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# Study on 3D printing process of continuous carbon fiber reinforced shape memory polymer composites

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**Abstract.** In this paper, shape memory polymer (SMP) and continuous carbon fibers (CCF) were utilized as raw materials to manufacture composites by means of 3D printing based on FDM. Various process parameters were systematically studied by the orthogonal experiment to find out the optimal process combination where interfaces and mechanical properties of printed composites specimens were treated as research objectives. The influence of each factor on the performance of the specimen was discussed. The results indicated that the mechanical properties were preferable when the printing temperature was 200°C, the printing speed was 4.5mm/s, the scanning pitch was 1.1 mm, and the ply stacking sequence was 0°. Furthermore, the fiber content had little effect on the shape memory performance of the polymer matrix. The rapid manufacture of shape memory carbon fiber composites has potential use in the field of aerospace.

## 1. Introduction

Shape Memory Polymer (SMP), as one of the new representatives in the field of smart materials, has the advantages of various driving methods, strong deformation ability, high recovery rate and being easy to process<sup>[1]</sup>. It has gradually become one of the hotspots in academic research<sup>[2]</sup>. Nevertheless, in order to solve the problem of low strength and stiffness of the shape memory polymer as structural components, carbon fibers are considered to be added as reinforcing material in practical applications<sup>[3]</sup>. What's more, preliminary studies show that the reinforcing effect of continuous fibers is better than that of short fibers<sup>[4]</sup>. Ning Fuda investigated that compared with pure plastic specimen, adding carbon fibers into plastic materials could increase tensile strength and Young's modulus, meanwhile, the longer the fiber was, the better the reinforcement effect would be<sup>[5]</sup>. Nevertheless, the traditional forming processes of carbon fiber reinforced composites are often very complex, and have a long production cycle and high cost<sup>[6]</sup>. Therefore, it is extremely urgent to develop a new fabrication process of continuous carbon fiber reinforced composites. At this time, 3D printing of composites, as an additive manufacturing technology, is a good choice to solve these problems.

In the present research, by using continuous carbon fiber as reinforcing phase and SMP as matrix, the CCF/SMP composites were fabricated by the modified FDM 3D printer with different process parameters. The mechanical properties of 3D printed specimens were systematically researched by



means of the orthogonal experimental to optimize the process parameters. And the interfaces between the fiber and resin were observed. The effect of fiber content on the shape recovery ratio of CCF/SMP composites was discussed as well.

## 2. Printer design

In this study, the fabrication of continuous carbon fiber reinforced SMP composites was achieved by use of the fused deposition modeling (FDM), which is one of the most commonly three-dimensional printing methods formed by layer stacking. The composite 3D printer was mainly composed of motion platform, printing nozzle, fiber feeding mechanism and control system. Compared with conventional printers, the nozzle of this printer can feed resin and fiber from two channels simultaneously and extrude them from the same nozzle, as shown in figure 1. In addition, the fiber feeding mechanism was designed to continuously feed the fiber at a suitable speed by adjusting the rotating speed of the stepping motor to avoid breakage of the fiber during printing.

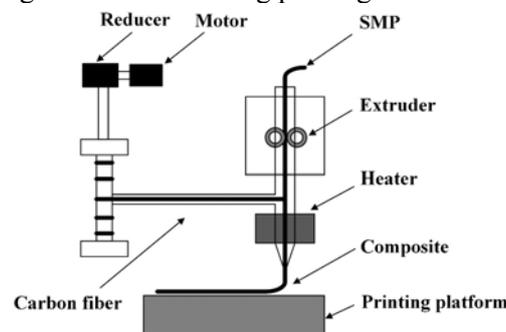


Figure 1. Printing principle of continuous carbon fiber reinforced SMP composites.

## 3. Study on 3D printing process of composites

### 3.1 Material

The shape memory polymer (SMP) developed by our research group was used as the matrix with glass transition temperature of  $76^{\circ}\text{C}$ , which was formed into a filament with a diameter of 1.75mm by the double-screw extruder. And the continuous carbon fiber (CCF, 1k HTA40, Toho Tenax Co., Ltd.) was used as the reinforcing phase to make the CCF/SMP composites.

