Parameter Optimization for Preparing Carbon Fiber/Epoxy Composites by Selective Laser Sintering

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Abstract

Carbon fiber (CF) reinforced thermosetting resin composites offer a wide range of high performance features including excellent strength, modulus and thermal resistance and light weight. Consequently, they are increasingly demanded by aerospace and automotive industries due to the tighter requirements of the transport vehicles for lightweight as well as higher payloads. Although thermoplastics and their composites have been widely used in additive manufacturing (AM), to date it is difficult to manufacture carbon fibers reinforced thermosetting composite parts via AM technologies. Therefore, this study developed a novel method based on selective laser sintering (SLS) to fabricate high-performance carbon fiber/epoxy resin composites. The response surface method was employed to study the processing parameters affecting the quality of final parts, and an optimized processing condition was obtained.

1 Introduction

Carbon fiber (CF) reinforced thermosetting resin composites offer a wide range of high performance features including excellent strength, modulus and thermal resistance and light weight [1, 2]. Consequently, they are increasingly demanded by aerospace and automotive industries due to the tighter requirements of the transport vehicles for lightweight as well as higher payloads [3, 4]. There are many ways to make carbon fiber reinforced composites, including prepreg lay-up, spray-up, compression molding, injection molding, structural reaction injection molding (SRIM), etc [5]. Most of these techniques involve a molding process, which exhibits high production efficiency and good product accuracy, but unfortunately suffers from a long preparation period and high production cost [5, 6]. Additionally, they are also facing a great challenge in manufacturing parts with high complexity customized geometries. Although hand lay-up has flexibility to make single piece or small batches of complex composite parts, this process is labor intensive and not environment friendly [5, 7]. Currently, additive manufacturing (AM) technology has been introduced to make carbon fiber reinforced composites. In 2014, Local Motors Company reported the first 3D-printed car made from short carbon fiber reinforced ABS through fused deposition modeling (FDM) process [8]. In the same year, Mark Forged Company developed their commercial AM machine Mark One ™ to fabricate

continuous carbon fiber reinforced thermoplastic composites [9]. Some efforts have been made on preparation of carbon fiber/polyamide composites powder for SLS [10, 11] and now it has been commercially available [12-14]. Although thermoplastics and their composites have been widely used in additive manufacturing (AM), to date it is difficult to manufacture high-performance carbon fibers reinforced thermosetting composite parts via AM technologies.

In this work, a novel method based on the combination of selective laser sintering (SLS) and infiltration technique was developed to fabricate high-performance carbon fiber/epoxy resin composites. The polyamide (PA)-12 coated carbon fiber was firstly prepared through the dissolution-precipitation process, then green parts with proper porosity and mechanical strength was built by SLS, and the final parts was obtained by the infiltration of the green parts with epoxy resin (EP) and subsequent post solidification.

In the whole process, the SLS processing parameters have great influence on the porosity and initial strength of the green parts, which finally affect the mechanical properties of the final parts. The aim of this study was to optimize the SLS processing parameters when making the carbon fiber reinforced epoxy resin composites. The processing parameters including laser power, scan speed and layer thickness affecting on the flexural strength of the composite parts was analyzed and optimized by the design of experiments (DOE).

2 Materials and experiments

2.1 powder preparation

The polyamide 12 coated carbon fiber (PA12/CF) composite powders were prepared by the dissolution-precipitation method as described in our previous work [10]. The process is briefly as follows: (1) firstly, carbon fibers (P06^{TM} from Jilin Fangda Jiangcheng Carbon Fiber Co., Ltd., China., 10-100 μm long, 1.76 g/cm^3) and PA pellets (L1640 from Degussa Co., Germany, 1.01 g/cm^3) mixture (1:4 vol/vol) was add to ethanol solvent (1:10 wt/wt) in a 10 L reactor; (2) secondly, the mixture was heated to 145 ^{o}C under vigorous stirring conditions and kept at this temperature for 2-3 h until the PA12 pellets was thoroughly dissolved and a homogeneous suspension was obtained; (3) the carbon fiber powder acted as heterogeneous nucleus of the dissolved PA12 when the mixture was cooled to room temperature, and then the mixed solvent was distilled out; (4) after vacuum drying and ball milling the PA12/CF composite powders were finally obtained.

2.2 SLS manufacturing

The manufacturing of specimens was performed on the HK S320^{TM} SLS machine (Wuhan Huake 3D Technology Co. Ltd., China). The factorial multilevel design of the experiment was summarized in **Table 1.** The lower and higher values for the layer thickness, laser power and scan speed were defined based on the pre-stage test.

