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3D printing of polymer matrix composites: A review and prospective

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The use of 3D printing for rapid tooling and manufacturing has promised to produce components with complex geometries according to computer designs. Due to the intrinsically limited mechanical properties and functionalities of printed pure polymer parts, there is a critical need to develop printable polymer composites with high performance. 3D printing offers many advantages in the fabrication of composites, including high precision, cost effective and customized geometry. This article gives an overview on 3D printing techniques of polymer composite materials and the properties and performance of 3D printed composite parts as well as their potential applications in the fields of biomedical, electronics and aerospace engineering. Common 3D printing techniques such as fused deposition modeling, selective laser sintering, inkjet 3D printing, stereolithography, and 3D plotting are introduced. The formation methodology and the performance of particle-, fiber- and nanomaterial-reinforced polymer composites are emphasized. Finally, important limitations are identified to motivate the future research of 3D printing.

1. Introduction

3D printing, also referred to as additive manufacturing(AM), rapid prototyping(RP), or solid-freeform(SFF), is a 'process of joining materials to make objects from 3D model data, usually layer by layer' [1], which was first described in 1986 by Charles Hull [2]. This technology creates objects by adding materials to reduce waste while reaching satisfactory geometric accuracy [3]. It begins with a meshed 3D computer model that can be created by acquired image data or structures built in computer-aided design (CAD) software. A STL (Surface Tessellation Language) file is commonly created. The mesh data will be further sliced into a build file of 2D layers and sent to the 3D printing machine.

Thermoplastic polymer materials such as acrylonitrile butadiene styrene(ABS) [4–6], polylactic acid(PLA) [4,6,7], polyamide(PA) [8] and polycarbonate(PC) [9] as well as thermosetting polymer materials like epoxy resins could be processed by 3D printing technology. Epoxy resins are reactive materials that require thermal or UV-assisted curing to complete the polymerization process, and they initially exhibit a low viscosity, which rises as the curing proceeds [10–12]. Therefore, epoxy resins are suitable for heat or UV– assisted printing process. Based on the various selections of materials, 3D printing of polymers has found their possible applications in aerospace industries for creating complex lightweight structures [13], architectural industries for structural models [14], art fields for artifact replication or education [15], and medical fields for printing tissues and organs [16]. However, most of 3D printed polymer products are still now used as conceptual prototypes rather than functional components, since pure polymer products built by 3D printing are lack of strength and functionality as fully functional and load-bearing parts. Such drawbacks restrict the wide industrial application of 3D printed polymers.

3D printing of polymer composites solves these problems by combining the matrix and reinforcements to achieve a system with more useful structural or functional properties non attainable by any of the constituent alone [17]. Incorporation of particle, fiber or nanomaterial reinforcements into polymers permits the fabrication of polymer matrix composites, which are characterized by high mechanical performance and excellent functionality. Conventional fabrication techniques of composites such as molding, casting and

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machining create products with complex geometry through material removal processes [18]. While the manufacturing process and performance of composites in these methods are well-controlled and understood, the ability to control the complex internal structure is limited. 3D printing is able to fabricate complex composite structures without the typical waste. The size and geometry of composites can be precisely controlled with the help of computer aided design. Thus, 3D printing of composites attains an excellent combination of process flexibility and high performance products.

Although 3D printing has attracted a lot of attentions over the past three decades, most of published review articles focused on introductions of processing techniques and printing of pure polymer materials. However, in the recent several years, there has been considerable achievements in developing printable polymer composites with improved performance. Hence, we present, analyze and summarize information pertinent to 3D printing of polymer composites in this review. We first provide a brief introduction of 3D printing technique used for polymer composites and their characteristics. Next, we investigate the detailed printing technique implementations and properties improvements of polymer composites. Biomedical, electronics and aerospace applications of polymer composites are explored then. In particular, work done in last five years are emphasized to show the progress in this area. Finally, we discuss the limitations of current technologies and future perspective.

2. 3D printing technology description

3D printing is a methodology that produces 3D haptic physical models layer by layer based on CAD models [19]. Various printing techniques have been employed to fabricate polymer composites. Among them, some techniques are well-established, such as fused deposition modeling, selective laser sintering, inkjet 3D printing, stereolithography and 3D plotting whereas others are still in development or used only by small groups of researchers. Each technique has its own advantages and limitations in producing composite products. The selection of fabrication technique depends on the starting materials, requirements of processing speed and resolution, costs and performance requirements of final products. Established rapid prototyping techniques are summarized in Table 1.

2.1. Fused deposition modeling(FDM)

Fused deposition modeling(FDM) printers are the most commonly used printers for fabricating polymer composites. Thermoplastics such as PC, ABS and PLA, are commonly used due to their low melting temperature. FDM printers work by controlled extrusion of thermoplastic filaments, as shown in Fig. 1(a). In FDM, filaments melt into a semi-liquid state at nozzle and are extruded layer by layer onto the build platform where layers are fused together and then solidify into final parts. The quality of printed parts can be controlled by altering printing parameters, such as layer thickness, printing orientation, raster width, raster angle and air gap. The effects of processing parameters have been discussed by Sood et al. [20].

One common drawback of FDM printing is that the composite materials have to be in a filament form to enable the extrusion process. It is difficult to homogeneously disperse reinforcements and remove the void formed during the manufacturing of composite filaments. Another disadvantage of FDM printers is that the usable material is limited to thermoplastic polymers with suitable melt viscosity. The molten viscosity should be high enough to provide structural support and low enough to enable extrusion. Also, complete removal of the support structure used during printing may be difficult. Notwithstanding these drawbacks, FDM printers also offer advantages, including low cost, high speed and simplicity. Another advantage of FDM printing is the potential to allow deposition of diverse materials simultaneously. Multiple extrusion nozzles with loading of different materials can be set up in FDM printers, so printed parts can be multi-functional with designed composition.

2.2. Powder bed and inkjet head 3D printing(3DP)

Powder-liquid 3D printing technology was developed at the Massachusetts Institute of Technology (MIT) in 1993 as a rapid-prototyping technology [21]. This technology is based on powder processing, as shown in Fig. 1(b). Powders are first spread on the build platform and then selectively joined into a patterned layer by depositing a liquid binder through inkjet printhead, which is able to move in X–Y direction. After a desired 2D pattern is formed, the platform lowers and the next layer of powder is spread. This process is repeated and finally unbounded powder should be removed to get final products. The internal structure can be controlled by altering the amount of deposited binder. Factors that determine the quality of final products are powder size, binder viscosity, interaction between binder and powder, and the binder deposition speed. The effect of processing parameters has been discussed in detail in a review by Ben et al. [22].

The key advantages of this technology are the flexibility of material selections and room temperature processing environment. Theoretically, any polymer materials in powder state could be printed by this technology. Removal of support structure is

Table 1
A summary of established rapid prototyping techniques.

Technique	State of starting materials	Typical polymer materials	Working principle	Resolution (Z direction,μm)	Advantages	Disadvantages
FDM	Filament	Thermoplastics, such as PC, ABS, PLA, and nylon	Extrusion and deposition	50–200 (Rapide Lite 500)	Low cost, good strength, multi-material capability	Anisotropy, nozzle clogging
SLA	Liquid photo-polymer	Photocurable resin (epoxy or acrylate based resin)	Laser scanning and UV induced curing	10 (DWSLAB XFAB)	High printing resolution	Material limitation, cytotoxicity, high cost
SLS	Powder	PCL and polyamide powder	Laser scanning and heat induced sintering	80 (Spo230 HS)	Good strength, easy removal of support powder	High cost, powdery surface
3DP	Powder	Any materials can be supplied as powder, binder needed	Drop-on-demand binder printing	100–250 (Plan B, Ytec3D)	Low cost, multi-material capability, easy removal of support powder	Clogging of binder jet, binder contamination
3D plotting	Liquid or paste	PCL, PLA, hydrogel	Pressurized syringe extrusion, and heat or UV-assisted curing	5-200 (Fab@home)	High printing resolution, soft materials capability	Low mechanical strength, slow