### 3.2 Path design and 3D printing

Due to the continuity of carbon fibers, the original G code generated by the commercial 3D printing slicing software can not meet the requirements, where the printing path should be continuous. Therefore, four printing paths were designed by programming according to the different ply angles of carbon fibers, as shown in figure 2. The ply stacking sequence was  $0^{\circ}$ ,  $90^{\circ}$ ,  $0^{\circ}/90^{\circ}$  and  $\pm 45^{\circ}$ , respectively. The specimens were printed with the size of  $175 \times 15 \times 1$  mm and  $175 \times 25 \times 2$  mm according to ASTM 3039.

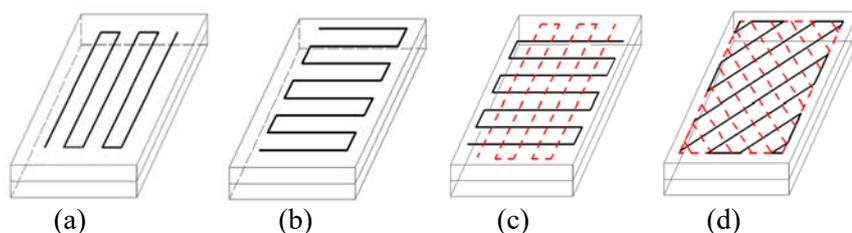


Figure 2. Four ply angles of carbon fibers, (a)  $0^{\circ}$  (b)  $90^{\circ}$  (c)  $0^{\circ}/90^{\circ}$  (d)  $\pm 45^{\circ}$ .

### 3.3 Orthogonal experimental design

Orthogonal experimental design is an effective method to study multi-factors and multi-levels, which is based on orthogonality to select some representative points from the all-parameter experiments. The influence factors in this research were printing temperature, printing speed, scanning pitch and ply angle, and each factor had four variables. The printing experiments of different combinations of process parameters were carried out according to the following table 1 to discuss the influence of different process parameters on the mechanical properties of the printed specimens. And the figure 3 shows partial printed specimens with different process parameters.

Table 1. Factors and levels of orthogonal experiment.

Printing Temperature	Printing Speed	Scanning Pitch	Ply Angle
185°C	1.5mm/s	1.1 mm	0°
200°C	3 mm/s	1.4 mm	90°
215°C	4.5 mm/s	1.7 mm	0°/90°
230°C	6 mm/s	2.0 mm	±45°

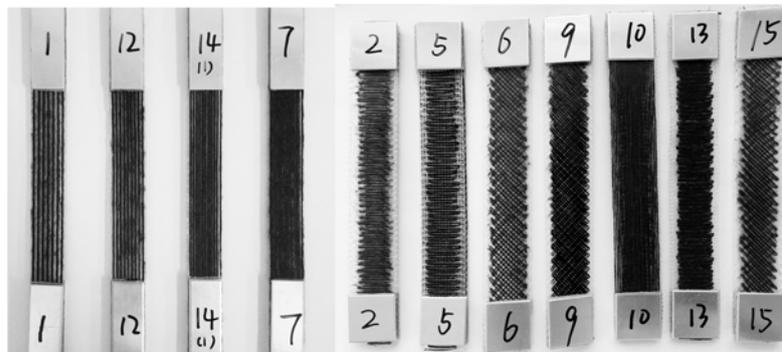


Figure 3. Partial printed specimens with different process parameters.

### 3.4 Mechanical test

To evaluate the performance of 3D printed CCF/SMP composites, tensile strength was measured using universal testing machine (Z050, Zwick Corp., Germany) according to the standard of ASTM 3039. There were five specimens in each experimental group, and the average value was measured as the final result. Furthermore, fracture surfaces of the tested specimens were observed by use of the scanning electron microscope (SEM) to evaluate the influence of various process parameters on the interface bonding between polymer and fibers.

### 3.5 Shape memory performance test

In order to research the effect of fiber content on the shape memory performance of the material, the shape memory performance test was carried out on the printed specimens with different fiber contents under the same printing parameters with the size of 60δ 60 δ 1 mm. The glass transition temperature of the SMP was about 76°C, which could be heated to deform in the water bath with the temperature of 80°C. The deformation and recovery results are demonstrated in figure 9.

## 4. Results and discussion

On the basis of the results of the orthogonal experiment, the range analysis was performed on each test index, and the results are shown in table 2.