Table 1 Summary of statistical design

Parameters	Unit	low level	Medium level	high level
layer thickness	mm	0.09 (-1)	0.12 (0)	0.15 (+1)
laser power	W	3.6 (-1)	4.5 (0)	5.5 (+1)
scan speed	mm/s	1500 (-1)	2000 (0)	2500 (+1)

2.3 Infiltration process

The novolac epoxy resin F51^{TM} was supplied by Jiangsu Sanmu Group Corporation, China. Hardener methylnadic anhydride (MNA) and accelerator 2, 4, 6-tri-(dimethyl aminomethyl) phenol (DMP-30) were provided by Shanghai Chengyi high-tech Co. Ltd., China. To reduce the viscosity, the epoxy resin was kept at 140-150 ^{o}C for several minutes, and then the hardener and accelerator were added and mixed according to the prescribed weight percentage. For infiltration, the SLS green parts was immersed into the liquid resin and kept the top surface exposed to air. The whole process was conducted in a vacuum oven at the state of constant temperature and high negative pressure, which ensured that the porous green parts was thoroughly saturated. The specimens were taken out after 5 min, cleaned their surfaces, and then placed in an oven to post cure at 120 ^{o}C for 10h, 150 ^{o}C for 5h and 200 ^{o}C for 3h.

2.4 Flexural testing

Tensile (ASTM D638) and flexural (ASTM D790) properties was evaluated using a universal testing machine (Zwick/Roell Z010, Ulm, Germany).

3. Result and discussion

Experimental design and statistical analysis of the results were performed using the software Design Expert (version 8.0). The Box–Behnken design (BBD) was employed and a quadratic model was selected for the response based on the experimental plans.

Table 2 Design layout and experimental results for flexural strength of the final composite parts

Run	A:Layer thickness (mm)	B:Laser power (W)	C:Scan speed (mm/s)	Response: Flexural strength (MPa)
1	0.09	4.50	2500	135
2	0.09	4.5	1500	118
3	0.12	4.5	2000	123
4	0.12	4.5	2000	113
5	0.12	3.6	2500	146
6	0.12	4.5	2000	109
7	0.12	4.5	2000	103
8	0.09	5.4	2000	114

9	0.15	3.6	2000	141
10	0.15	4.5	2500	120
11	0.12	4.5	2000	99
12	0.15	4.5	2000	111
13	0.12	5.4	2500	132
14	0.12	5.4	1500	105
15	0.12	3.6	1500	119
16	0.15	5.4	2000	113
17	0.09	3.6	2000	138

3.1 Analysis of variance

The analysis of variance (ANOVA) revealed the most significant input factors, and their combination, in term of their influence on the results.

Table 3 Results for ANOVA applied to flexural strength

Term	Sum of squares	DF	Mean squares	F ratio	P value
A-Layer thickness	50	1	50	0.59	0.4661
B-Laser power	800	1	800	9.50	0.0177
C-Scan speed	800	1	800	9.50	0.0177
AB	4	1	4	0.048	0.8337
AC	16	1	16	0.19	0.6760
BC	0	1	0	0.000	1.0000
A^2	167.2	1	167.2	1.99	0.2017
B^2	491.12	1	491.12	5.83	0.0464
C^2	118.27	1	118.27	1.41	0.2745
Lack of fit	242	3	80.67	0.93	0.5041
Pure error	347.2	4	86.8		
Cor Total	3114.94	16			

ANOVA design layout and tested flexural strengths of the final composite parts are shown in **Table 2**. A quadratic model was applied to obtain the correlation with the experiment data. From **Table 2**, it is observed that P-values of laser power and scan speed and quadratic factor of laser power are 0.0177, 0.0177 and 0.0464 (less than 0.05), indicating a significant influence of these parameters on the flexural strength of the composite parts. The other factors in the model do not have significant effect on the response. Coefficients of determination, namely R^2, is defined as the ratio of the explained variation in the model to the total variation [15, 16], which indicates the degree of fit. When the R^2 close to 1, it means the that the selected model fit exactly with the actual results. The obtained R^2 value for the output is 0.81, showing that the quadratic equation model has a good correlation between the predicted and experimental values. The final regression euqstion of this model for response of flexural strength in terms of coded factors is shown as follows:

$$\text{Flexural strength} = 109.4 - 2.5 \times A - 10 \times B + 10 \times C - 1 \times A \times B - 2 \times A \times C + 6.3 \times A^2 + 10.8 \times B^2 + 5.3 \times C^2 \quad \text{..... Eq.1}$$

3.2 Normal probability

A normal probability plot can be used to analyze the distribution of the experimental points. **Fig.1a** and **b** show the normal plot of residuals and the plot of residual vs. predicted response, respectively. It can be seen that the residuals are fall on the straight line and the predicted responses are close to the baseline, which infer that the errors are distributed normally. Therefore, the proposed quadratic model is adequate.