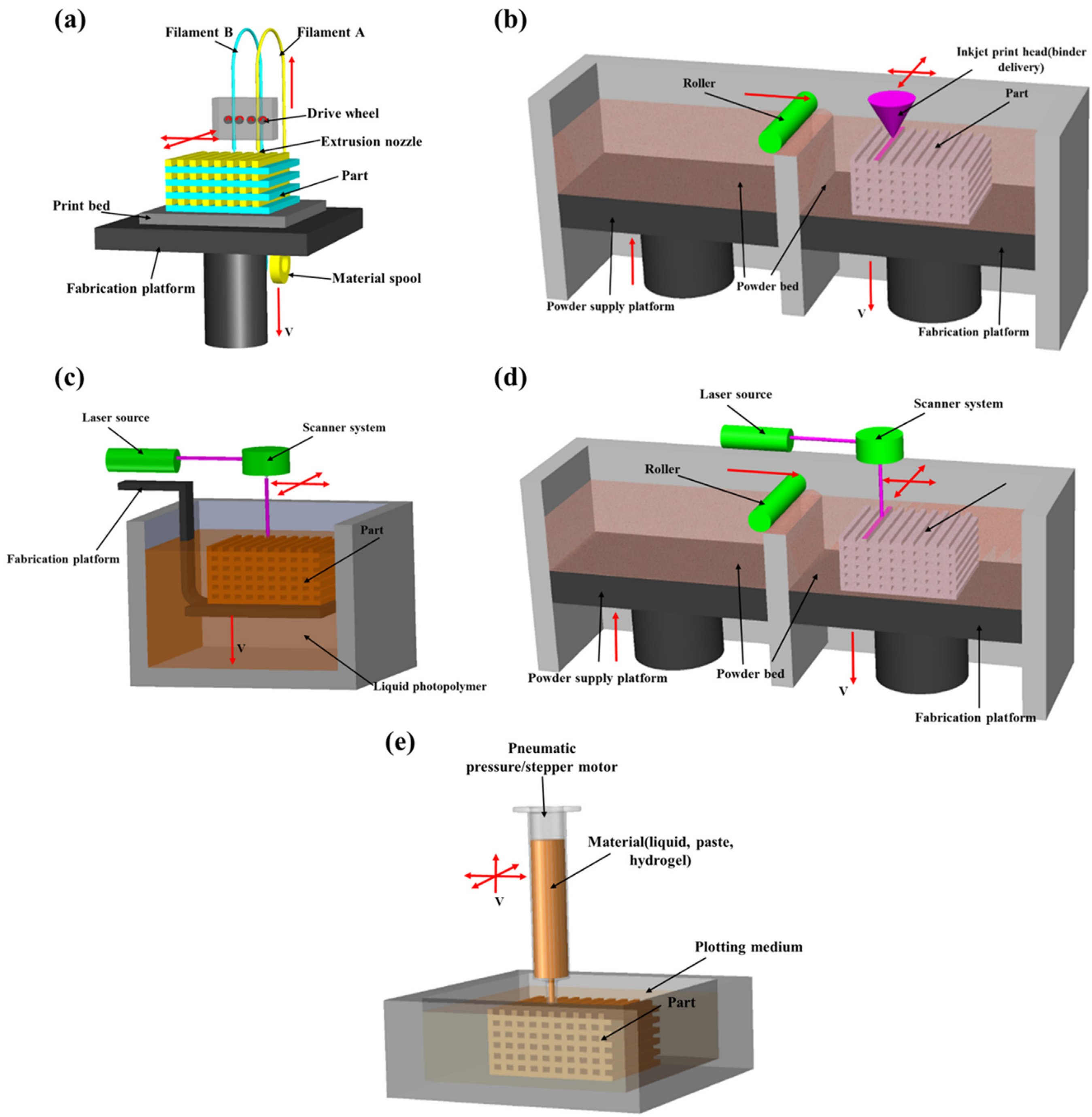

Fig. 1. Schematic representation of a typical (a) FDM setup (b) 3DP setup (c) SLA setup (d) SLS setup (e) 3D plotting setup.

relatively easy with this technique. However, the binder used may incorporate other contaminations and the printing resolution is very limited for this technology.

2.3. Stereolithography(SLA)

Stereolithography uses photopolymers that can be cured by UV laser. An UV-laser is controlled in a desired path to shoot in the resin reservoir, and the photocurable resin will polymerize into a 2D patterned layer. After each layer is cured, the platform lowers and another layer of uncured resin is ready to be patterned [23], as shown in Fig. 1(c). Typical polymer materials used in SLA are acrylic and epoxy resins.

Understanding the curing reactions occurring during polymerization is critical to control the quality of final printed parts. Intensity of laser power, scan speed and duration of exposure affect the curing time and printing resolution [24]. Photoinitiators and UV absorbers can be added to the resin to control the depth of polymerization [25]. The main advantage of SLA printing technology is the ability to print parts with high resolution. Additionally, because SLA is a nozzle-free technique, the problem of nozzle clogging can be avoided. Despite these advantages, the high cost of this system is a main concern for industrial application. Possible cytotoxicity of residual photoinitiator and uncured resin is another concern.

2.4. Selective laser sintering(SLS)

Selective laser sintering technique is similar to previously mentioned 3DP technique and they are both based on powder processing. Instead of using a liquid binder, in SLS, a laser beam with a controlled path scans the powders to sinter them by heating, as shown in Fig. 1(d). Under high power lasers, neighboring powders are fused together through molecular diffusion and then processing of next layer starts. Unbounded powder should be removed to get final products [26]. The feature resolution is determined by powder particle size, laser power, scan spacing and scan speed [27].

Although theoretically any thermoplastic polymer in powder form could be processed by SLS technique, the complex consolidation behavior and molecular diffusion process during sintering have limited the choice of materials used in SLS process [28]. To the date now, polycaprolactone(PCL) and polyamide(PA) are widely used laser sintering materials.

2.5. 3D plotting/direct-write

3D plotting is based on extruding a viscous material from a pressurized syringe to create 3D shape of materials, as shown in Fig. 1(e). The syringe head can move in three dimensions, while the platform keeps stationary where extruded materials are joint together layer by layer. Curing reactions can be performed by dispensing two reactive components using mixing nozzles or be induced either by heat or UV light [29]. In certain cases, materials can be delivered to a plotting medium to finish the curing reaction. Material viscosity and deposition speed correlate with the quality of final printed parts [16].

The key advantage of this technique is material flexibility. Solutions, pastes and hydrogels can all be loaded into 3D plotting printers. A temporary, sacrificial material may be needed to support the printed structure since raw viscous materials have low stiffness that may result in the collapse of complex structures.

2.6. Other techniques

Recently, several new techniques are developed for 3D printing of composites, such as PolyJet which works by polymerization of deposited droplets of photopolymer ink [30], digital light processing(DLP) which is based on selective polymerization of an entire surface of photopolymer by a projector light [31], liquid deposition modeling(LDM) which consists in the additive deposition of material layers directly from a solution in a volatile solvent [32], and fiber encapsulation additive manufacturing(FEAM) which involves directly encapsulate fiber within an extruded flowable polymer matrix [33]. Compared to traditional 3D printing techniques, these methods have either more material selections or less processing time. However, due to their high cost and complexity, there is only a few research adopting these new techniques.

3. 3D printing of polymer matrix composites

Polymer materials with low melting point or in liquid state are widely used in 3D printing industry due to their low weight, low cost and processing flexibility. Although 3D printed polymer products could have geometric complexity, lack of mechanical strength and functionality is a big challenge for their wide applications. Combining various materials for achieving desired mechanical and functional properties is a promising way to solve these problems. Therefore, in recent years, development of composite materials that are compatible with the available printers has attracted tremendous attentions. Many promising results in developing new printable composite materials reinforced by particles, fibers or nanomaterials have been demonstrated. These results are discussed below in detail.

3.1. Particle reinforced polymer composites

Due to their low cost, particle reinforcements are widely used to improve the properties of polymer matrix. Particles are easy to be mixed with polymers, either in powder form for SLS or in liquid form for SLA, or further to be extruded into printable filaments for FDM process. Table 2 summarizes various particle reinforced polymer materials used for 3D printing and the properties improvements of resulting composites. The key issues for consideration in the 3D printing of particle reinforced composites including improved tensile/storage modulus by adding glass beads [34], iron or copper particles [35], improved wear resistance by adding aluminum and aluminum oxide(Al_2O_3) [36], and improved dielectric permittivity by adding ceramic [37,38] or tungsten [39] particles. In these cases, cuboid or cylinder-shaped parts were fabricated through FDM, SLS or SLA technique and improved properties were observed, but no further structural application was demonstrated.

An exciting development in 3D printing of particle reinforced composites is the capability to print structural components for potential real-world applications. Recently, Kalsoom et al. [40] employed SLA technique for the fabrication of a heat sink composite structure, as shown in Fig. 2. This composite structure consists of up to 30%(w/v) microdiamond particles in acrylate resins. The temperature of the composite heat sink was higher compared to that of the pure polymer heat sink when the sinks were heated at the same temperature, thus demonstrating the improved heat transfer rates by the addition of diamond particles. In another work, Castles et al. [41] demonstrated the printing of diamond photonic crystal structures using barium titanate($BaTiO_3$)/ABS by FDM(Fig. 3). Improved and adjustable dielectric relative permittivity was observed by incorporation of $BaTiO_3$ particles. At 70 wt% of $BaTiO_3$ loading, the relative permittivity of the printed composite increased 240% compared to that of pure polymer. Additionally, due to the flexibility of 3D printing technology, the principal components of the effective permittivity tensor can be tuned by adjusting the dimensional patterning in this work.

The addition of particles into polymers also helps address some difficulties in the printing process. One obstacle for FDM printing process is the distortion of final printed parts, which is caused by the thermal expansion of polymer. Embedding metal particles into polymers was proved to be an efficient solution to this problem [42]. When combing with copper and iron particles, ABS composites showed a large reduction in coefficient of thermal expansion, thus the distortion of printed part reduced a lot. Another characteristic for FDM printing process is the anisotropy properties of the 3D printed part, which, depending on the application, can be advantages or limitations. If the printed part needs to be used under isotropic loading condition, the low tensile strength and modulus in the direction perpendicular to building orientation [43] may cause the failure of the printed part. Thermoplastic elastomer(TPE) is a promising additive to reduce the mechanical property anisotropy. Perez et al. [44] prepared ABS-based composites with TPE, and the tensile test results demonstrated a reduction in the difference between tensile strength in two perpendicular directions, which indicating a reduction in mechanical property anisotropy. In another recent study, Kokkinis et al. [45] developed a novel magnetically assisted 3D printing platform and control of particles orientation was realized by incorporating magnetized alumina platelets into polymer matrix(Fig. 4). Because of the alignments of anisotropic particles, the target properties of printed composite parts in particular directions have been enhanced. Similarly,

Table 2
A summary of techniques and materials used for 3D printing of particle reinforced polymer composites and properties improvement of resulting composites.