Table 2. The results of range analysis.

Levels	Printing Temperature(A)	Printing Speed (B)	Scanning Pitch (C)	Ply Angle (D)
1	41.41	33.39	51.57	92.89
2	48.10	45.82	44.25	19.35
3	40.22	50.98	36.20	11.84
4	40.28	39.81	38.00	45.93
Range	7.89	17.59	15.37	81.04
Rank	4	2	3	1

It can be seen that the ply angle of fibers had the greatest influence on the tensile strength of the specimens, which was the main factor. The influence of each factor on the performance of the specimens followed the sequence: D>B>C>A, and the optimal process parameter combination was D1B3C1A2.

In order to further analyze the influence of process parameters on the mechanical strength of the specimens, the peak signal-to-noise ratio (SNR) was adopted based on the test results. The maximum tensile strength was taken as the research objective to maximize the response. As can be seen from the figure 4, when the printing temperature was 200°C, the printing speed was 4.5mm/s, the scanning pitch was 1.1 mm, and the ply stacking sequence was 0°, the tensile strength of the specimen was the largest, which was consistent with the results of range analysis.

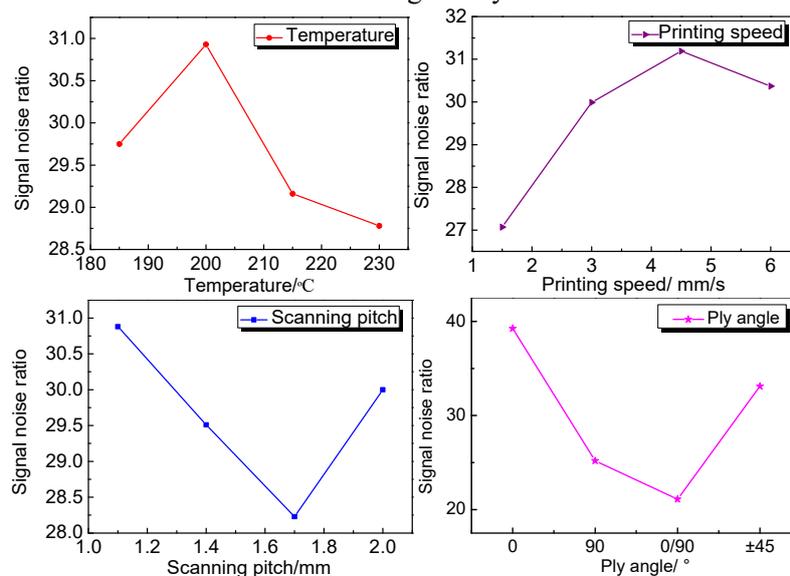


Figure 4. Influence of process parameters on mechanical properties of specimens.

The test results showed that the ply angle of carbon fibers was one of the key factors affecting the mechanical properties of the composites. Figure 5 shows the effect of changing the ply angle of carbon fibers on the tensile strength and modulus of the specimens under the same conditions of other process parameters. The reason for this result was that when the ply angle was 0 degrees, the tension direction was along the fiber direction, and due to the large specific strength and modulus of carbon fibers, the tensile strength of the specimens with this ply angle was the largest. While the ply stacking sequence was 90 degrees, the direction of tension was perpendicular to the fiber direction, which led to limited fiber reinforcement. In consequence, the tension could easily break the adhesion between resins.

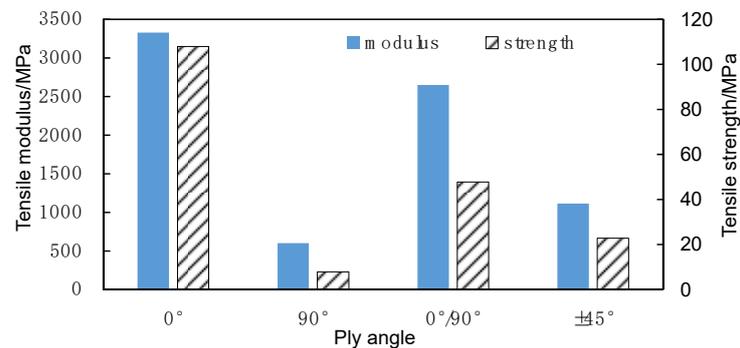


Figure 5. Relationship between tensile strength and modulus and fiber ply angle.