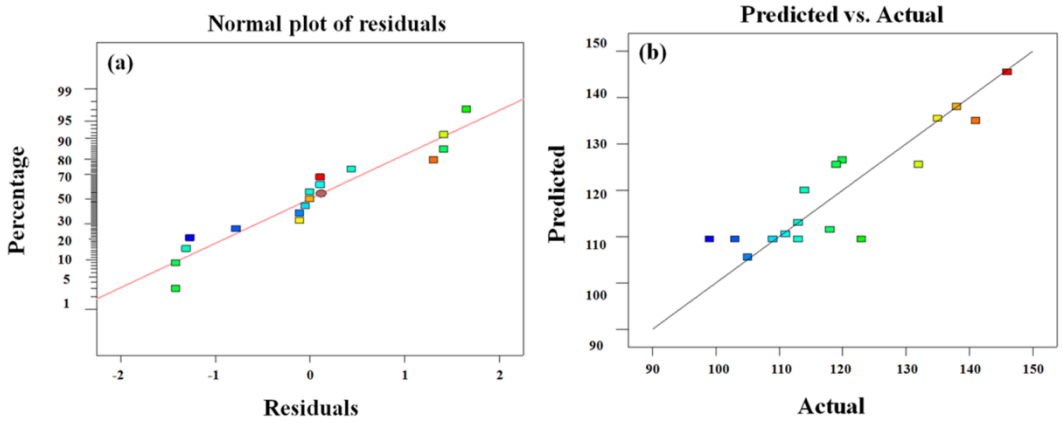

Fig. 1 Normal plot of rersidual (a) and residual vs. the predicted response (b) for flexural strength

3.3 Response surface

Figure 2 shows the effect of the single factor on the flexural strength, when keeping the other two factors at constant value. It can be seen that the flexural strength first decreased with the layer thickness at a lower level and then increased **(Fig.2a)**. When the layer thickness was at the lower level, the laser beam can easily penetrate into the thin powder layer and form a dense green part, leading to less liquid resin infiltration in to the porosity. when the layer thickness increses, less porosity would be exist in the parts, which hinders the later infliation process. As the layer thickness further incresed, the gap between two adjacent layers increases, which facilitates the resin infiltration into the gap, and thus enable the increase of the flexural strength. **Figure 2b** and **c** depict the influence of laser power and scan speed on the flexural strength of the final parts, respectively. It is shown that the flexural strength deceases with increasing the laser power, but increases with the scan speed. When the layer thickness was at a fixed value, increaing the laser power or decreasing the scan speed will result in higher energy input per unit area of the powder. Thus, denser green parts will be obtained and less thermosetting resin can be penetrated, leading to the lower flexural strength of final parts.

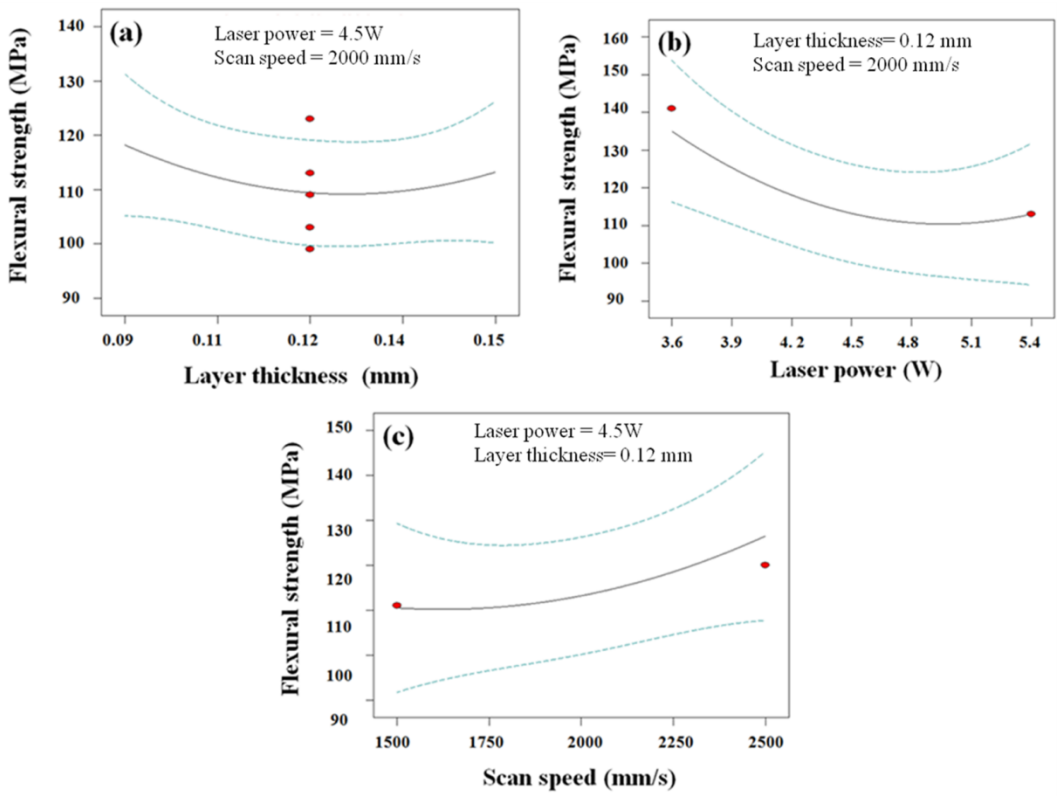

Figure 2 Effects of (a) layer thickness, (b) laser power and (c) scan speed on the flexural strength

