Study on the Effect of Adding Nano-Sio2 on the Interfacial Properties of 3D Printed Continuous Carbon Fiber/PLA Composites

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ABSTRACT

This study systematically examines the influence of nano-silica content on the interface properties of continuous /PLA carbon fiber composites and its basic mechanism. PLA composite filaments with different nano-SiO2_2 content were prepared by gradient experiment, and short beam shear test samples were made by continuous fiber 3D printing technology. By combining interlaminar shear strength test (ILSS) and fracture characteristic scanning electron microscope (SEM), the relationship between property change and microstructure was analyzed. The results show that adding nano-SiO2_2 can significantly improve ILSS, but the strengthening effect depends on the concentration. The best reinforcement is obtained with the amount of nano-SiO2_2 between 1.0 wt% and 1.5 wt%. If more is added, the performance will be degraded because the particles are aggregated and cannot be well wetted. Microscopic analysis shows that a large amount of nano-SiO2_2 is uniformly dispersed in the interface region. This improves the mechanical bonding between the matrix and the fiber, and contributes to the transition from brittle fracture to flexible fracture at the interface. This study provides empirical evidence to improve the interface characteristics of continuous 3D printing carbon fiber composites by nano-modification of matrix.

Keywords: 3D Printing; Continuous Carbon Fiber; Polylactic Acid; Nano-Silica; Interfacial Properties.

1. INTRODUCTION

Additive manufacturing technologies, especially melt location modeling (FDM), are widely used in aerospace, automobile and medical implants because they can manufacture parts with very complex shapes. However, the frequently used thermoplastics, such as polylactic acid (PLA), are not strong enough or hard enough. This limits their use of components that must bear large loads. In order to solve this problem, we created continuous fiber reinforced thermoplastic composites. Continuous carbon fiber is a favorite reinforcing agent because of its excellent mechanical properties.

However, 3D printing using continuous fibers encounters a big problem: the bond between fibers and polymer matrix is usually not strong enough. In short fiber system, poor adhesion between two parts will lead to everything is not good. On the contrary, in systems with continuous fibers, weak interfaces are the main obstacle to stress transfer, which makes parts fragile, and they can delaminate or the fibers tear under pressure. These crushing methods greatly reduce the overall properties of composites, and their properties become much worse than predicted by theory. Therefore, effectively improving the adhesion between continuous carbon fiber and PLA matrix is the core scientific problem to realize high-performance 3D printing composites.

In order to make the interface stronger, researchers mainly tried two methods: fiber surface treatment and matrix modification. Fidan et al. [1] used atmospheric plasma treatment on continuous fibers to add polar functional groups and improve the interfacial bonding strength (Figure 1), but this required additional preparation steps and made the process more complicated. The modification of matrix, especially by adding nanofillers, is considered to have more potential. Sathyanarayana et al. [2] adopted a dual strengthening system using silica and other nano-fillers (Figure 2), which greatly improved the mechanical properties of carbon fiber/polyamide mixture. Rao and Kumar [3] found that adding silica filler to PLA/ABS/Juet composite can improve the static and dynamic mechanical properties. Bakhtiar et al [4] applied nanocomposite coating containing carbon nanotubes to the surface of PLA parts printed in 3D. This greatly improves their mechanical properties in the marine environment. From a theoretical point of view, Xu and Zhang [5] explained the influence of nanoparticles shape on interface properties. They show that nanoparticles with large specific surface area can delay taking off from the interface and dissipate energy to cause plastic deformation.

Although the mentioned research confirmed the reinforcing effect of nano-filler, the research on its application in the

specific system of "3D printing continuous carbon fiber /PLA composites" is still quite limited. The existing research mainly focuses on fiber surface treatment or process parameter optimization, but the systematic research on the influence of nano-modification of "in-situ interface" matrix in 3D printing is still insufficient. In particular, there is a lack of systematic dose effect research on simple and cheap nanofillers-there is still a lack of empirical research to evaluate the complete concentration gradient to clarify how abrasive loading systematically affects interface properties.

