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## Highly oriented carbon fiber–polymer composites via additive manufacturing



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#### ABSTRACT

Additive manufacturing is distinguished from traditional manufacturing techniques such as casting and machining by its ability to handle complex shapes with great design flexibility and without the typical waste. Although this technique has been mainly used for rapid prototyping, interest is growing in direct manufacture of actual parts. For wide spread application of 3D additive manufacturing, both techniques and feedstock materials require improvements to meet the mechanical requirements of load-bearing components. Here, we investigated short fiber (0.2–0.4 mm) reinforced acrylonitrile–butadiene–styrene composites as a feedstock for 3D-printing in terms of their processibility, microstructure and mechanical performance. The additive components are also compared with traditional compression molded composites. The tensile strength and modulus of 3D-printed samples increased  $\sim$ 115% and  $\sim$ 700%, respectively. 3D-printing yielded samples with very high fiber orientation in the printing direction (up to 91.5%), whereas, compression molding process yielded samples with significantly lower fiber orientation. Microstructure–mechanical property relationships revealed that although a relatively high porosity is observed in 3D-printed composites as compared to those produced by the conventional compression molding technique, they both exhibited comparable tensile strength and modulus. This phenomenon is explained based on the changes in fiber orientation, dispersion and void formation.

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#### 1. Introduction

Rapid prototyping (RP) is a technology in which a part can be built layer by layer to a desired geometry based on a computeraided design (CAD) model. With RP, complex parts can easily be built in reasonable timeframes [\[1–3\]](#page--1-0). Therefore, use of this technology as a manufacturing process along with conventional manufacturing techniques can significantly improve and boost the manufacturing industry.

Fused deposition modeling (FDM), a leading RP technique, accomplishes the layer-by-layer build by depositing a material extruded through a nozzle in a raster pattern (i.e., in a pattern that is composed of parallel lines) in each layer  $[1,2,4,5]$ . However, because only a limited number of materials, such as thermoplastics and some engineering plastics, have been used as a feedstock for FDM, the final products have limited mechanical properties [\[6,7\].](#page--1-0) Therefore, to render this technology suitable for producing functional, load-bearing parts, FDM protocols are needed for materials development and for the manufacturing of composite products.

Fiber reinforcement can significantly enhance the properties of resins/polymeric matrix materials [\[8–11\].](#page--1-0) Although continuous fiber composites offer high mechanical performance, their processing is not commonplace. More commonly used for traditional low-cost composite part fabrication are the short fiber-reinforced polymers (SFRPs) with moderately improved mechanical properties [\[3,12–14\]](#page--1-0). SFRPs are typically produced by extrusion compounding and injection molding processes [\[15–20\]](#page--1-0). The mechanical properties of these SFRPs depend significantly on the fiber length distribution and fiber orientation distribution of the final parts [\[3,14,21\].](#page--1-0) During processing, fiber breakage occurs [\[3\]](#page--1-0), affecting the mechanical properties of the final composite part. As fiber loading increases, the fiber breakage, due to increased fiber–fiber interaction [\[15,22,23\],](#page--1-0) increases. Fiber breakage during processing also arises from the interaction of fibers with polymers, and processing equipment surfaces  $[3]$ . Therefore, the matrix material, the process conditions, and the fiber loading determine the final fiber length

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distribution of the composite. Similarly, fiber orientation distribution and void fraction of final SFRPs are also affected by the aforementioned factors.

Only a few studies report FDM of fiber-reinforced feedstock. Among these, Gray IV et al. [\[4\]](#page--1-0) added thermotropic liquid crystalline polymer fibrils into polypropylene in order to prepare a composite feedstock for FDM. A capillary rheometer was used to simulate the FDM process, and, subsequently, the tensile properties of the extruded strands were measured. Zhong et al. [\[2\]](#page--1-0) studied FDM processing of short glass fiber-reinforced acrylonitrile–butadiene–styrene (ABS) resin. Additions of plasticizer and compatibilizer improved feedstock processibility. Shofner et al. [\[6\]](#page--1-0) investigated the effect of vapor-grown carbon fibers into ABS as an FDM feedstock. An average of 39% increase in tensile strength was observed at 10 wt% loading of nanofiber. To the best of our knowledge, FDM processing of  $5-7 \mu m$  diameter short carbon fiber-reinforced resin has not been reported, despite its high potential to reach desired mechanical, electrical and thermal properties, and low density [\[24–26\].](#page--1-0) Thermoplastic matrix composites further provide improved toughness and recyclability [\[25,27\].](#page--1-0)

In this study, carbon fiber-ABS composites were successfully prepared and used as FDM feedstocks. Short carbon fiber-reinforced ABS composites at different fiber loadings were prepared by both compression molding (CM) and FDM to assess the strengths and weaknesses of the FDM process (in comparison with the more conventional CM process). Effects of the process and fiber loading on void formation, average fiber length, and fiber orientation distribution, and eventually their effect on the tensile strength and modulus of the final printed product, were investigated.