Technique	Materials	Enhancement in properties	Ref.
FDM	Iron/ABS, and Copper/ABS	Improved storage modulus and thermal conductivity, reduced coefficient of thermal expansion	[35,42]
	Al and Al_2O_3/Nylon-6	Reduced frictional coefficient	[36]
	$BaTiO_3$/ABS and $CaTiO_3$/Polypropylene	Improved dielectric permittivity and controllable resonance frequency	[37]
	Tungsten/PC	Improved dielectric permittivity, x-ray attenuation factor and impact resistance	[39]
	$BaTiO_3$/ABS	Improved and tunable dielectric permittivity, periodic and graded structures were printed for demonstration	[41]
	Thermoplastic elastomer/ABS	Reduced anisotropy of printed components	[44]
SLA	Al_2O_3/UV cured resin	Improved dielectric permittivity and reduced dielectric loss tangent	[38]
DLP	Diamond microparticle/acrylate based resin	Improve heat transfer, heat sink and cooling system were printed for demonstration	[40]
Magnetically assisted-DLP	Alumina/UV-sensitive resin	Controlled orientation of magnetized particle and bioinspired composite micro-architectures were printed	[46]
Magnetically assisted-direct writing	Alumina/polyurethane acrylate	Controlled local composition and orientation of magnetized particle	[45]
SLS	Glass bead/Nylon-11	Improved tensile modulus and compressive modulus, whereas reduced elongation at break	[34]

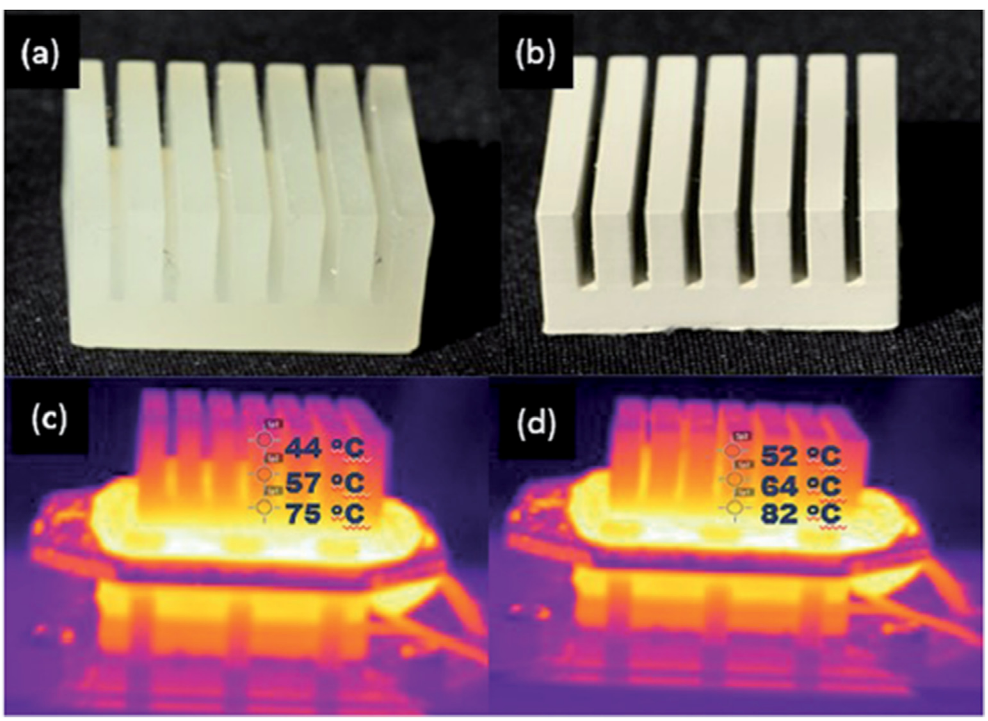

Fig. 2. 3D printed heat sinks using (a) Acrylate resin, (b) 30% (w/v) composite material; IR images of (c) Polymer heat sink (d) Composite heat sink heated for 10 min at 100 °C. Reprinted with permission from Ref. [40].

magnetized alumina particles could also be blended in UV-sensitive resins for SLA printing and aligned under magnetic fields during the printing process [46]. Bioinspired composite micro-architectures were designed and realized by controlling the orientation of magnetized alumina particles, and the resulting mechanical properties were demonstrated to be dependent on their mircostructures.

3.2. Fiber reinforced polymer composites

Fiber reinforcements can also significantly enhance the properties of polymer matrix materials. Common 3D printing technologies to produce fiber-reinforced polymer composites are FDM and direct write technique. For FDM processing, polymer pellets and fibers are mixed in a blender first and then delivered to extruder to be fabricated into filaments. A second extrusion process could be conducted to make sure the homogenous distribution of fibers. For direct writing processing, polymer paste and fibers were mixed first and directly extruded out. Powder based technologies are not ideal for creating fiber-reinforced composites because making a smooth layer of powder-fiber mixture is difficult [47]. Table 3 summarizes the materials used for 3D printing of fiber reinforced polymer composites and mechanical property improvements of resulting composites.

Typical short fibers including glass fibers [48] and carbon fibers(CFs) [49–53] are common-used reinforcements to improve the mechanical properties of polymer composite in 3D printing area. Fiber orientation and void fraction of composites play an important role in determining the properties of final composite parts. Tekinalp et al. [49] investigated the fiber orientation and

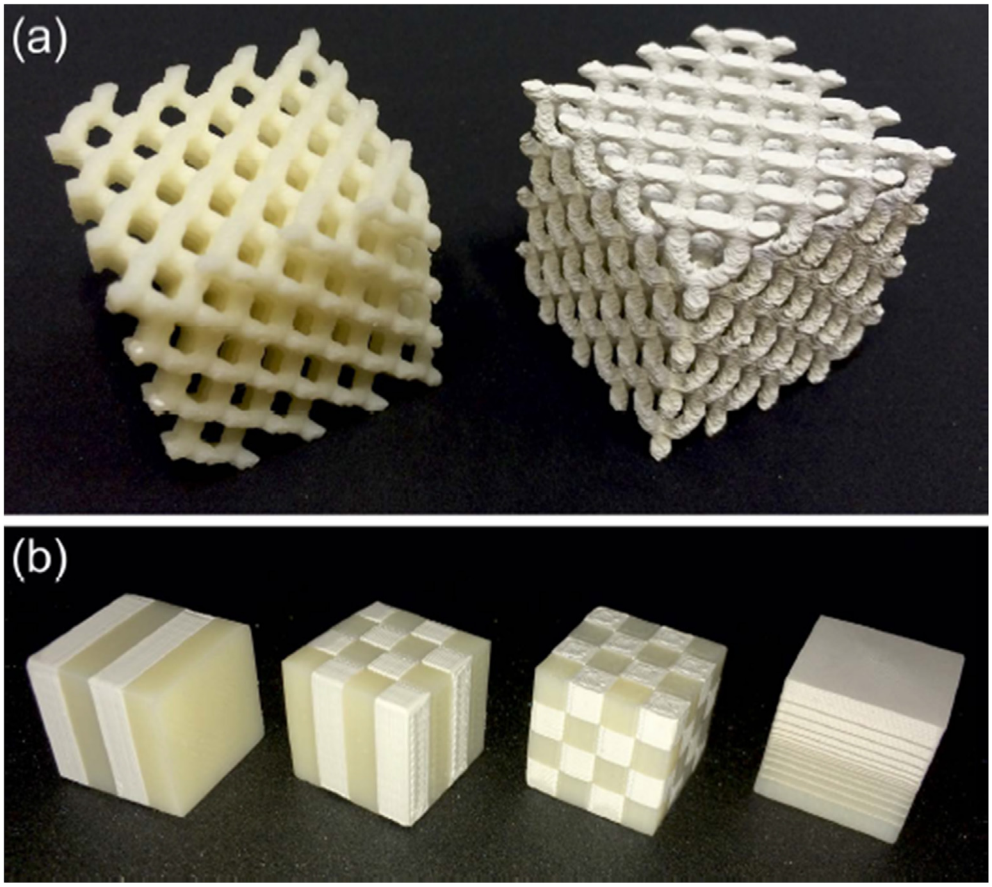

Fig. 3. a) Rod-connected diamond photonic crystal structures printed in ABS polymer (left, $\varepsilon' = 2.57$) and 50 wt% $BaTiO_3$/ABS polymer composite (right, $\varepsilon' = 4.95$). (b) 1D, 2D, and 3D periodic structures and a 1D graded structure printed using a combination of ABS polymer and 50 wt% $BaTiO_3$ in ABS polymer composite. Reprinted with permission from Ref. [41].

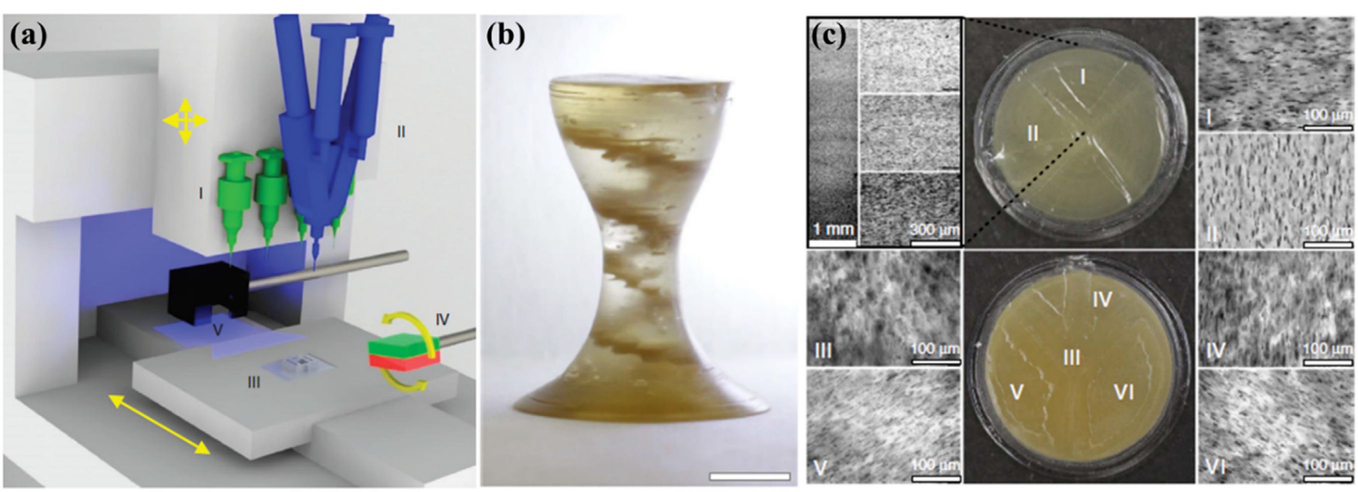

Fig. 4. (a) Schematics of the magnetically-assisted 3D printing platform for the creation of heterogeneous composites. (I) multiple dispensers (II) a mixing unit (III) movable head and table (IV) a magnet (V) a curing unit (b) Printed object with internal helicoidal staircase. Scale bar, 5 mm. (c) Photograph and optical microscope of the structure, highlighting the successful realization of the programmed gradient in platelet concentration and locally different platelet alignment. Reprinted with permission from Ref. [45].

porosity effect on the properties of FDM printed carbon fiber reinforced ABS composite parts. Composite parts are also made by compression molding(CM) as a comparison. Due to the presence of gaps between deposition lines and poor bonding between polymer and fiber, 3D printed composite samples showed significant void formation(~20%), as shown in Fig. 5(a), whereas compression molded samples showed almost no pores. However, the tensile strength improvement for printed samples is close to that for compressive molded samples. That is because more fibers are aligned in the load-bearing direction during printing, which compensates the negative effect of porosity. Compton et al. [54] also demonstrated that fillers could be oriented along printing direction

Table 3
A summary of techniques and materials used for 3D printing of fiber reinforced polymer composites and mechanical properties improvement of resulting composites.