The scanning pitch refers to the center distance between two adjacent print lines, which controls the fiber content. The smaller the scanning pitch is, the more the fiber content is. The tensile strength and modulus increased with the increase of fiber content, as shown in figure 6. When the scanning pitch was 1.1 mm, that is, the fiber volume fraction was 7.26%, the tensile strength and modulus of the specimens were 118.05MPa and 4.4GPa respectively.

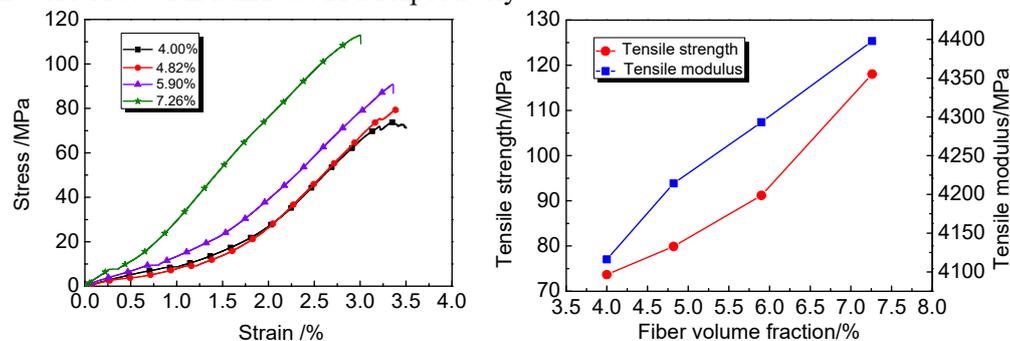


Figure 6. Stress-strain and strength/modulus of different fiber volume fractions.

The printing temperature had an important influence on the impregnation of the fibers and resin, and also affected the bonding strength between the layers. It was found that when the temperature was lower than 180°C, the polymer had poor fluidity and it was difficult to extrude from the nozzle. When the temperature was higher than 235°C, the polymer almost melted into liquid and naturally flowed out of the nozzle, which was not suitable for 3D printing. Therefore, the printing temperature was controlled between 185°C and 230°C in this study. The experimental results showed that the mechanical properties of the specimens were relatively better when the printing temperature was controlled at about 200°C. At the same time, the printing speed also affected the impregnation effect of the fibers and resin. Excessive printing speed prevented fibers from being fully permeated by the resin and caused the deviation of fibers from their original position by traction, resulting in printing failure or poor printing quality.

In order to further investigate the influence of printing temperature and printing speed on the interface bonding force, the failure surfaces of the specimens were observed under a scanning electron microscope. As can be seen from the figure 7(a), when the printing temperature was 185°C, obvious delamination occurred between the printing layers because the polymer was not fully melted and bonded to each other. When the printing temperature was 200°C or higher, as shown in figure 7(b) and (c), the fluidity of the polymer was better and the delamination disappeared.

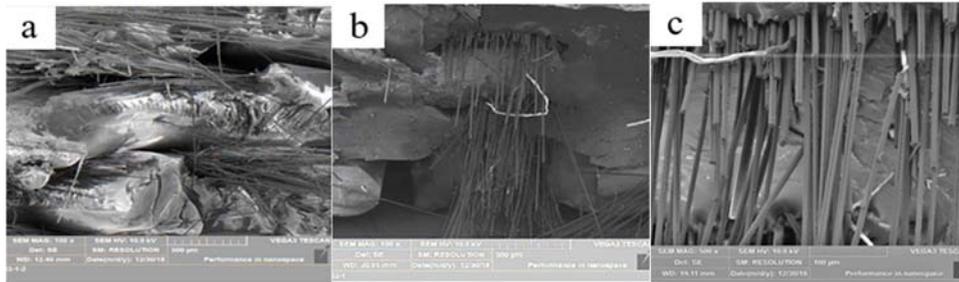


Figure 7. SEM of fracture cross section of specimens under different temperatures.

Figure 8 shows the scanning electron microscopy of the specimens at different printing speeds. It was found that when the printing speed reached 6mm/s, the fibers were not fully covered by resin and almost exposed, as shown in figure 8 (b), and the fibers were pulled away from their original position when the resin was not completely cooled with the overquick printing speed, as shown in figure 8 (c). And the fibers were well coated when the speed was 4.5mm/s, as shown in figure 8 (a).