#### 2. Experimental

#### 2.1. Materials and processing

ABS copolymer (GP35-ABS-NT) was obtained from M Holland Co., IL. Chopped Hexcel AS4 carbon fibers (CF) with epoxy-based sizing of 3.2 mm length were obtained from E&L Enterprises Inc., TN.

The carbon fibers and ABS resin were compounded with a Brabender Intelli-Torque Plasti-Corder prep-mixer at 220 °C and 60 rpm rotor speed until the torque reading became constant. Mixtures of 10, 20, 30, and 40 wt% CF were prepared. A neat ABS resin was also run through the mixer at the same conditions as the control. Average mixing time was 13 min, including the feeding time. Next, these mixes were extruded as preforms at  $220^{\circ}$ C using a plunger type batch extrusion unit. For CM preforms, a slit-shaped die and for FDM printing preforms (i.e., filament), a cylindrical die of 1.75 mm diameter were used. During the process, the barrel temperature ranged between 220 and 235  $°C$ .

FDM dog-bones were prepared by feeding the extruded filaments into a commercial desktop FDM unit (Solidoodle 3 from Solidoodle Co., NY) and printing. ASTM D638 type-V dog-bone dimensions were followed [\[28\]](#page--1-0). During printing, nozzle temperature was maintained at 205  $°C$ , while printer table temperature was 85  $\degree$ C. The layer height was set to 0.2 mm with the deposition direction being parallel to the loading direction in the gage section. The nozzle diameter of the FDM unit was 0.5 mm and the radius of curvature at the corners of the dog-bones was around 1.2 mm. The printed dog-bones were precise and no post-processing machining was required/performed. Although all samples up to 30 wt% CF were printed successfully, only several layers of the 40 wt% CF samples could be printed owing to nozzle clogging. Thus, the reader should note that the results for this sample were only included for completeness.

For the preparation of the CM dog-bones, slit-extruded preforms were cut into shorter pieces to fit the mold, and they were compression molded at  $220 \degree C$  based on ASTM standard D4703 [\[29\]](#page--1-0) to make rectangular bars. Next, dog-bones (ASTM D638 type-V) were cut from these bars by use of a Tensilkut template (special template for ASTM D638 Type V, Sieburg International Inc., TN), and a router (Tensilkut 10-21, serial No. 100590, Sieburg International Inc., TN).

### 2.2. Testing and analysis

The tensile properties of the CM and FDM samples were determined by testing at least five dog-bone samples of each composition, performing displacement-controlled tensile tests in a servo-hydraulic testing machine at a strain rate of 0.0254 mm/s. A 12.5 mm gage-length extensometer was used for strain measurements.

Fibers were extracted from dog-bone samples using acetone. A small portion of each extracted sample was transferred onto a glass petri dish, and the acetone was allowed to evaporate. Images of the extracted fibers were taken at  $20\times$  magnification, and fiber length distributions from these images were obtained using a code developed in our laboratories. Mostly, around 1000 fibers were measured in order to obtain reliable fiber length data.

A piece from a dog-bone representing each composition was cut and mounted in epoxy. Next, these samples were polished for imaging clarity. After taking images of the polished surfaces for void fraction analysis, the surface was plasma etched to reveal the fiber orientation for imaging. Afterwards, a technique developed by Velez-Garcia and automated by Kunc [\[30\]](#page--1-0) was used to calculate fiber orientation. The images were taken from the regions of the samples that were most representative of the gauge region of the dog-bones.

Fracture surfaces of the tested dog-bones were first sputtercoated with carbon. Next, SEM micrographs of the fracture surfaces were taken with a Hitachi S4800 FEG-SEM at an acceleration voltage of 5 kV and an emission current of 20  $\mu$ A.

#### 3. Results and discussion

The purpose of this research was to understand challenges and opportunities of fiber-reinforced composites made by 3D printing and to specifically evaluate the potential for load-bearing components. Our results show that composites with highly dispersed and highly oriented carbon fibers can be printed by FDM process as illustrated in Fig. 1. Both tensile strength and modulus increased



Fig. 1. Schematic presentation of 3D-printed fiber-reinforced composite by fused deposition modeling.

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