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The 3D Printed Filaments Manufactured by various Methods and Composites

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ABSTRACT

New metal/polymer composite filaments for fused deposition modeling (FDM) processes were developed in order to observe the thermo-mechanical properties of the new filaments. The acrylonitrile butadiene styrene (ABS) thermoplastic was mixed with copper and iron particles. The percent loading of the metal powder was varied to confirm the effects of metal particles on the thermo-mechanical properties of the filament, such as tensile strength and thermal conductivity. The printing parameters such as temperature and fill density were also varied to see the effects of the parameters on the tensile strength of the final product which was made with the FDM process. As a result of this study, it was confirmed that the tensile strength of the composites is decreased by increasing the loading of metal particles. Additionally, the thermal conductivity of the metal/polymer composite filament was improved by increasing the metal content. It is believed that the metal/polymer filament could be used to print metal and large-scale 3-dimensional (3D) structures without any distortion by the thermal expansion of thermoplastics. Commodity thermoplastics and thermoplastic composites are staples in Additive Manufacturing (AM). Their use is widespread and accounts for the largest volume of 3D printed materials.

Key words: Metal/polymer composite filament, 3D printing, fused deposition, Modeling, Additive Manufacturing, Thermoplastic Composites

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INTRODUCTION

Polymer materials are of central importance to Additive manufacturing (AM) and eventually for fabrication of original equipment manufacturer (OEM) parts. The majority of materials currently in use can be divided into two classes, thermoplastics and thermoplastic composites. The methods for employing high volume polymer thermoset and elastomer materials for AM are just recently emerging. The aim of this review paper is to provide an overview of recent advances of thermoplastic and composite materials in 3D printing, including the need to improve their performance and properties which is key to growing the technological impact and marketability of polymer-based AM. Additive manufacturing or 3D-printing is characterized as a process of joining layers of materials to create 3D objects from a Surface Tessellation Language (.stl) file designed from a computer-aided design (CAD). AM was first demonstrated by Charles Hull in 1986 who created an apparatus capable of generating 3D objects. Since then, various technologies have been introduced in additive manufacturing. There is a wide range of 3D printing technologies and processes such as fused deposition modeling (FDM), electron beam melting (EBM), stereo lithography (SLA), selective laser melting (SLM), selective laser sintering (SLS), ink-jet printing, aerosol-jetting, micro-dispensing deposition write (MDDW), and oil pressure dispensing. An important challenge in the field of AM is the lack of advanced polymer materials and available Nano composites to match the performance and fabrication requirements. The introduction and development of novel polymers, additives, and other thermoplastic composites for AM, aims to extend the impact of AM on the fabrication of end-product components and real parts replacement. A common issue in using commodity thermoplastic products in AM is their lack of strength and functionality as high load-bearing parts.

The use of engineering plastics and high-performance polymers (HPP) is desirable, but the cost and the requirement for more demanding processing conditions (such as higher temperatures) present a challenge, especially if high build volumes are desired. Common matrix materials used in AM are thermoplastic materials, to which filler components such as fiber-, particle- or nanoparticle-based materials can be added for new material formulations. Traditional polymer blending has been used to combine the strength of two polymers or reinforce the weaker polymer with a highperformance polymer.

VARIOUS ADDITIVE MANUFACTURING PROCESSES

Abbreviations	**AM Type**	**Defination**
FDM	Fused Deposition Modeling	FDM involves deposition of melt extruded layers of material through a nozzle using a feedstock filament.
SLS	Selective Laser Sintering	SLS involves sintering of layer-bylayer Powdered materials.
SLM	Selective Laser Melting	SLM involves polymer powder and laser beam-based manufacturing Process.
LDM	Liquid Deposition Modeling	LDM deposits polymer using a solvent or a UV photo curable resin.