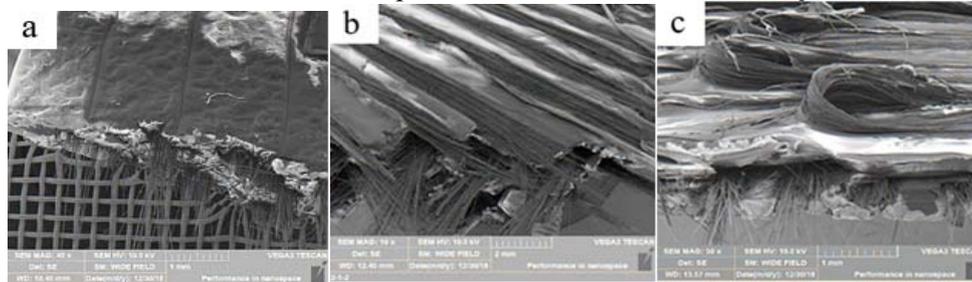


Figure 8. SEM of the surfaces of the specimens at different printing speeds.

As for the shape memory performance test, the printed specimens with different fiber contents were shaped in the water bath with the temperature of 80°C, and then cooled down. They almost all returned to their initial state when they were heated again to the glass transition temperature regardless of the fiber content, as shown in figure 9. This phenomenon indicated that the fiber content had little effect on the shape memory performance of the matrix.

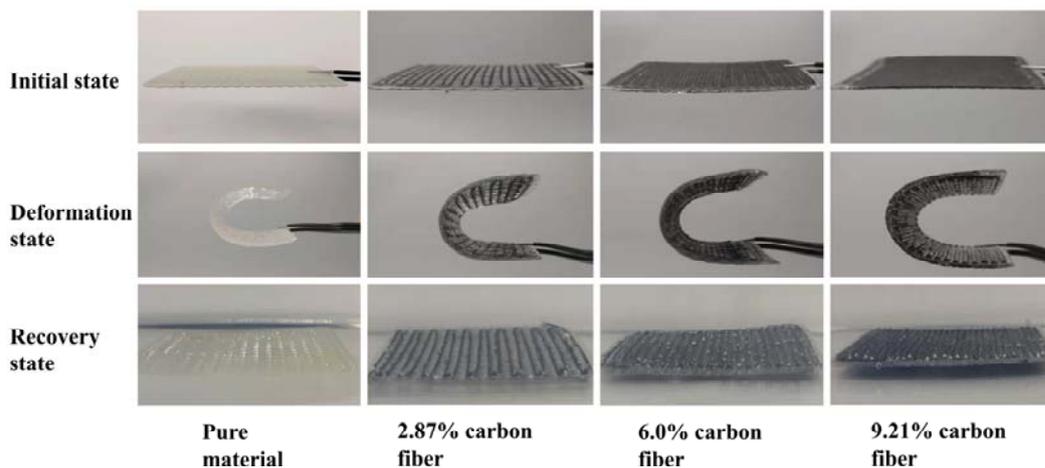


Figure 9. Shape memory performance test with different fiber contents.

### 5. Conclusion

Additive manufacturing technology (3D printing) based on FDM was utilized to fabricate the composite specimens in this paper, where the continuous carbon fiber and the shape memory polymer acted as the reinforcing phase and the matrix respectively. From the orthogonal test results, it was found that the specimens had relatively large tensile strength and modulus when the printing

temperature was 200°C, the printing speed was 4.5mm/s, the scanning pitch was 1.1 mm, and the ply stacking sequence was 0°. In addition, the tensile strength and modulus increased with the increase of fiber content. And when the fiber volume fraction reached 7.26%, the tensile strength and modulus of the specimens were 118.05MPa and 4.4GPa respectively, which was about three times larger than that of pure polymer. Printing temperature and printing speed were critical parameters to the interface bonding and impregnation of the fibers and resin, which also determined the mechanical performance of composites. According to the SEM and the tensile test results, good impregnation effect could be guaranteed with the printing temperature above 200°C and the printing speed below 6 mm/s. As for the shape memory effect, the fiber content had little effect on the shape memory performance of the polymer matrix.

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