Ultrasound-assisted Vat Photopolymerization 3D Printing of Preferentially Organized Carbon Fiber Reinforced Polymer Composites

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Abstract

In this study, we present a new vat photopolymerization 3D printing process that uses acoustic radiation forces from ultrasonic standing waves to organize carbon short fibers within a photocurable resin. A chamber was developed to generate the standing bulk acoustic wave in the resin to align the carbon fibers along the nodes of the standing wave. The resin was then selectively cured to create constructs in the shape of a dog bone specimen by exposing to UV. The effect of fiber concentration (0.5%, 1%, 2%, and 4% w/v) and direction of alignment (parallel, perpendicular) on tensile strength of the carbon fiber reinforced polymer composites was determined. The constructs with 1% w/v showed the highest gain in tensile strength due to the fiber alignment. For two-layered constructs with 1% fiber concentration, 0°-0° constructs (fibers aligned along the uniaxial testing direction) demonstrated significantly higher tensile strength followed by 0°-90°constructs compared to constructs with randomly distributed fibers and without fibers.

Keywords: Additive manufacturing; Vat photopolymerization; Ultrasound; Bulk acoustic wave; Carbon fiber reinforced composites

Nomenclature

AM	Additive Manufacturing
CAD	Computer Aided Design
CFRP	Carbon fiber reinforced polymer
SLA	Stereolithography
UV	Ultraviolet

1. Introduction

AM is a manufacturing technology that is capable of creating three-dimensional parts layer-by-layer using data from CAD models [1]. AM processes are capable of producing parts with high geometric complexity by adding material layer by layer as opposed to subtractive manufacturing processes which require removal of material to create a desired shape [2][3]. AM provides users the ability to customize their design and build parts based on their requirements [4]. Over two decades, several AM processes have been developed and are being applied in automotive, aerospace, fashion, biomedical, and consumer products [5].

Vat photopolymerization is the one of the most commonly used AM techniques which involves curing of photopolymer resin using UV radiation [6]. In this process, the UV laser is used to create a pre-programmed geometry on to the surface of photopolymer resin filled in a tank. The resin, upon irradiation, gets solidified and forms a single layer of the part. Then, the build platform is lowered by one layer and this process is continued for each layer until the complete part is built [7]. The main limitation associated with vat photopolymerization process is the poor mechanical properties of manufactured parts [8]. The process of crosslinking induces brittleness in the parts which limits their application in high load applications [9]. Moreover, the polymer resin is light sensitive and undergoes warpage over time, needing significant post-processing to retain the part geometry. Herein, reinforcing the resin by adding short fibers could create composite parts with improved mechanical properties.

Some studies have been conducted to reinforce the resin in vat photopolymerization process with various reinforcement constituents including graphene oxide, cellulose nanocrystals, nanosilica and carbon nanotubes [10][11][12][13][14][15][16][17]. These studies have shown promising results, and considerable improvement in mechanical properties has been achieved. However, these approaches typically involve random dispersion of reinforcement constituents into the resin matrix. Not only the concentration of the reinforcements but also their organization within the resin matrix can be expected to affect the mechanical properties. The control over alignment in resin matrix can provide better directional mechanical properties to the parts for specific load bearing applications.

The alignment of particles in matrix media using ultrasonic assembly has been an area of interest in the recent past. An acoustic radiation force field can cause the arbitrarily shaped particles to be drawn towards the pressure nodes [18]. Ultrasonic waves of a pre-defined frequency can be transmitted using piezoelectric transducers to create a standing wave in resin media. The standing wave creates nodal planes where the particles in the media tend to aggregate. The nodes are planes parallel to the piezoelectric transducer-reflector surfaces and are separated by half the ultrasound wavelength. This technique allows control of both the alignment and position of the particles in media. Using such a technique, some researchers have been able to produce composites by manipulating and aligning the reinforcement particles into various photocurable or chemically curable matrix media including polysiloxane, acrylics, agar, epoxy and polyester [19][20][21][22][23][24][25][26][27]. However, the major focus of these studies was to produce the alignment of the reinforcements, and there is insufficient information about the mechanical properties and structural performance of ultrasonically-formed composites. In addition, layer-by-layer 3D printing and control of fiber orientation along the thickness of the specimen were also not the foci of those studies.