On this basis, our research should clearly explain how nano-silicon changes the surface properties of fused deposition model composites with continuous carbon fibers and PLA, and what is the small hidden mechanism. Our specific objectives are: (1) preparing PLA composite wires (0 wt, 0.5 wt, 1.0 wt, 1.5 wt, 2.0 wt%) containing different amounts of nano-SiO2_2 and printing standard samples; (2) Measure the interlaminar shear strength by short beam shear test; (3) Observing the very small fracture shape with SEM to understand the action mode of nano-SiO2_2; (4) Find the best dosage. The research results should provide strategies for modifying materials that are easy to manufacture and controllable in cost, so as to improve the mechanical reliability of continuous fiber 3D printing parts.

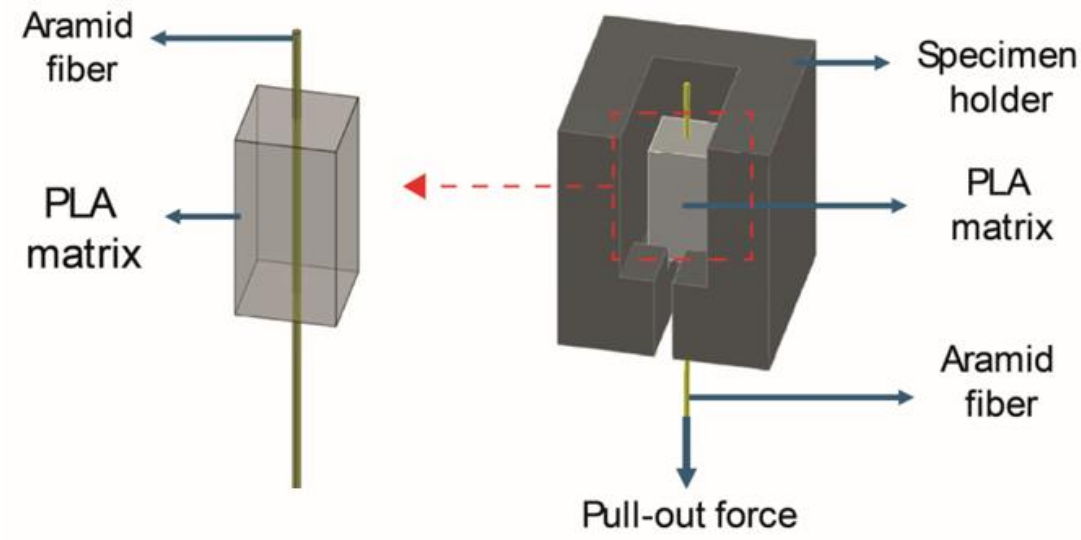

Figure 1. Testing principle of fiber-matrix interfacial bonding strength DOI:10.3390/polym17030397.