Technique	Materials	Fiber loading	Maximum tensile strength (MPa)	Tensile strength improvement(%) compared to that of pure polymer	Ref.
FDM	Short glass fiber/ABS	18 wt%	58.6	140	[48]
	Short carbon fiber/ABS	40 wt%	70	115	[49]
		5 wt%	42	24	[50]
		13 wt%	70.69	194	[51]
Direct write	Short carbon fiber/Silicon carbide whisker/epoxy	35 wt%	66.2	127	[54]
FDM based co-extrusion	Continuous carbon fiber/nylon	34.5 vol%	464.4	446	[57]
	Continuous carbon fiber/PLA	6.6 vol%	185.2	335	[60]

and a lightweight cellular composite was obtained by direct writing technique, as shown in Fig. 6. They produced a bio-inspired wood structure with controlled architecture and mechanical properties using silicon carbide whiskers and carbon fibers reinforced epoxy. Through adjusting the alignment of fibers and whiskers reinforcements, highly optimized structure with desired stiffness and toughness was obtained.

Since the voids that formed during the printing process significantly impair the mechanical properties of printed composites, researchers have made many efforts to investigate how to reduce the void formation. Recently, it was found that the expandable microspheres could be added into polymer to reduce the porosity in printed parts [55]. At 11 wt% of microsphere loading, the porosity of printed parts reduced from 17% to 7%. It is promising to largely increase the mechanical properties of printed composites by using this additive. In another interesting work, Le Duigou et al. [56] reported that printed fiber reinforced composites may take advantage of formed voids. A wood fiber reinforced biocomposite was prepared by FDM process and it exhibited an increasing of swelling ability compared to parts without void. This hygroscopic property could be tuned into an advantage in design of a programmable moisture-actuated biocomposite.

The effect of fiber content on the mechanical properties of printed parts is another interesting research topic. Tekinalp et al. [49] demonstrated that ABS/carbon fiber composites prepared by FDM showed an increasing of tensile strength and modulus as the fiber content increased, and a maximum 115% and 700% increase could be obtained at 40 wt% fiber loading. Fig. 5(b) shows the effect of fiber content and preparation process on tensile strength of ABS/CF composites. Ning et al. [50] also studied the effect of fiber content on the mechanical properties of FDM printed ABS/carbon fiber composites. The best performance of the printed parts was obtained at 5 wt% fiber loading and higher loading of fiber deteriorated the performance of printed parts due to the higher porosity. Actually, there is a significant variation in fiber loading content to achieve the maximum improvement of mechanical properties between different cases of study, mostly because fiber distribution condition and interfacial bonding strength vary a lot between each case. A basic standard for design and processing may need to be established.

Up to now, the adding content of fibers is up to 40 wt% and the composites with more fibers are not able to be printed due to nozzle clogging issues. In addition, composites with higher fiber loading are difficult to be made into continuous filaments for FDM due to the loss of toughness. Therefore, the properties of the resulting composites are limited by the low fiber content. It is essential to further understand the rheological properties of printing materials and increase the fiber content. Applying plasticizers and compatibilizers could be a possible way to improve the feedstock processability [48].

Another challenge for fiber based printing is the difficulty of the addition of continuous fibers. Most of the research until now only present the addition of short fibers in polymer matrix. Recently, several researchers reported continuous fiber based printing. A study was conducted to evaluate the mechanical properties of continuous fiber reinforced thermoplastic composites printed by a commercial available Mark One printer [57]. The printed part has a sandwich construction that consists of carbon fiber reinforced thermoplastic(CFRP) in the middle and nylon polymer at the top and bottom. Two print heads were employed to extrude CFRP and

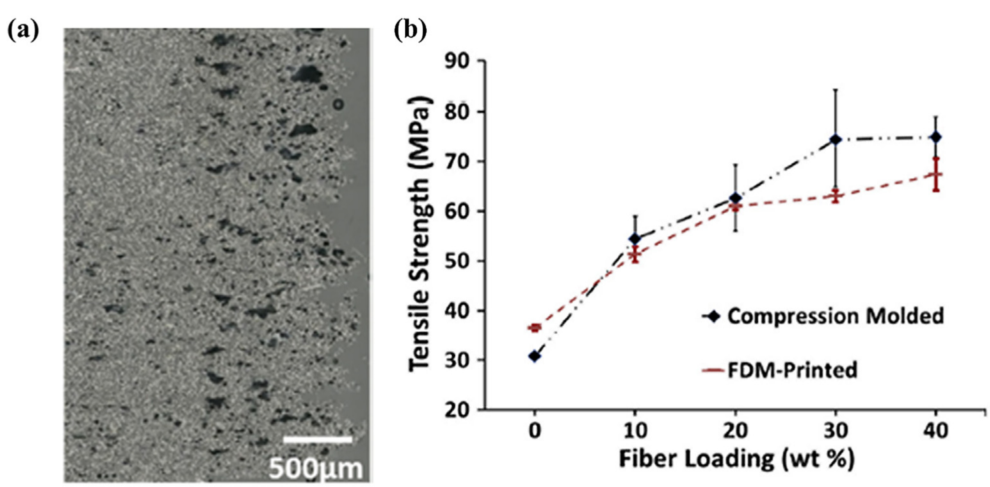

Fig. 5. (a)Micrograph of the polished surface of the printed ABS/30 wt% carbon fiber composite (b) Effect of fiber content and preparation process on tensile strength of the printed ABS/carbon fiber composite. Reproduced with permission from Ref. [49].

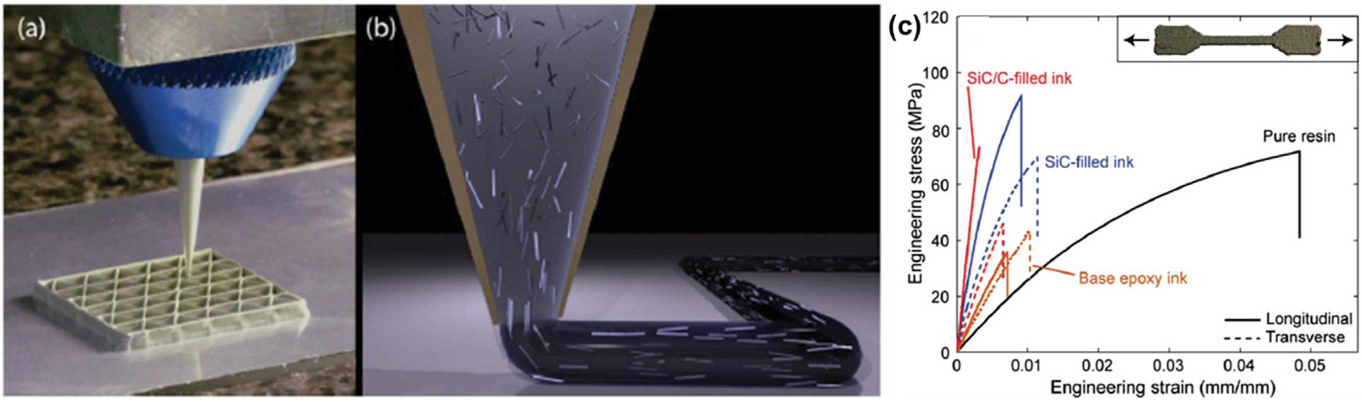

Fig. 6. (a) Optical image of 3D printing of a triangular honeycomb composite. (b) Schematic illustration of the progressive alignment of high aspect ratio fillers within the nozzle during composite ink deposition. (c) Representative tensile stress versus strain curves for 3D printed tensile bars of varying composition and control samples cast from pure epoxy resin. Reprinted with permission from Ref. [54].

nylon respectively. In other research, in-site fiber impregnation was adopted to print continuous fiber based composites [58–60]. As shown in Fig. 7, PLA filaments and continuous carbon fibers were separately supplied and co-extruded. Deposition layer thickness, temperature of liquefier, hatch spacing and printing speed were found to affect the mechanical properties of the continuous fiber reinforced PLA composites [58]. Matsuzaki et al. [60] reported that the tensile modulus and strength of 3D-printed continuous carbon fiber reinforced PLA composites are 19.5 ($\pm$2.08) GPa and 185.2 ($\pm$24.6) MPa, respectively, which are 599% and 435% of the tensile modulus and strength of the pure PLA specimens. This mechanical improvement is much larger compared to that of short fiber reinforced PLA composites. However, in some cases, irregularity and discontinuity of fiber still exist in the printed samples. Although the mechanical properties of composites were largely improved compared with pure polymer, the improvement was still lower than the theoretical value calculated by the ruler of mixture [57,59].

Shape memory polymer composites are attractive for great research interest due to their ability to store and recover deformation for deployable and morphing structures [61–63]. Therefore, shape memory polymers could enable 3D printing to 4D printing, where the geometry or property of a 3D printed component could be controlled in time. Ge et al. [30,64] printed active composites by directly embedding shape memory polymer fibers in an elastomertic matrix using PolyJet printing technique. Through designing the lamina and laminate architecture, the thermomechanical response of their printed composites was programmable. A self-folding/opening box was produced for the demonstration of the shape memory effect of printed active composites. Except for temperature-responsive shape memory polymer composites, water-responsive shape memory polymer composites were also obtained by 3D printing [65]. The printed composite that is composed of cellulose fibrils and acrylamide exhibited shape change on immersion in water. The localized and anisotropic swelling behavior was programmable by controlling the alignment of cellulose fibrils along the printing path, thus the printed composites could morph into given target shapes.

3.3. Nanocomposites

Nanomaterials such as carbon nanotube [66–68], graphene [69,70], graphite [70,71], ceramic [72,73] and metal nanoparticle [74,75] often exhibit unique mechanical, electrical and thermal properties. Thus, the addition of nanomaterials into polymers for printing could enable the creation of high-performance functional composites. The promising results in 3D printing of nanocomposites are summarized in Table 4 and briefly discussed below.