This study focuses on a new vat photopolymerization 3D printing process that uses acoustic radiation forces from ultrasonic standing waves [28] to organize carbon short fibers within a UV-curable resin. We demonstrate process-structure interrelationships that govern the alignment characteristics of the fibers within the resin, and then study the effect of fiber concentration and direction of alignment on tensile strength of the CFRP composites.

2. Materials and methods

2.1. Specimen fabrication

A chamber consisting of a piezoelectric transducer (2 MHz, STEMINC) opposite to a glass reflector was used to generate the standing bulk acoustic wave in the polymer matrix (Clear Resin, Formlabs) to organize the carbon fibers (average Ø7.2 x 100 µm, Zoltek Corp.) along planes parallel to the transducer surface. The schematic diagram of the chamber is shown in Fig. 1.

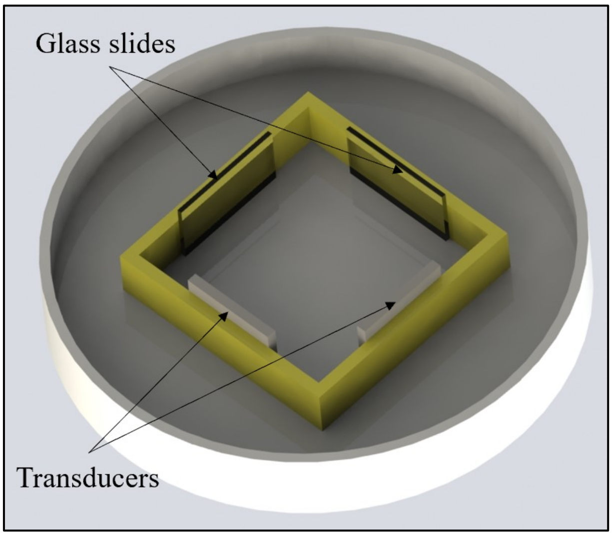

Fig. 1. Chamber used to generate the standing bulk acoustic wave in the polymer matrix

The resin, diluted with acetone in 40:60 ratio to lower the viscosity to reduce the viscous drag forces on the fibers during alignment, and mixed with the fibers was added to the chamber. The piezoelectric transducer was actuated by applying a sinusoidal voltage signal produced from a function generator (Keysight Technologies Inc.) and amplified from an RF amplifier (Electronics & Innovation Ltd.) to align the randomly dispersed carbon fibers along pressure nodes. A 1 min wait period was used to allow the fibers to get aligned before the resin was selectively cured in the shape of a dog bone specimen with entrapped fibers by placing a mask (geometry as per ASTM D1708-18) over the chamber and exposing to 405 nm UV light at 10 mW/cm^2 for 10 min. After one layer was cured, resin was added on top and the other piezoelectric transducer was actuated to create alignment in the second layer. Fig. 2 shows a schematic of the entire process workflow.

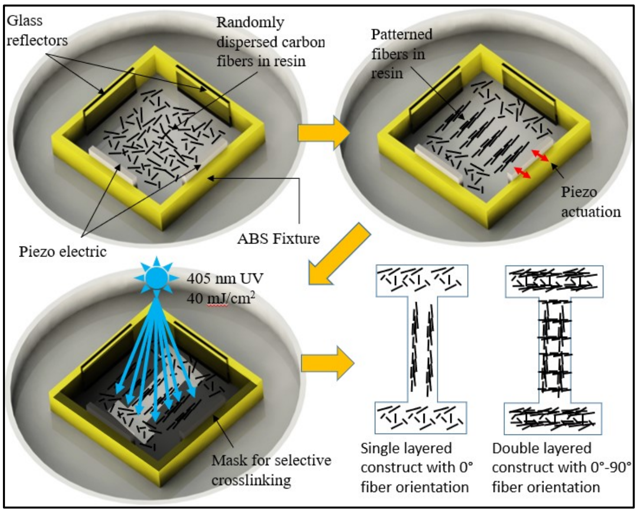

Fig. 2. Illustration of alignment of fibers using ultrasonic actuation and curing of resin in the shape of dog bone specimen using a UV light source.