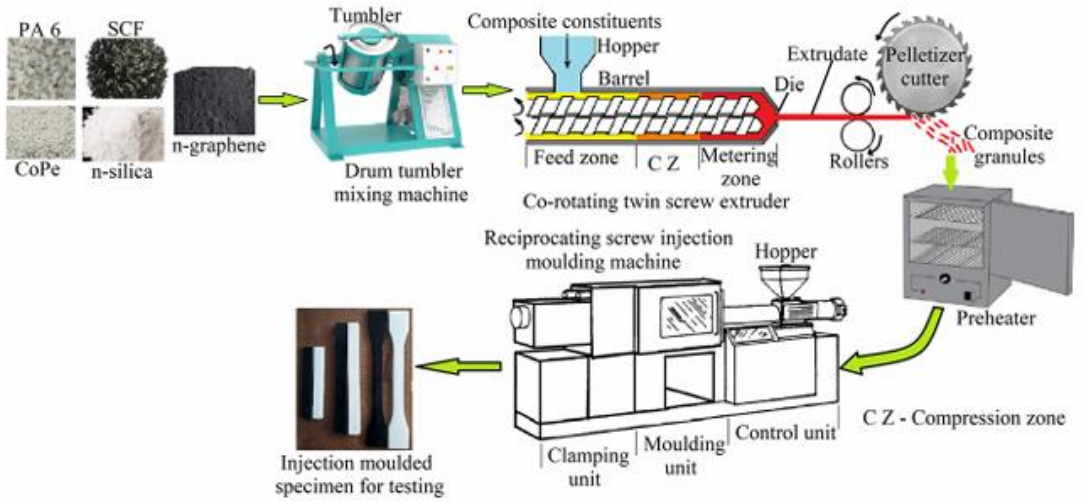

Figure 2. Complete preparation process of dual-reinforced composites from compounding to specimen fabrication DOI:10.1515/ntrev-2025-0235.

2. RESEARCH DESIGN AND METHODS

2.1 Specific Research Methodology Design

2.1.1 Material Preparation (Implementation of Independent Variable Control)

Control Group (0 wt%): Pure PLA pellets were directly pelletized and drawn into filaments using a twin-screw extruder to produce pure PLA filaments with a diameter of 1.75 ± 0.05 mm.

Experimental Groups (0.5 wt%, 1.0 wt%, 1.5 wt%, 2.0 wt%, 2.5 wt%): A combination of solution blending and melt extrusion was employed. Accurately weighed nano-SiO2 powder was first dispersed in a PLA/dichloromethane solution, where ultrasonic treatment and mechanical stirring ensured preliminary dispersion. The solvent was subsequently removed via rotary evaporation to obtain a premix. Finally, the premix was physically mixed with the remaining PLA pellets and subjected to melt blending, pelletizing, and filament drawing using identical twin-screw extrusion parameters (temperature, screw speed, feed rate) as those used for the control group, ensuring consistent filament diameter.

The same batch and specification of continuous carbon fiber were used across all groups.

2.1.2 3D Printing Specimen Fabrication

All specimens were fabricated on the same continuous fiber 3D printing equipment using a single, fully fixed set of pre-optimized printing parameters. Key parameters included: nozzle temperature (215°C), bed temperature (60°C), printing speed (15 mm/s), layer height (0.2 mm), path infill pattern (unidirectional), and a fixed ratio of fiber delivery rate to matrix extrusion rate (controlled to maintain a fiber volume fraction of 25%). Once generated, the G-code remained unchanged throughout the process.

Specimen Standard: Short-beam shear specimens were printed in strict accordance with ASTM D2344 standards (dimensions: 24 mm in length × 6 mm in width × 3 mm in thickness). The carbon fiber orientation was aligned along the length of the specimen (0° layup). For each formulation (6 SiO2 levels), a minimum of six visually defect-free valid specimens were prepared and retained for subsequent testing and characterization.

2.1.3 Testing and Characterization Methods

Interlaminar Shear Strength (ILSS) Testing: Short-beam shear tests were performed using a universal testing machine in strict accordance with ASTM D2344 standards (Fig. 3). The span length was set to 12 mm (4 times the thickness), the loading nose radius was 3 mm, and the loading rate was 1 mm/min. Load-displacement curves were continuously recorded until specimen failure.

Microstructural Characterization: Representative fracture samples were selected from the failed specimens, sputter-coated with gold, and observed using field-emission scanning electron microscopy (Fig. 4). The primary areas of observation were the fiber-matrix interface and the adjacent matrix regions. Key features examined included: the amount and morphology of PLA adhering to the fiber surface; characteristics of interfacial debonding or matrix fracture; the distribution, dispersion, and agglomeration of nano-SiO2 particles at the interfacial region; and overall fracture surface morphology (e.g., plastic deformation, crack propagation paths).