Nanomaterials have been utilized for improving the mechanical properties of printed composite parts. The addition of 5 wt% nano-titanium dioxide(TiO_2) [44], 10 wt% carbon nanofiber [76] or 10 wt% multi-walled carbon nanotube [77] showed a 13.2%, 39% and 7.5% increase in the tensile strength of printed composite parts compared with unfilled polymer parts, respectively, but all printed composite parts showed reduced elongation and more brittle feature in these cases. Lin et al. [78] demonstrated a SLA fabricated graphene oxide/photopolymer composites with a good

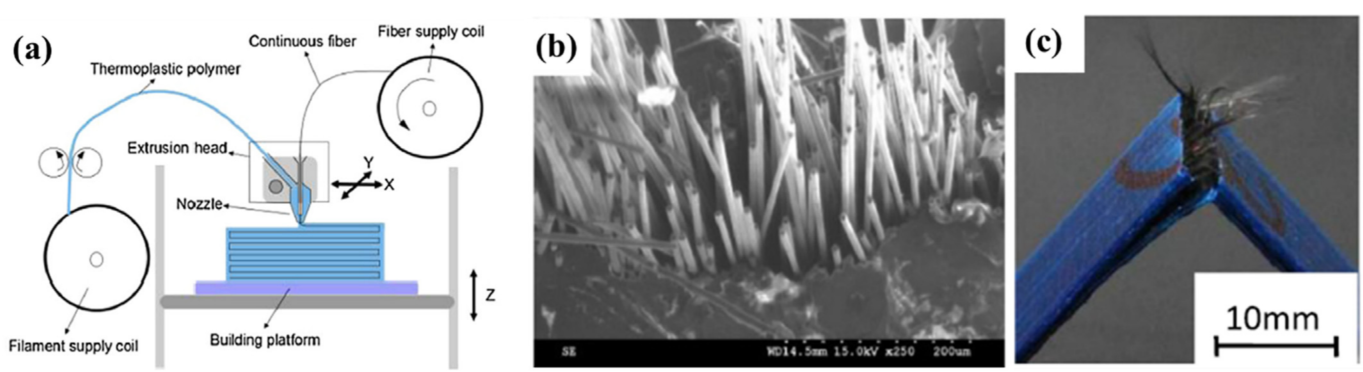

Fig. 7. (a) The setup for the 3D printing of continuous fiber reinforced polymer composites. (b) Interface microstructures (c) Fracture pattern of fractured cross section of carbon fiber reinforced PLA composites. Reprinted with permission from Ref. [58].

Table 4
A summary of techniques and materials used for 3D printing of polymer nanocomposites and properties improvement of resulting composites.

Technique	Materials	Enhancement in properties	Ref.
FDM	TiO_2/ABS	Improved tensile modulus and strength, reduced elongation	[44]
	Carbon nanofiber/ABS		[76]
	Montmorillonite/ABS	Improved tensile strength and modulus, flexural strength and modulus, and thermal stability, reduced thermal expansion coefficient	[85]
	Graphene/ABS	Improved electrical conductivity and thermal stability	[83]
	Carbon nanofiber/Graphite/polystyrene	Better voltammetric characteristics, less capacitive background current	[81]
SLA	CNT/epoxy	Improved tensile strength, reduced elongation	[77]
	Graphene oxide/photopolymer	Improved tensile modulus, strength and elongation	[78]
	TiO_2/epoxy acrylate	Improved tensile strength and modulus, flexural strength, hardness and thermal stability	[84]
	$BaTiO_3$/PEGDA	Improved piezoelectric coefficient	[89]
	CNT/acrylic ester	Improved absorption of electromagnetic energy	[80]
	BST/epoxy	Ultralow thermal conductivity and high energy conversion efficiency	[86]
DLP	Silver/PEGDA	Improved electrical conductivity	[90]
SLS	Carbon black/nylon-12		[82]
	Al_2O_3/polystyrene	Improved tensile strength and impact resistance	[87]
	TiO_2/nylon-12 and graphite/nylon-12	Improved tensile modulus, reduced elongation	[88]
	Silica/Nylon-11		[91]
Solvent-cast direct writing	CNT/PLA	Improved electrical conductivity	[79]

combination of increased strength and increased ductility. Their specimens showed a 62.2% increasing of tensile strength and a 12.8% increasing of elongation with only 0.2% GOs. The authors claimed that the increased ductility was due to the increasing in crystallinity of graphene oxide in reinforced polymers. Except for the mechanical properties improvement, enhanced electrical properties could be obtained by the addition of carbon-based nanomaterials like carbon nanotube [79,80], carbon nanofiber [81], carbon-black [82] and graphene [83]. Wei et al. [83]demonstrated for the first time, a graphene reinforced ABS composite could be FDM printed into computer-designed models(Fig. 8) and the enhanced electrical conductivity was observed. With the loading of 5.6 wt% graphene, the electrical conductivity of ABS nanocomposites showed four orders of magnitude improvement. Furthermore, incorporation of nano-TiO_2 [84] and nano-clay [85] into polymer matrix could greatly improve the thermal stability of printed nanocomposites. In another study, He et al. [86] produced a thermoelectric composite by blending $Bi_{0.5}Sb_{1.5}Te_3$(BST) into photoresins using SLA process, and the resulting composites exhibited an ultralow thermal conductivity of 0.2 W $m^{-1}K^{-1}$, which is favorable for thermoelectric applications.

Homogenous dispersion of nanoparticles into polymers is essential for manufacturing a composite with desired performances by 3D printing technique. To avoid the agglomeration of nanoparticles and ensure a good interfacial bonding between nanoparticles and polymers, chemical surface treatment of nanoparticles [84,87–89] before printing processes was adopted. Nano-Al_2O_3 particles coated with polystyrene by emulsion polymerization were processed by SLS printers [87]. The polystyrene nanocomposites printed by sintering treated particles showed a dense structure and 300% increase in tensile strength, whereas samples produced by untreated particles showed almost no improvements on properties. Adding linker molecules that cross-link with polymer matrix on the surface of nanomaterials is another efficient treatment to improve interfacial bonding between polymers and nanomaterials. For example, after nitric acid treatment, oxidized graphite nanoplatelets were more efficient in enhancing the ultimate strength and Young's modulus of SLS printed nylon/graphite nanocomposites [88]. Very recently, an in-situ generation process of silver nanoparticle after the printing process was proposed. Fantino et al. [90] dissolved the metal salts in the starting polyethylene glycol diacrylate(PEGDA) liquid photopolymer and then digital light processing technique was employed to fabricate 3D structures. The in situ generation of metal nanoparticles was finally induced by a thermal treatment. The electrical conductivity of the produced silver reinforced nanocomposites is three orders of magnitude higher than that of neat polymers. Through using this novel method, printing difficulties that brought by embedding nano-fillers in a polymeric matrix could be addressed to some extent.

3D printing is also an ideal technique to create functionally graded polymer nanocomposites. It realizes this by delivering different volume fractions of nanomaterials to the specific building areas. The ability to design composition enables the optimization of the properties of printed parts. A 3D nylon/nanosilica nanocomposite with spatially varying mechanical properties was produced by Chung et al. [91] with 1D nanosilica composition gradient (Fig. 9) using SLS technique, and the printed components realized the optimized functional values.

4. Application of 3D printed polymer composites

4.1. Biomedical application

With the development of Computed Tomography(CT) and Magnetic Resonance Imaging(MRI) technology, three-dimensional images of tissues and organs have become more informative with higher resolution [92]. Using the acquired image data, patient specific tissues and organs with intricate 3D microarchitecture could be produced by 3D printing technology. Polymer materials currently used for printing in the field of biomedical applications are based on naturally derived polymers(gelatin, alginate, collagen, etc.) or synthetic polymer molecules(polyethylene glycol(PEG), poly lactic-*co*-glycolic acid(PLGA), polyvinyl alcohol(PVA), etc). The desired traits of printable materials for biomedical applications are printability, biocompatibility, good mechanical properties and structural properties [16]. It is essential for successful transplantation and function to ensure the printed 3D parts have a good interaction with endogamous tissues. Table 5 summarizes the materials used by various 3D printing techniques for fabricating bio-composites and their improved properties.

In tissue engineering, scaffolds are critical to provide a physical connection for cell infiltration and proliferation [93]. Traditional technologies are lack of ability to incorporate internal architecture