2.2. Mechanical testing

To evaluate the effect of fiber concentration and alignment on tensile strength, single-layered constructs were fabricated with or without aligned fibers at four different concentrations (0.5%, 1%, 2%, and 4% w/v) and tensile tested (n = 3 constructs/group) as per ASTM D1708-18 standard in a universal testing system. Then, for the fiber concentration that showed the largest gain in strength due to fiber alignment, tensile strength of two-layered constructs with 0°-0° (fibers aligned along the testing direction in both layers) and 0°-90° was assessed in comparison to constructs of the virgin resin and resin with randomly distributed fibers.

3. Results and discussion

Ultrasonic patterning was successfully applied to control the alignment of carbon fibers within individual layers in polymer resin. The fibers aligned along planes parallel to the transducer at their nearest pressure nodes and the separation between the neighboring bands of fibers was closely correlated to half the wavelength of ultrasound across all concentrations. Fig. 3 depicts a 3D printed construct and representative microscopic images of aligned fibers in both single and two-layered constructs.

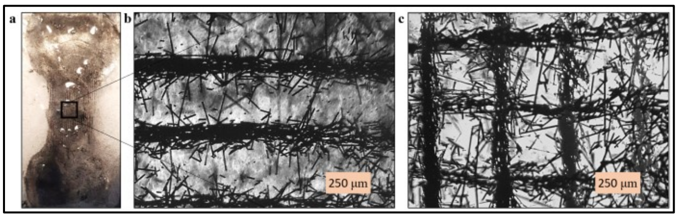

Fig. 3. (a) 3D-printed construct; (b) alignment of fibers in single layer; (c) alignment of fibers across two layers in the 0.5% fiber concentration group

Distinct alignment could be observed at 0.5% and 1% fiber concentrations. However, at 2% and 4% concentrations, the large quantity of fibers led to large widths of adjoining strands of clustered fibers, which inevitably obscured the overall alignment pattern. In such scenarios, to create distinct patterns, higher radiation forces generated by increasing the voltage applied to the transducers, could be utilized to enhance the compactness of the fibers in individual strands, which will be in the scope of future studies. Fig. 4 shows the mechanical testing setup to evaluate the tensile properties of the constructs.

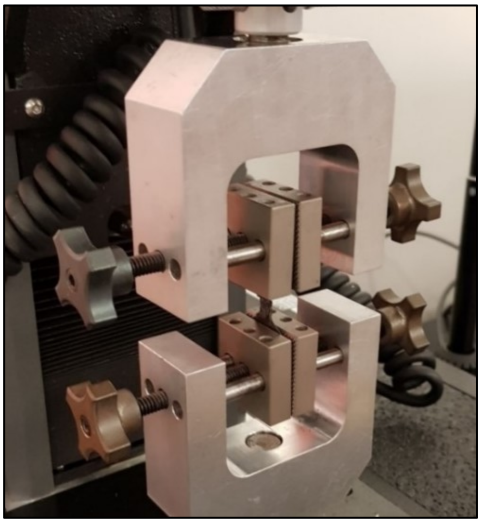
Fig. 4. Tensile testing setup

The results are summarized in Table 1. The tensile strength of single-layered constructs in the 0.5%, 1%, 2% and 4% fiber concentration groups with alignment were 2.1, 2.78, 1.71, and 3.57 MPa, respectively. In comparison, tensile strength of corresponding constructs without fiber alignment were 1.88, 2.17, 1.69, and 3.43 MPa. The constructs with 1% w/v fiber concentration showed the highest gain in tensile strength (28%) due to the alignment. This was expected since 1% was the highest concentration at which distinct alignment could be observed. These results were analyzed using two-way ANOVA with alignment and fiber concentration as independent factors. The fiber concentration was a significant factor. Tukey post hoc test showed that differences in tensile strength between 0.5% and 4% groups, and 2% and 4% groups were statistically significant.