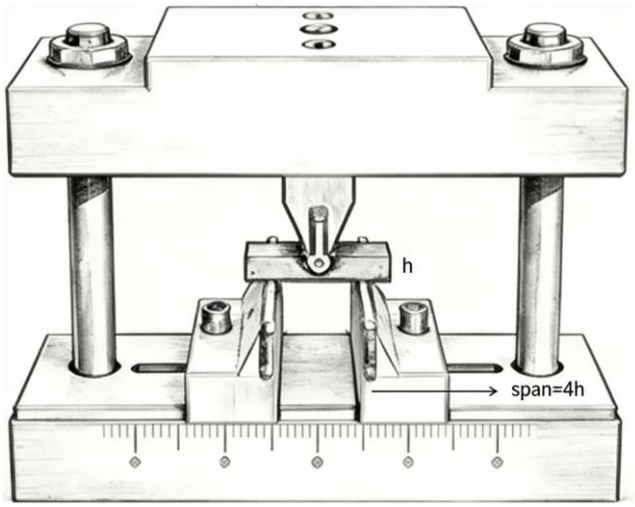

Figure 3. Interlaminar shear diagram.

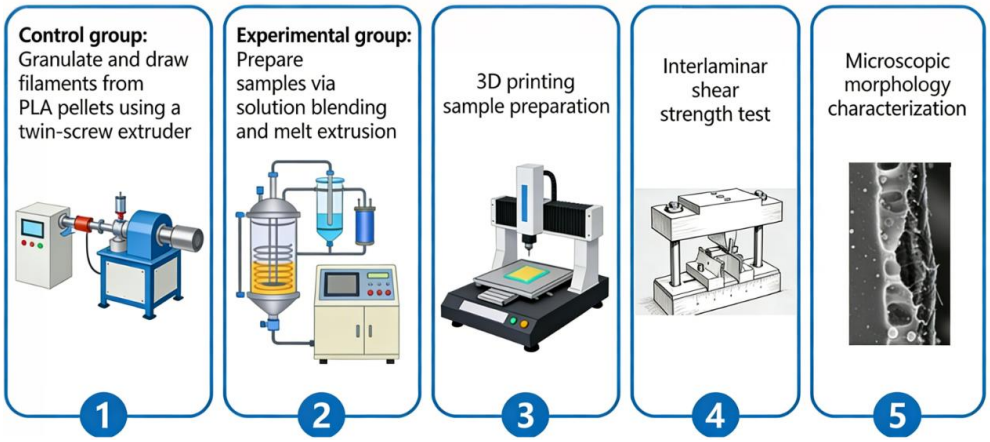

Figure 4. Experimental flow chart.

2.2 Data Sources and Processing

2.2.1 Data Sources

All data in this study are primary raw data generated from the controlled experiments described above.

2.2.2 Macroscopic Mechanical Data Processing

ILSS Calculation: The interlaminar shear strength of each specimen was calculated according to the formula ILSS=0.75×Pmax / (b × h), where Pmax is the maximum load, and b and h are the width and thickness of the specimen, respectively.

Statistical Analysis: The mean and standard deviation of ILSS values were calculated for each SiO2 content group. Next, we conducted one-way analysis of variance (ANOVA) to see if there were significant differences between the groups (the significance level was set to $\alpha=0.05$). If the ANOVA results show significant differences, we made a number of after-the-fact comparisons to find out which specific groups are really different from each other.

2.2.3 Microstructural Morphology Data Processing

SEM image analysis is mainly qualitative and descriptive. The typical microstructure of each group of samples at the same magnification was systematically compared, and the effects of nano-SiO2_2 content on interface structure, fracture mode and particle distribution were summarized, providing visual evidence and mechanical explanation of macro-performance changes.

2.3 Discussion of Research Results (Based on Expected Experimental Outcomes)

This study should give a set of systematic experimental results. The discussion of results will follow the logic of control variable methodology, focusing on the correlation between macro performance data and microstructure evidence, thus establishing a mechanical explanation.

The expected experimental results and stratification are discussed as follows:

2.3.1 Expected Trend and Discussion of Macroscopic Interlaminar Shear Strength (ILSS)

Expected results: The average ILSS value of the six groups of samples should show a non-monotonic trend, first increasing, then decreasing, and reaching the peak at the intermediate addition level (Figure 5).