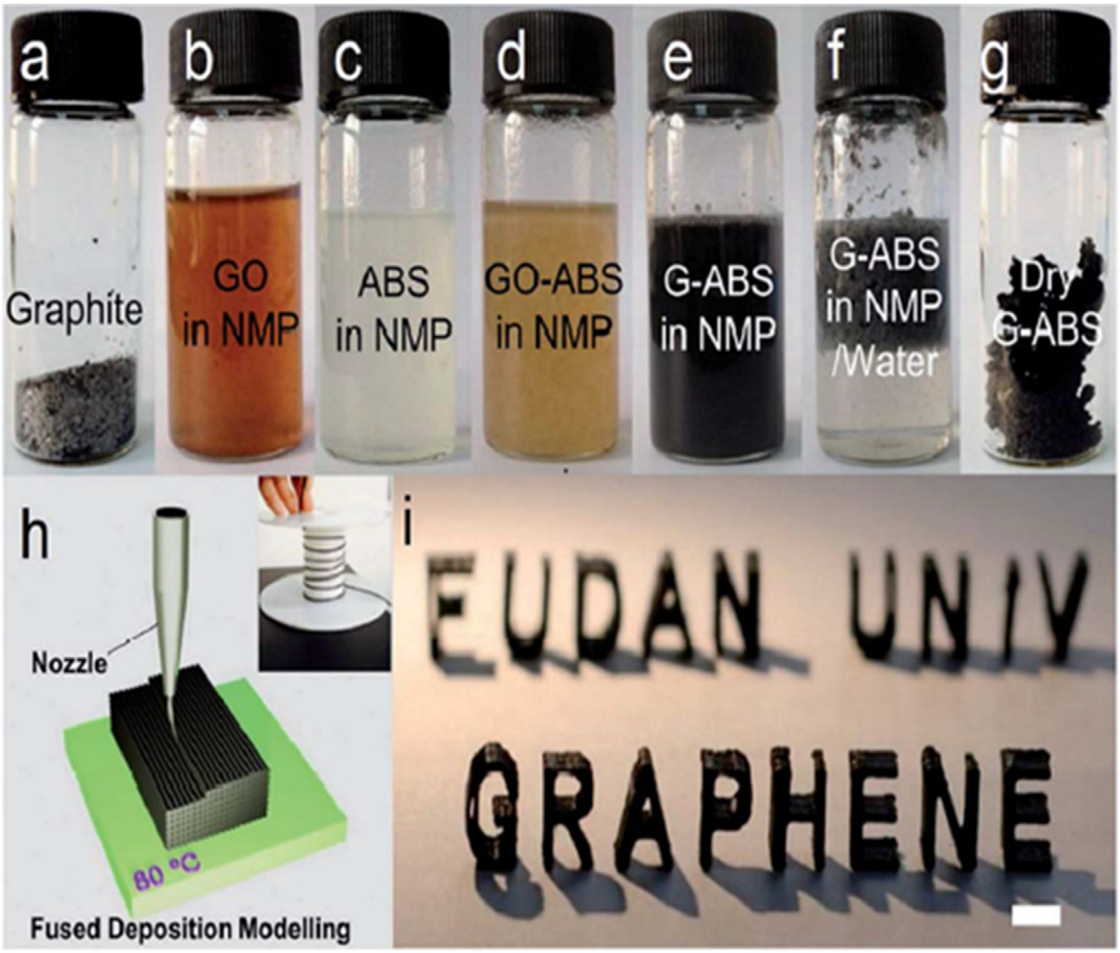

Fig. 8. Picture of (a) Graphite flakes, (b,c) Dispersions of graphene oxide(GO) and ABS in *N*-Methylpyrolidone(NMP) solvent, (d,e) A homogeneous mixture of GO-ABS in NMP before and after chemical reduction, (f) Graphene(G)-ABS coagulations obtained after isolation (e) with water, (g) G-ABS composite powder after washing and drying, (h) Schematic illustration of fused deposition modeling 3D printing process. Inset is the graphene-based filament winding on a roller, (i) A typical 3D printed model using 3.8 wt% G-ABS composite filament, scale bar: 1 cm. Reprinted with permission from Ref. [83].

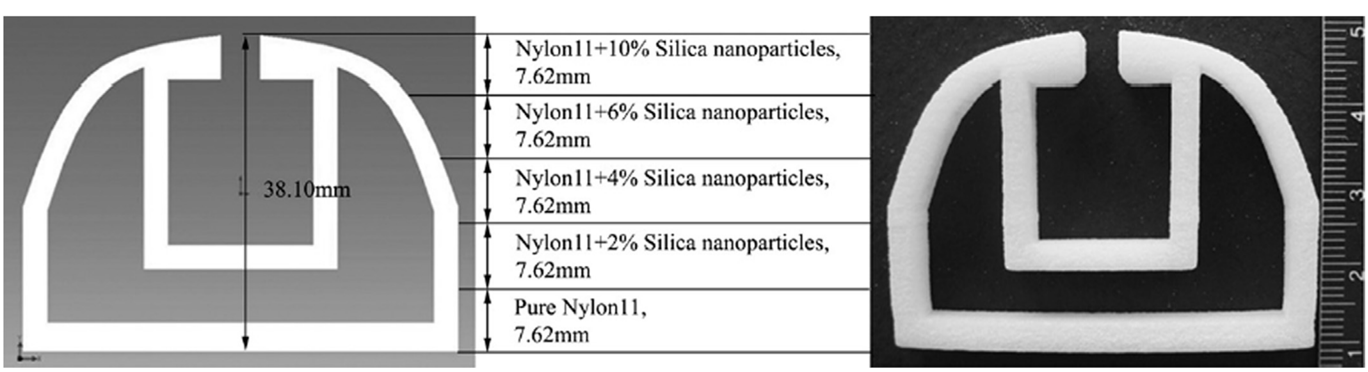

Fig. 9. Schematic description and physical image of a printed compliant gripper. Reprinted with permission from Ref. [91].

and control porosity of scaffolds. 3D printing has addressed these problems by allowing for control of the pore size and pore distribution of scaffolds [94]. Printing of composite scaffolds with high biocompatibility was achieved through adding bioactive particles into polymer. The biodegradable and biocompatible polymers could maintain the toughness of the printed scaffolds while the brittle bioceramic particles could increase the biocompatibility. Serra et al. [95] employed a nozzle-based printing system to create highly porous PLA/bioglass 3-D biodegradable scaffolds. Fig. 10(a) and (b) is the scanning electron microscopy of printed 3-D scaffolds, which shows a controlled and repetitive architecture. The incorporation of glass particles improved cell adhesion of PLA polymer by increasing both the roughness and the hydrophilicity of the scaffolds. The immunofluorescence studies clearly demonstrated this result, as shown in Fig. 10(c) and (d). Also, other synthetic calcium phosphates such as hydroxyapatite(HA) [96–99] and

Table 5
Composites fabricated by various 3D printing techniques for bio-medical application.

Technique	Materials	Applications	Ref.
3D plotting	TCP/PCL	Biodegradable scaffold with improved hydrophilicity and cell adhesion, and improved compressive strength	[102]
	TCP/alginate		[101]
	HA/PLA		[99]
	Bioglass/PEG/PLA		[95]
	CNT/alginate		[103]
	Silica/collagen/alginate		[105]
	HA/CNT/PCL	Biodegradable scaffold with improved electrical conductivity allowing the application of electric stimuli	[96]
	Graphene/PLGA		[104]
	Fe_3O_4/bioactive glass/PCL	Biodegradable scaffold that can generate local heat for hyperthermia therapy	[107]
	Alginate/epoxy	Artificial hydrogel meniscus cartilage	[110]
	Agar/alginate		[111]
	Cell seeded hydrogel/silver nanoparticle	Bionic ear	[112]
	Cell seeded alginate/PCL/PEG		[114]
	Cell seeded hydrogel/PCL		[113]
	Cell seeded gelatin/collagen	Vasculature	[115]
	Cell seeded gelatin/alginate	Aortic valve	[116]
	Cell seeded alginate/nanofibrillated cellulose	Cartilage constructs	[117]
	Cell seeded multicellular spheroids/agarose	Liver tissue constructs	[118]
FDM	HA/PLA	Biodegradable scaffold with improved crack resistance during cyclic loading	[108]
	HA/PEG/PLA	Biodegradable scaffold with improved hydrophilicity and cell adhesion, and improved compressive strength	[97]
	HA/TCP/PLGA		[100]
SLS	HA/PCL		[98]
	$CaSiO_3$/PVA		[106]

tricalcium phosphate(TCP) [100–102] have been used extensively by tissue engineers to fabricate biocompatible composite scaffolds through variable 3D printing technologies. These materials effectively generate nano/microscale topology in composite scaffolds and greatly improve the hydrophilicity of the scaffolds, thus increase the bioactivity of scaffolds. In vivo studies have also been done with the printed composite scaffolds. For example, FDM printed PLGA/TCP/HA composite scaffolds have been successfully implanted into rabbit femoral bone defect to support bone deposition and the scaffolds then successfully biodegraded over 12

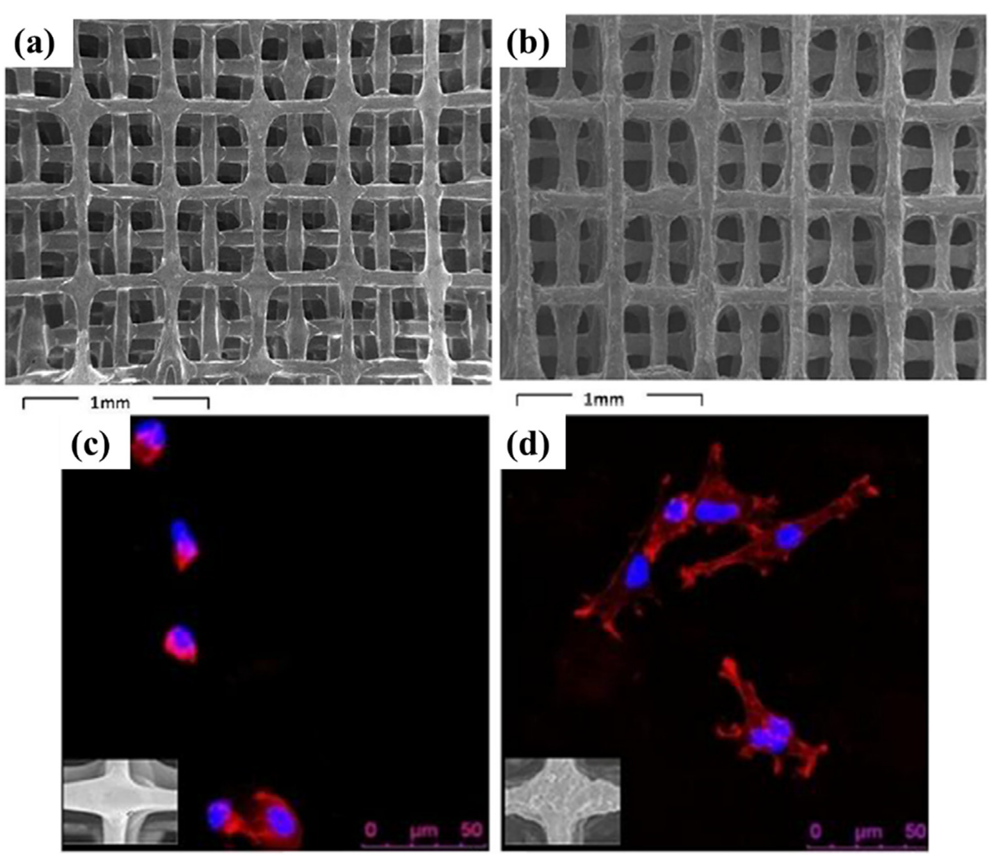

Fig. 10. Top view SEM micrographs of 3-D printed (a) PLA/PEG and (b)PLA/PEG/G5 scaffolds; Fluorescence images of attached cells on (c) PLA/PEG and (d) PLA/PEG/G5 scaffolds. Reprinted with permission from Ref. [95].

weeks [100].