Table 1. Effect of fiber concentration and alignment on tensile strength of 3D-printed constructs

Fiber concentration	Alignment NA: Not aligned A: Aligned	Tensile strength (MPa)	Percentage improvement in tensile strength
0.5%	NA	1.88 ± 0.46	11
	A	2.10 ± 1.13	
1%	NA	2.17 ± 0.40	28
	A	2.78 ± 0.77	
2%	NA	1.69 ± 0.39	1
	A	1.71 ± 0.39	
4%	NA	3.43 ± 0.8	4
	A	3.57 ± 1.3	

The 1% fiber concentration group was used to evaluate the effect of reinforcement and direction of alignment on tensile strength of the two-layered constructs since it showed the highest gain in tensile strength. The tensile strength of groups was ranked in following descending order: 0°-0° constructs, 0°-90° constructs, constructs with randomly distributed fibers, virgin resin constructs (Table 2). As such, the 0°-0° constructs were expected to possess higher tensile strength than the other three groups. This is because the fibers in both layers of the 0°-0° constructs were oriented along the direction of tensile loading, which increased the overall friction between the fibers during stretching, leading to higher strength in tension. A one-way ANOVA showed that the difference in tensile strength between the groups was statistically significant ($p < 0.05$). Tukey post hoc test showed that the differences in all groups other than 0°-90°constructs and constructs with randomly distributed fibers, were significant.

All three subgroups with reinforced constructs had higher tensile strength as compared to that of virgin resin group. This is in agreement with established literature [13][15] that reinforcement of resin with carbon fibers can significantly improve the tensile strength of polymers. In addition, we also show that controllably organizing carbon fibers within resins through our ultrasound-assisted VAT photopolymerization process also had a significant effect on the mechanical performance of the constructs. Herein parallel (0°-0°) fiber organization across the bi-layered constructs showed higher tensile strength than orthogonal (0°-90°) constructs and constructs with randomly dispersed fibers.

Table 2. Effect of reinforcement and direction of alignment on tensile strength of the constructs at 1% fiber concentration

Reinforcement type	Tensile Strength (MPa)
Virgin resin	1.63 ± 0.14
Randomly dispersed fibers	2.16 ± 0.08
0°-0° constructs	2.95 ± 0.12
0°-90°constructs	2.36 ± 0.14

In future studies, the effects of different frequencies and ultrasound amplitudes on structural and mechanical characteristics of CFRP composites can be studied. In theory, higher ultrasound amplitude can lead to higher radiation forces, resulting in constructs with more closely packed strands of clustered fibers. This could lead to improved mechanical strength. In addition to strength, the effect of fiber concentration, ultrasound parameters and resulting organization on maximum elongation should also be investigated. Finally, fiber length is another important parameter that needs to be studied. In theory, there is no limit to the fiber length that could be oriented. As such, an elongated fiber length could increase the inter-fiber friction and enhance the tensile strength. However, an increased fiber length would also require higher radiation forces, generated by increasing the voltage applied to the transducer, which would be accompanied by increased temperature of the transducers due to increased impedance heating. This would, in turn, limit the process capabilities in order to prevent damage to the transducer. These studies will be a part of our future work.

4. Conclusion

Ultrasonic patterning was effectively used to control the alignment of carbon fibers within individual layers in CFRP composites, with composites containing 1% carbon demonstrating the most benefit out of the organization. Both reinforcement and direction of alignment had significant effect on tensile strength of bilayered constructs at 1% fiber concentration. This approach has the ability to be scaled up into DLP and SLA processes to create functional mechanically anisotropic composites parts. In future, the effects of fiber length, ultrasound frequency and amplitude, and layer thickness on the mechanical properties and sustainability of such composites will be investigated.

Acknowledgements

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