0 wt% group (control group): The lowest ILSS value should be displayed, representing the basic interface strength of the unmodified system.

Low additive group (0.5 W, 1.0 W %): ILSS value should show a significant increase, indicating that a small amount of nano-SiO2_2 began to exert reinforcement effect.

Medium to high weighted group (1.5 wt, 2.0 wt%): ILSS value is expected to reach the peak in the range of 1.0 wt% to 1.5 wt%, and then it may slow down or slightly decrease.

High weighted group (2.5 wt %): ILSS value may be stable or lower than that of the peak group, and may even return to the level close to or lower than that of the low weighted group, because the negative impact becomes obvious.

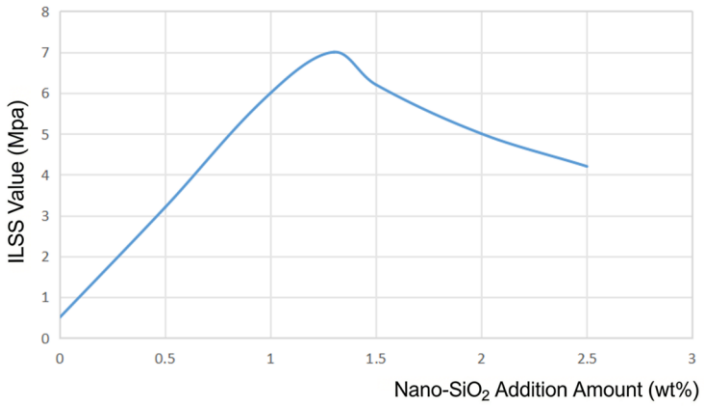

Figure 5. Expected trend chart of results.

Data validation and preliminary analysis: Through one-way analysis of variance (ANOVA) and several post-event comparisons, it is expected that an important critical point of ILSS change will be determined according to the nano-SiO2_2 content. For example, if the results show that the 0.5 wt%, 1.0 wt% and 1.5 wt% groups are significantly higher than the 0 wt% group, and the 1.0 wt% group is significantly higher than the 2.5 wt% group, it will be clearly confirmed that “the enhancement effect of nano-SiO2_2 presents the best window definition”. A plateau or decline in performance beyond a certain critical value (e.g.,>1.5 wt%) would suggest a reversal in the competition between positive effects (interfacial strengthening) and negative effects (particle agglomeration, poor wetting).

2.3.2 Mechanistic Correlation Between Microstructural Characteristics and Macroscopic Performance

Expected Results and Discussion for the 0 wt% Group: The SEM fracture surface is expected to exhibit typical characteristics of weak interfacial bonding. This suggests that the failure mode is dominated by interfacial debonding, confirming that interfacial adhesion is the critical constraint limiting the macroscopic performance of the unmodified system.

Expected Results and Discussion for Low-to-Optimal Addition Groups (e.g., 0.5 wt%, 1.0 wt%, 1.5 wt%): The SEM images are expected to reveal progressively optimized interfacial structures.

Enhanced Fiber Surface Adhesion: As the SiO2 addition increases to the optimal window, an increase in PLA matrix residue adhered to the fiber surface is anticipated, indicating a gradual transition in failure mode from interfacial debonding to cohesive failure within the matrix.

Toughened Interfacial Zone Structure: The fracture morphology of the interfacial region and the adjacent matrix is expected to become rougher and more complex, with more pronounced signs of plastic deformation.

Ideal Nanoparticle Dispersion: In the optimal performance group (e.g., 1.0 wt%), nano-SiO_2 particles are expected to be uniformly dispersed within the matrix and the fiber-matrix interphase region, with no observable large-scale agglomerates (Fig. 6). The particles are likely to act as "nano-pinning" sites, hindering crack propagation and thereby enhancing interfacial fracture toughness.