Materials Used for Selective Laser Sintering (SLS)

The most common materials used in SLS are thermoplastic polymers such as polyurethane (TPU), polycaprolactone (PCL) and polyamide (PA). These thermoplastics can be filled with glass, aluminum, or fibrous materials to cater to certain applications. Industry-grade SLS machines also use ceramics and metals. A drawback is that the surface porosity is high in SLS-printed products since the sintering process of SLS creates a porous mass of these materials without reaching the point of liquefaction (melting). Otherwise, it will be called Selective Laser Melting (SLM). SLS machines are more expensive than other 3D printing machines due to the high cost entailed by using a high-power laser.

Selective Laser Sintering (SLS)

Selective Laser Sintering (SLS) or Selective Laser Beam Melting (SLBM) is a Powder Bed Fusion (PBF) technique through directed heating. It has become the most popular PBF process since it was the first technique to be commercialized under this category of AM. In this method, a power-based heating source such as CO2 or Nd-YAG laser is selectively aimed at a bed of powder metal, thermoset or Thermoplastic that created a solid object made of fused powder. A hybrid additive manufacturing technique for SLS has been introduced in the research of Wudy and Drummer in which combined reactive liquids of thermoset resin and thermoplastics were used. Thermoplastics, such as polyamide 12, polyamide 11, polypropylene and thermoplastic elastomers, are the conventional materials being used in a typical SLS setup. The chamber is usually heated just below the melting point of the powder material before the sintering process is applied. The printing is complete when the entire object is formed inside the printing chamber. To get the final shape, the excess powder is 26 shaken off. The print quality of SLS depends on the power and the scan speed of the laser, the particle size of the powder material and the layer thickness.

Selective Laser Melting (SLM)

Selective laser melting (SLM) uses a high-power-density fiber laser (ytterbium fiber laser) to melt and fuse metallic powders to form a 3-dimensional part. This process is similar to Selective Laser Sintering (SLS); the only difference being that the material is melted while in SLS it is heated to a temperature just enough to allow fusion of the powder. First, the CAD model is sliced into 2-dimensional layers and transferred to the SLM machine. Moreover, then, an even layer of powder material is distributed using a coater. Under a tightly-controlled inert atmosphere, the laser then scans and fuses the layer following the geometric information of the sliced model. The substrate is then lowered by one layer thickness. These scanning/fusing and layering processes are repeated until the final part are complete. The finished part is removed from the powder bed and undergoes post-processing steps such as heat treatment.

Laser beam melting (LBM)

Laser beam melting (LBM) is another additive manufacturing technology that allows layer-based Production of geometrically complex parts from metal powder. The CAD drawing is sliced into 2D+1D representation. A thin powder layer of 20 µm is deposited on top of the platform and melted using the two-dimensional geometry. After the process is done, the platform is lowered and allows another layer of powder to be deposited on top. The process is repeated until the whole 3D objects if finished.

Materials used for SLM and LBM

Polystyrene has been used to produce spherical micron-sized particles having adequate flow ability for LBM and SLM printing. The process included wet grinding, rounding, and dry coating. Drummer *et al.* investigated the density and flow ability of new and used PA12 powders for SLM printing. Other than the particle parameters, they found out that the density of laser-molten parts depends on the heating rate and applied coating parameters and mechanisms. The authors also used polyamide 12 to investigate the influence of different ways of energy input on the resulting melt pool for SLM printing. Also, time dependent effects during exposure have been studied. Lastly, they reported on the morphological structure of PA12 blended with PEG and polyvinyl alcohol. Laumer *et al.* reported on several requirements for compatibility between different polymeric materials for LBM printing of polymers. They also provide a matrix of possible material combinations for composite structures. Schmidt *et al.* developed a process chain to produce spherical polymeric particles made out of poly (butylene terephthalate) (PBT). Their process included wet grinding of polymer micro particles, and then the particle is rounded in a heated downer reactor, and lastly, the particle is dry-coated with fumed silica.