Except for biocompatibility, good mechanical properties are important for 3D printed scaffolds. Good mechanical stability would help scaffolds to support cellular activity. To improve the strength of scaffolds, most of composite scaffolds were prepared by blending TCP or HA with polymer matrix. For example, Davila et al. [102] fabricated the composite scaffolds using a mixture of PCL and TCP by 3D extrusion printing process. In the case of scaffold with 20 wt% TCP, the compressive modulus increased 107% compared to that of the pure polymer scaffolds. Xia et al. [98] selected hydroxyapatite(HA) as reinforcement and fabricated a PCL based scaffold by SLS technique. A 130% improvement on the compressive strength of fabricated composite scaffolds was demonstrated. Other additives such as CNT [103], graphene [104], silica [105], $CaSiO_3$ [106] and Fe_3O_4 [107] were also reported to increase the compressive modulus/strength of composite scaffolds. In a recent study, the fatigue behavior of a 3D printed PLA/HA composite scaffold was investigated [108]. During the cyclic fatigue test, the formation of defects and the accumulation of plastic deformation were detected. It is demonstrated that the introduction of HA particles reduced the rate of accumulation of defects and increased the crack resistance of composite scaffolds. Besides improved mechanical properties, incorporation of Fe_3O_4 or graphene into the printed scaffolds also provide special thermal and electrical properties. Magnetic Fe_3O_4 nanoparticles in printed scaffolds can generate local heat under an alternating magnetic field thus facilitate the cancer treatment by hyperthermia therapy [107]. Graphene could largely increase the electrical conductivity of printed scaffolds thus allows for the fabrication of bioelectronics [104].

Many soft tissues can be thought of as fiber reinforced hydrogel composites like articular cartilage. A two-step process to print fiber reinforced hydrogel composites was first developed, in which fiber scaffold was printed first, and then the composite structure was completed by immersing the scaffold into hydrogel precursor solutions and polymerizing the gel [109]. Recently, one step printing process of hydrogel composites was developed [110]. A 3D-bioplotter with a commercial UV-curing system was used to deposit and cure the fiber reinforcement and matrix at the same time. An artificial hydrogel meniscus cartilage was successfully printed to demonstrate the potential real-world bio-application of this composite structure. With the control of fiber distribution in composites, printed parts could exhibit a variable swelling behavior and mechanical property. An extremely tough composite hydrogel for meniscus was also obtained by one step printing process [111]. The agar and alginate were pre-mixed and printed by an extrusion-based printer. The entanglement of the alginate toughens the agar gel and largely improves the mechanical performance of printed composite.

Biofabrication using living cells for tissue and organ transplantation is another new paradigm of 3D printed polymer composite applications in the biomedical industry. Several tissues and organs, such as ears [112–114], vasculatures [115], aortic valves [116], cartilage constructs [117] and liver tissue constructs [118] have already been successfully printed to meet the functionality requirement for transplantation. Lee et al. [114] employed a 3D bioplotter to print ear with complex structure, which consists of PCL and cell-seeded hydrogel. The printed composite ear satisfies expectations for both the geometry and anatomy of the native ear. Tissue generations have also successfully occurred in the fabricated composite structure. In another study, silver nanoparticle was incorporated into the cell-seeded hydrogel matrix to enhance the auditory sensing for printed ear(Fig. 11) [112].

4.2. Electronics

The use of 3D printing technology can provide geometrically appropriate electronic prototypes with reduced development time [119]. When combined with electrical conductive materials, 3D printed polymer composites are able to function as electronic devices that can be used in numbers of ways. Electronic sensors ranging from piezoresistive sensors to capacitive sensors were fabricated by Simon. et al. [120] through FDM printing of carbon black/PCL composites, as shown in Fig. 12. The piezoresistive sensors were able to sense mechanical flexing through the change of electrical resistances, and the capacitive sensors could be printed as a part of custom interface devices or embedded into smart vessels to detect the presence and absence of water. Similarly, the 3D printing of CNT/epoxy nanocomposites with ultraviolet-assisted direct write technique has also been described for application in printed electronics [121]. Utilizing this composite material, a piezoresistive sensor with high electromechanical sensitivity(gauge factor~22) was realized. These functional sensors demonstrated the promising application of 3D printing technology on electronic devices.

Traditional printed electrical circuits were based on 2D flat surface printing[122]. For example, silver-capsuled composite particles were developed as conductive toners for electrostatic printing on flexible substrate[123]. The printed conductive tracks have a conductivity of ~10^{-4} Ω cm. However, in real-world applications, electronic prototypes may need to be embedded within more appropriate shapes in order to authenticate prototype products earlier in the development cycle[124]. Recently, efforts have been made to develop 3D structural electronics. A 3D connector of an electrical circuit was fabricated by digital light processing printer using silver and cross-linked photopolymer [31], as shown in Fig. 13. The 3D porous structure fabricated using the printable oil-in-water emulsion was dipped in a dispersion of silver nanoparticles, then sintered to obtain conductive percolation paths. The porosities and total surface areas of printed 3D structures can be controlled by altering the processing parameters, thus the electrical conductivity could be potentially designed. Liquid deposition modeling printing of CNT/PLA composite was also adopted to fabricate 3D structural electronics [32]. Direct deposition of a homogeneous dispersion of CNT in PLA using a high volatility solvent was performed, and then a rigid 3D microstructure was formed after the evaporation of the solvent. A 3D flexible woven conductive structure printed using this material was employed to set up a simple electrical circuit to turn on a commercial LED, thus giving a further practical demonstration of the potential applicability in the field of microelectronics. There has also been efforts to develop 3D electronic devices by encapsulating metal wires into polymer matrix during the printing process. This printing process is similar to the technique employed to fabricate continuous fiber reinforced composites. Copper wires and molten styrene block copolymers were separately supplied and co-extruded. The printed two-phase composites were demonstrated to be used as an open membrane switch [33], which is activated when the membrane is deformed by pressure contact, resulting in the copper wires on adjacent polymer layers to be shorted together.

4.3. Aerospace applications

Most aerospace components have complex geometries which are time-consuming and costly to be manufactured. Therefore, 3D printing is highly suitable for development of these components. Up to now, most aerospace components like engine exhaust and turbine blade are 3D-printed with metal materials[125, 126], since usually metals are stronger and more flame retardant than polymer

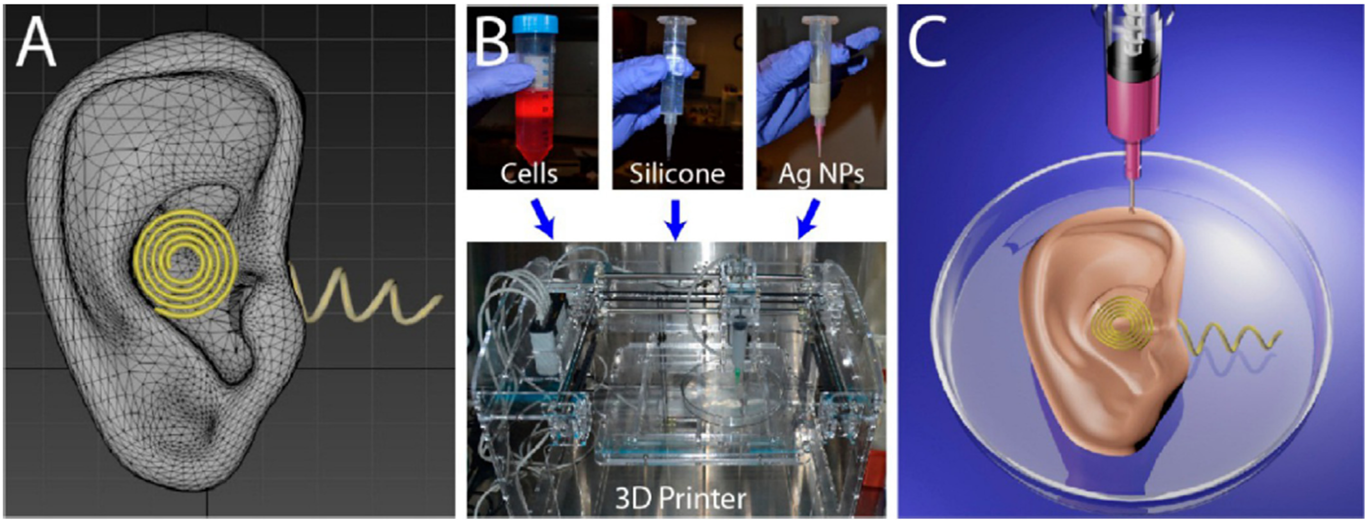

Fig. 11. Three-dimensional interweaving of biology and electronics via additive manufacturing to generate a bionic ear. (A) CAD drawing of the bionic ear. (B) (top) Optical images of the functional materials, including biological (chondrocytes), structural (silicone), and electronic (AgNP infused silicone) used to form the bionic ear. (bottom) A 3D printer used for the printing process. (C) Illustration of the 3D printed bionic ear. Reprinted with permission from Ref. [112].

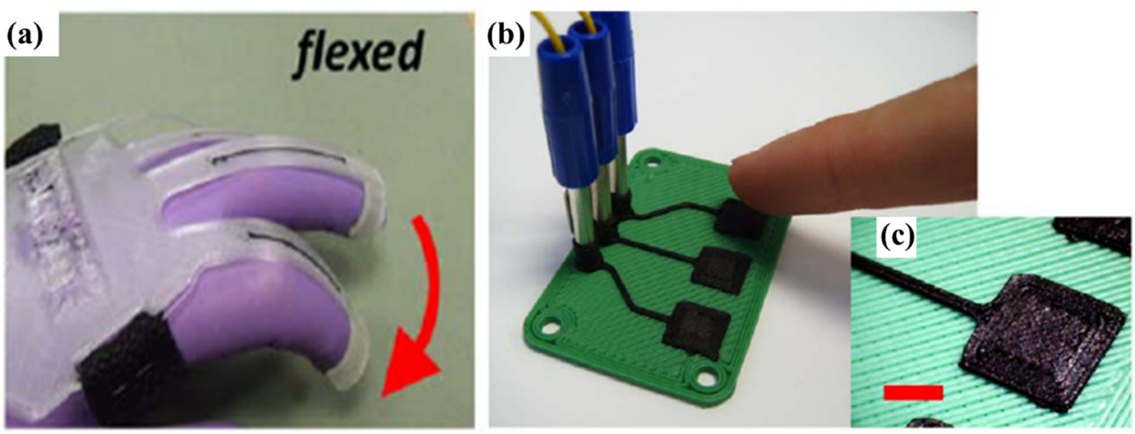

Fig. 12. The 3D printed carbon black/PCL composite for (a) Piezoresistive sensors (b) Capacitive sensors (c)A macro image of the printed sensor pads (scale bar 5 mm). Reprinted with permission from Ref. [120].

