fibers;
GF	Glass fibers;
rPET	Recycled polyethylene terephthalate;
MAPE	Maleated polyethylene;
MAPP	Maleic anhydride polypropylene;
CNF	Cellulose nanofibril;
POE-g-MA	Maleic anhydride polyolefin;
MCC	Microcrystalline cellulose;
rPP	Recycled polypropylene;
rPS	Recycled polystyrene;
EPS	Expanded polystyrene foam;
PEG/PEO	Polyethylene glycol/polyoxyethylene;
PE-g-MAH	Polyethylene graft maleic anhydride;
PHBV	Poly(3-hydroxybutyrate-co-3-hydroxyvalerate);
PLA/S-co-MMA	Poly(styrene-co-methyl methacrylate);
PBAT	Poly(butylene adipate-co-terephthalate);
NR	Natural rubber;
BioPBS	Poly(butylene Succinate);

CLTE	Coefficient of linear thermal expansion;
UHMWPE	Ultrahigh molecular weight polyethylene;
SEBS	Styrene ethylene butadiene styrene;
TPU	Thermoplastic polyurethane;
MFCs	Microfibrillar composites;
SMA	Poly(styrene-maleic anhydride);
PA	Polyamide;
CPT	Cold plasma treatment;
AFM	Atomic force microscopy;
TPE	Thermoplastic elastomer;
PEI	Polyetherimide.