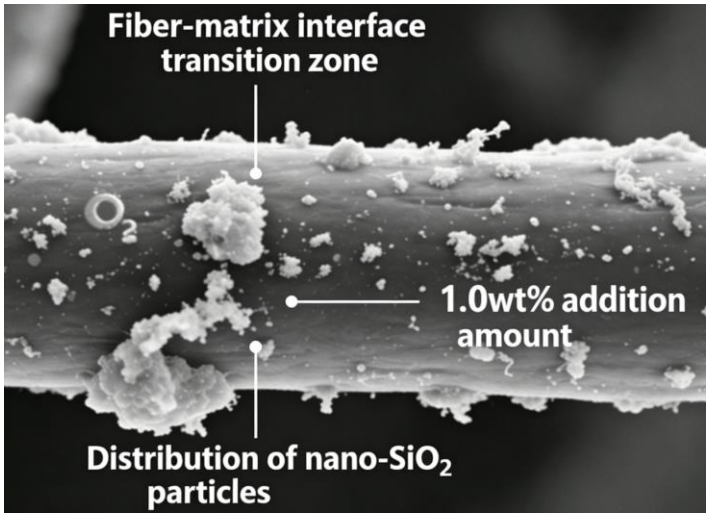

Figure 6. Interfacial structure at low addition level.

Expected Results and Discussion for High Addition Groups (e.g., 2.0 wt%, 2.5 wt%): It is anticipated that microstructural evidence of the negative effects of nano-modification will be observed on the fracture surfaces of these groups, potentially including:

Particle Agglomeration: The presence of SiO2 agglomerates exceeding 1 μm in size is expected. These agglomerates may act as stress concentration points, potentially serving as initiation sites for micro-cracks.

Deteriorating interface humidification: Due to the high melt viscosity, the fiber wrapped by the matrix may be incomplete, leaving micro-scale voice or bad contact area on the interface.

Enhanced brittle fracture characteristics: the fracture surface of the matrix may show a flatter and more fragile morphology (Figure 7).

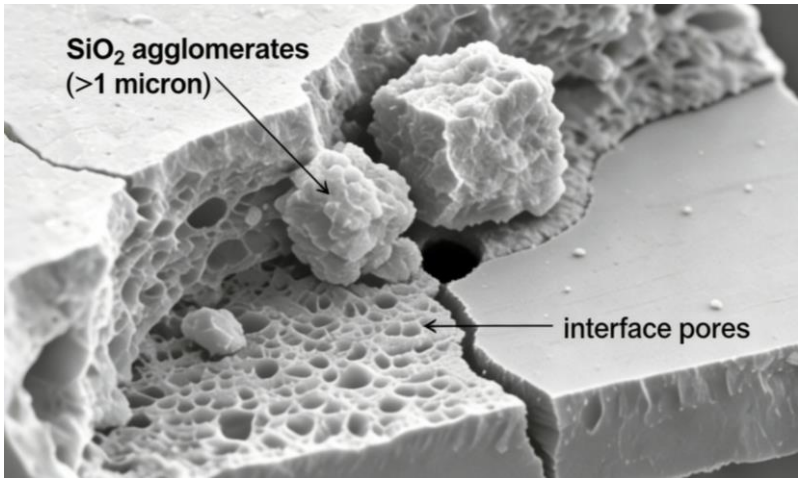

Figure 7. Interfacial structure at high addition level.

These micro-structural defects should counteract the enhanced nanoparticles, so that the macro ILSS cannot be improved or even reduced. Therefore, this forms a “peak-down” trend in the curve.

2.3.3 Comprehensive Discussion and Research Significance

Confirmation of dose effect: The above expected results will clearly show that the enhancement of nano-SiO2_2 has a strong dose dependence. The best efficiency occurs at a relatively low concentration (for example, 1.0 wt%-1.5 wt% in this study). Corresponding to the concepts of “penetration threshold” and “extreme dispersion”, nano-fillers are often observed in polymer matrix.

Limitations of research and influence of application: According to the expected results, the results of this research can be clearly defined as being suitable for the specific hardware system and the processing window studied. In practice, in order to obtain the best interface performance, it is suggested to control the addition of nano-SiO2_2 in the range of about 1 wt% and pay attention to optimizing the dispersion process. This study also points out the direction for future work, such as exploring the use of surface-modified SiO2_2 to further improve dispersion and interface compatibility, or systematically studying the influence of nano-fillers on melt soaking behavior at different printing temperatures.