Liquid Deposition Modeling (LDM)

Liquid deposition modeling (LDM), also called direct ink writing, has been garnering more Attention in recent years. Similar to FDM, this technique fabricates 3D structures by consecutive addition of layers of extradites into computer-designed geometries; however, LDM involves the extrusion of viscous liquids rather than melted filaments. The preparation of viscous liquids even at 31 room temperature offers LDM the highest versatility among all the 3D printing techniques. The disadvantage of LDM is the selection of solvent and polymer concentration to achieve appropriate flow parameters without compromising rapid evaporation of the solvent in the solution. Printing parameters must be optimized to achieve the desired material extrusion of the dissolved polymer.

Materials Used for Liquid Deposition Modeling (LDM)

A wide range of materials, including ceramics, metals, hydrogels, carbon-based materials, polymers, and biomaterials can be 3D printed using LDM, once it is successfully formulated into a viscous liquid. Very porous structures can be fabricated using this method.3D-printed micro-lattice of grapheme aerogel that possesses a porous structure. Minas *et al.* demonstrated the fabrication of hierarchical porous ceramic foams with LDM, in which viscous liquid ink is prepared by emulsified ceramic powders and PVA binders. Shah has reported 3D printing of various metal materials into complex architectures where metal powders were suspended in a solvent mixture to form viscous liquid inks. A PDMS elastomer was filled with silica nanoparticles to form viscous inks which were 3D printed into different lattice structures to exhibit designed mechanical properties. In order to 32 successfully 3D print by LDM the highly filled materials described above (vide supra), materials with thixotropic rheological properties are required. When extruded from the nozzle, inks are under high shear stress; thus, shear thinning behavior is required so that the flowing of ink is consistent and controllable. The deposited ink should be able to retain its shape, and also be mechanically strong to support the weight of subsequent layers without deforming its original shape. Owing to the high versatility of materials processed in LDM, the curing procedures are very diversified and determined by the nature of the ink materials. Liquid deposition modeling includes the selection of solvent and polymer to be combined in a Solution. Setup for LDM must be optimized in order to ensure flow ability of the solution during the extrusion and rapid evaporation of the solvent after the extrusion.

Comparison of FDM, SLS, and LDM

The comparison of thermoplastic composites used in FDM, SLS, and LDM. FDM has been used in extrusion and layer-by-layer deposition of thermoplastic material. The process of mixing fillers with polymers at its molten state is much easier as compared using the dissolution precipitation method. Therefore, a wide range of fillers are available for FDM such as particle, fiber, nanomaterial and polymer blends except for natural fibers. Natural fibers tend to degrade at elevated temperatures [40]. SLS creates a solid object by fusing powder materials through directed heating and partial melting. An SLS printer has a required particle size of powder to be used in printing objects. This is the reason why the applicable fillers for SLS are limited to the particle, short fiber, and nanomaterial. Powder materials used in SLS can be categorized into separate grains, composite grains, and coated grains. Liquid deposition modeling (LDM) extrudes viscous liquid instead of melted filaments. Preparation of viscous liquids with LDM is the challenging part since it involves proper selection of solvents applicable to polymers and its fillers without compromising the rheological requirements for printing such as storage modulus ($G' > 1{,}000$Pa) and yield stress (_ > 200Pa) [81].

CHALLENGES WITH THE USE OF THERMOPLASTIC COMPOSITES IN AM

Defects due to fillers. The formation of voids and micro voids is a common problem when fillers are introduced into a thermoplastic. Poor adhesion and vaporization of volatile compounds with fillers themselves during melt blending and material extrusion causes the formation of voids and microvoids. The quality of the finished product depends on the quality of mixing process. Under stress loading, defects propagate crack nucleation and can cause inconsistent quality and properties in printed parts [40]. For particle-based and nanomaterial-based composites, agglomeration and non-uniform dispersion is a common problem. Further increase in filler loading in composites with agglomerations and non-

uniform dispersion may not be cost-effective especially with higher cost fillers and Nano fillers. Also, a variety of particle size 47 within the thermoplastic composites can cause inconsistent thermo-mechanical properties of the thermoplastic composites. For fiber-reinforced based composites, commercially available 3D printers are limited to printing short fiber reinforced thermoplastic composites. The use of continuous fiber reinforcement in a printed part requires complex print head designs and computer programming. Also, poor adhesion between fiber and thermoplastic can cause inconsistent quality and properties of the thermoplastic composites. Preprocessing is needed to ensure good fiber-matrix adhesion. Natural fibers can thermally degrade at an elevated
Temperature that caused the formation of voids.