Figure 3 shows the 3D response surface for the flexural strength. The curvilinear profiles show that the higher flexural strength is obtained at low levels of laser power, high level of scan speed, high levels and low or high levels of layer thickness, which is in agreement with the observation of the influence of the single factor in **Figure 2**.

The optimum processing parameters that yields miximum flexural strength are layer thickness of 0.15 mm, laser power of 3.61 W and scan speed of 2481 mm/s. At this optimized processing condition, the flexural strength of the composite parts is 147.2 MPa.

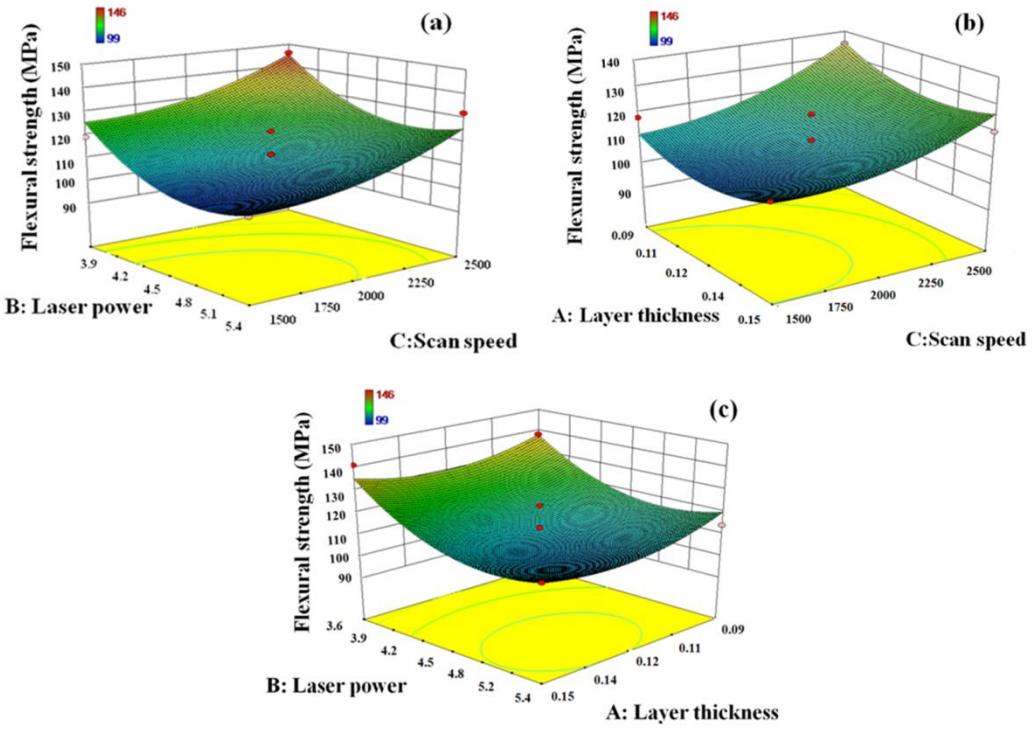

Figure 3 Response surfaces of flexural strength

Conclusion

This paper proposed a new method based on the combination of selective laser sintering and infiltration for manufacturing carbon fiber reinforced thermosetting resin. The response surface methodology and ANOVA were applied to analyze the effect of SLS parameters (layer thickness, laser power and scan speed) on the mechanical property (flexural strength), and obtained the optimal processing parameters for the desirable performance. The following conclusions were drawn from the present research:

(1) The Box–Behnken design was adopted for experimentation owing to benefit of reducing the number of the experiments required.

(2) A quadratic regression equation was established to describe the relationship between the regressor - responses using the response surface method. The results of ANOVA suggested that the predicted value of flexural strength well fits with the quadratic model.

(3) From ANOVA results and the response surface graph, it was concluded that laser power and scan speed are two major factors that influence the flexural properties. The flexural strength increases with the increase of scan speed, and the decease of the laser power.

(4) The maximum flexural strength of 147.2 MPa was obtained through the optimized processing parameters, and the optimized processing condition is: layer thickness of 0.15 mm, laser power of 3.61 W and scan speed of 2481 mm/s.

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