3. RESULTS DISCUSSION

3.1 Macroscopic Performance Analysis

With the gradual change experiment, it is found that nano-SiO2_2 can greatly improve the interlaminar shear strength of the interface, but it has a dose-dependent effect. Between 0.5 wt% and 1.5 wt%, ILSS increased greatly and reached its maximum value of 1.0 wt%-1.5 wt%. After 1.5 wt%, ILSS shows a plateau or decline, which means there is an optimal window (Figure 5). This result proves that the enhancement of nano-SiO2_2 in continuous fiber 3D printing system has nonlinear characteristics, and adding more does not necessarily produce better performance.

3.2 Microstructural Mechanism Analysis

Microstructure analysis shows how nano-SiO2_2 works. At low to medium concentration (0.5-1.5 wt%), nano-SiO2_2 is uniformly dispersed in the interface region (Figure 6). It acts as a “nano-splicing” site to improve the mechanical adhesion between matrix and fiber. This helps to change the interface, from fragile takeoff to stronger fracture. At high concentration (≥2.0 wt%), strong aggregation of nanoparticles was observed (Figure 7). This leads to stress concentration points, and at the same time, the viscosity of molten materials is too high, and fiber immersion is poor. This leads to micro-video in the interface, which reduces the enhancement effect.

3.3 Comparison with Existing Research

This study confirmed the reinforcing effect of nano-SiO2_2 in polymer matrix composites, and added the results of Sathyanarayana et al [2] and Rao & Kumar [3]. This research provides empirical support for the idea of single interface in 3D printing, which is consistent with Zhang et al.’ s [6] viewpoint, that is, “poor interfacial al adhesion is the main bottle

neck in FDM-CFRP", and proposes a solution, which can directly solve this problem by modifying materials rather than just optimizing the process. Xu and Zhang [5] suggest that the shape of nanoparticles affects the interface takeoff and plastic response; This study verifies the feasibility of nano-SiO2_2 to improve the performance by improving the microstructure of the interface during dynamic printing.

Different from the existing research, this study systematically explores the dose effect of nano-SiO2_2, and determines its optimal addition window (1.0 wt%-1.5 wt%) in continuous fiber 3D printing, thus providing specific guidance for the practical application of this technology.

4. CONCLUSIONS AND FUTURE WORK

4.1 Main Conclusions

This study shows that the amount of nano-SiO2_2 greatly changes the interface properties of continuous carbon fiber/PLA_3D printing composites. The change of force first rises and then falls. Between 0.5 wt% and 1.5 wt%, the interlaminar shear strength (ILSS) is greatly increased. Its peak value is between 1.0 wt% and 1.5 wt%, and the interface shows signs of failure. More than 1.5 wt%, there are more defects on the interface, because the aggregation of nanoparticles and melt immersion are not good. This leads to the stagnation or decline of ILSS, indicating that adding too much nano-SiO2_2 will eventually weaken the interface performance.

4.2 Research Limitations and Future Directions

This study has the following limitations: the nano-SiO2_2 used is an unmodified hydrophilic powder, which depends on physical dispersion, so it is difficult to completely avoid aggregation. We didn't use the transmission of electron microscope or the direct characterization of the three-dimensional structure and distribution of chemical components in the interface area of atomic force microscope; The investigation only focuses on interlaminar shear strength, but does not study tensile, bending, impact or fatigue behavior.

The future research can be carried out in the following directions: (1) Surface modification of nano-SiO2_2 with silane coupling agent to further improve dispersion and interface compatibility; (2) Integrating multi-scale characterization technology to establish the quantitative relationship among the distribution of nanoparticles, interface structure and macro-properties; (3) Study the applicability of modification strategy in complex structural parts, different types of fibers or different matrix systems, so as to promote the transition from laboratory research to engineering application.

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