3D Printer Design Limitations. Commercially available printers are not designed to handle
Thermoplastic composites with different filler types and loadings. For SLS printing, thermoplastic composites should be prepared into a fine powder in order to ensure good flow ability during printing. For FDM, it is difficult to introduced fibers in 3D printing due to limited types of the commercially available nozzle. The use of more anisotropic and larger fillers can cause nozzle clogging and abrasion [32]. In LDM, the use of a highly volatile solvent is needed to allow the material to flow through the nozzle. Further research is needed in order to explore the applicability of other solvents in LDM. It is important to consider the type of solvents that are being used since it is highly dependent on the polymer solubility.

MANUFACTURING OF METAL/POLYMER COMPOSITE FILAMENTS

Copper and iron powders were mixed with the thermoplastics ABS. The size of copper and iron particles was less than 24 lm (_625 mesh, 99%; Alfa Aesar) and 43 lm (_325 mesh, 98%; Alfa Aesar), respectively. The contents of metal powder were varied from 10 wt. % to 50 wt.% in order to observe the effects of metal particles on thermo mechanical properties of the final products. The metal/ABS mixture was formed into pellets, and the pellets were extruded by a filament extruder (Filastruder) to make a metal/polymer composite filament for a FDM 3D printer. The final diameter of the filaments was 1.75 mm. Metal filaments were printed by a NP-Mendel (Open creators) which is a type of FDM 3D printer. Dog bone-shaped structures were manufactured to analyze their tensile properties. Tensile properties of the filaments were measured by a tensile strength tester (Micro Tester 5548; Instron). The specimens were designed according to ASTM D638, and the speed of testing was set to 50 mm/min. Microstructures of the cutting plane of specimens were analyzed to see the effect of metal particles on tensile properties. A video optical microscope (SV-35; Sometech) was used to observe the microstructure of specimens.

SUMMARY AND CONCLUSION

In this review, an overview and discussion of the different types of processing techniques and materials for 3D printed thermoplastic composites are provided. The most common method used in the preparation of thermoplastic composite materials is melt blending and extrusion due to its simple approach and ease of preparation. Preparation of composite powders or coated powders can be performed either by melt blending/milling or dissolution-precipitation. Composite or coated powders improve the adhesion between fillers and matrix. Current efforts focus on the improvement of the print-head design of FDM printers through in-nozzle impregnation and dual print heads. In the future, this should allow for the 3D printing of 48 a larger variety of thermoplastic composites without the need for preprocessing or mixing. Also, new variants of AM techniques (such as LDM) are developed that allow for with higher filler loading in thermoplastic composites. Because of the need for solvents, the application of LDM in 3D printing thermoplastic composites remains limited, and further research is needed.

While thermoplastics and thermoplastic composites have been successfully applied in AM (including particle, fiber-, nanomaterial, and polymer-polymer composites) the application of AM in filled systems still needs to be optimized for each new type of printer head that is being developed. Even with the addition of fillers, there is no assurance that the thermo-mechanical properties of the printed material are bound to improve. As noted, filler addition can result in the unpredictable formation of defects. To control and reduce defect formation, it is important to understand the nature of compatibilization, miscibility, and surface migration to improve the properties. Aggregation and agglomeration of fillers is a frequent problem in filled systems. Also, understanding the quenching properties and their effect on the semi-crystalline nature of the extruded material and adhesion between deposited layers is a primary concern.

In summary, the extension of AM to filled polymers holds the promise of novel applications and improved properties of AM fabricated parts. However, the required optimization of process parameters depending on materials system and AM techniques presents a challenge for the AM of filled polymers and defines the need for future research.

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