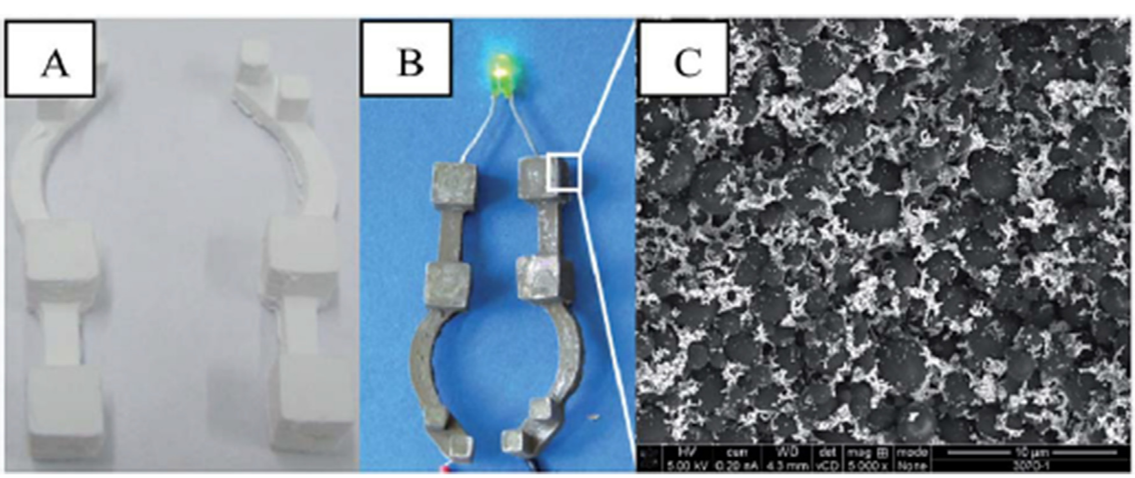

Fig. 13. Images of electric circuit printed from emulsion of 70 wt% dipropylenglycol diacrylate as oil phase (A) image of the clean porous 3D structure (B)image of the porous structure after inserting Ag in silver dispersion, sintering and connecting to 3 V and LED, (C) UHR-SEM cross-section image of the printed structure with embedded Ag nanoparticles. Reprinted with permission from Ref. [31].

materials. Recently, several research institutes are exploring ways to apply 3D printing of polymer composites to aerospace applications due to the fuel efficiency of usage of polymer composites.

Airfoil and propeller have been demonstrated to be printed with glass fiber and carbon fiber reinforced photopolymer composites using a UV-assisted 3D printing system(Fig. 14)[127]. High-fidelity replicas of the digital model and excellent reproducibility were achieved by using these materials to produce aerospace components. Excellent interlayer bonding between successive layers favors the good mechanical property in the printed components. Recently, polymer composites that can withstand a high temperature were printed for aerospace applications. Glenn Research Center[128] used FDM process to fabricate an inlet guide vane with Ultem 1000 and chopped carbon fiber, and the operating temperature of this composite structure could reach 400 °F. Similarly, Impossible Objects company[129] also announced its ability to manufacture high performance carbon fiber reinforced polyether ether ketone(PEEK) composites for aerospace applications. The fabricated composite parts have a heat resistance of 482 °F and are 50% lighter than traditional aluminum parts while maintaining 2/3rds of the aluminum's strength. Airfoil, rotor support arm and air intake have been demonstrated to be printed with this composite material.

5. Future research

Although 3D printing of polymer composites has undergone significant developments in recent years, it is still not widely

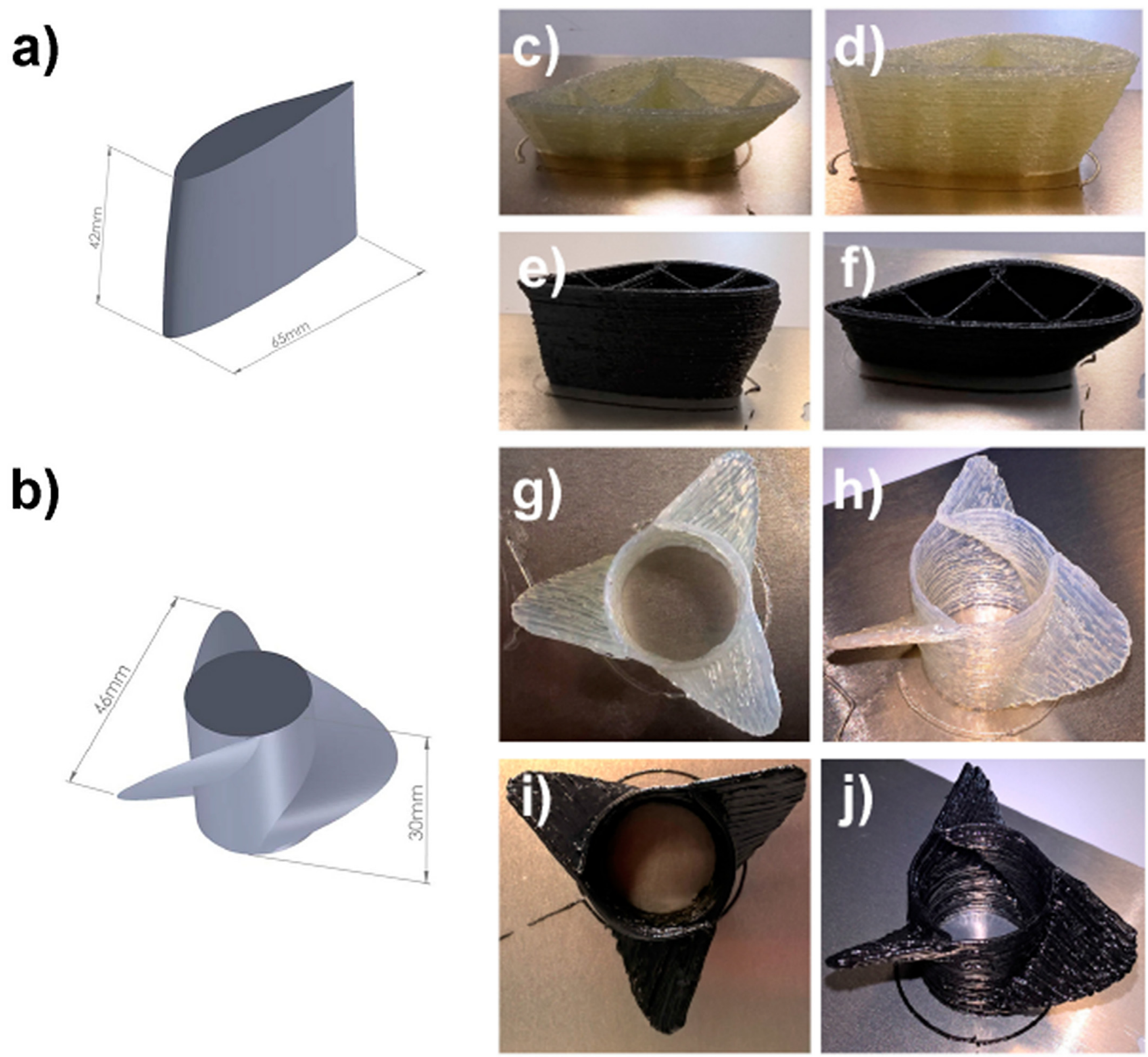

Fig. 14. Axonometric projection of the airfoil (a) and the propeller (b) 3D models used to demonstrate the printability of the glass fiber and carbon fiber composite formulations developed in this work. UV-3D printed reproduction of the airfoil (c–f) and the propeller (g–j) 3D models based on the glass fiber (c, d, g, h) and carbon fiber (e, f, i, j) polymer composite formulations developed. Reprinted with permission from Ref.[127].

accepted by most industries. Several limitations of this technology need to be overcome.

- Material: The wide application of 3D printing is severely limited by printable materials. Currently, only thermoplastic polymer with low glass transition temperature and suitable melting viscosity, powder formed materials and a few photopolymers could be used in 3D printing. However, these limited materials could not meet the variety requirements of industry application and thus the diversity of materials must increase. Synthesis of matrix materials with special properties, discover of new reinforcement and discover of suitable mixing composition are critical to increase the versatility of composites printing technology. Sustainable materials are also promising to be developed to reduce material cost and environmental impact.
- Performance: Although reinforcement helps improve the performance of polymer composites, compared with polymer composites manufactured by traditional molding methods, most of the printed composites still have low mechanical strength and are not able to meet the functional requirement. Therefore, additional post-treatment steps involving infiltration or consolidation have been used to improve the performance of printed products. However, these steps further increase the cost and processing time. A main reason for the low mechanical strength is the presence of void in printed parts. The addition of reinforcement may further increase the porosity due to the poor interfacial bonding with matrix. Therefore the improvement brought by reinforcement may be compensated by the induced porosity. How to eliminate the formation of void during printing and ensure good interfacial bonding between matrix and reinforcement requires further significant research. Moreover, the repeatability and consistency of manufactured parts cannot be guaranteed by printing, thus approaches to ensure the uniform properties of printed parts need to be investigated in depth.
- Machine: Most printing processes are time-consuming now, and it is difficult to fabricate parts that have large volume. These inhibit their industry adoption. New printing techniques based on scalable and fast processing of materials should be developed. For example, digital light processing is an efficient improvement of SLA process. A layer of photopolymer is fabricated during one time projection, which greatly reduces the processing time. Similar improvement should be done for other techniques. Another area of growth centers on the need for feedback systems. If an error occurs during printing now, the process needs to be suspended, which causes the waste of time and materials. Feedback systems should be built in the printer to have a response to the process change. Additional progress for 3D printers is to increase the printing resolution without extending printing time or sacrificing geometry complexity of products.

While many limitations still exist now, 3D printing of polymer composites already develops fast in recent years. As evidenced by the above-mentioned publications, researchers are exploring new materials and new application of 3D printing of polymer composites. This paper gives a platform based on which further researchers will progress more. In terms of materials, processing control, processing scalability and product performance, there are new avenues of research with 3D printing of polymer composites.

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