Preparation and Properties of Tobacco Leaf Residue/PLA Composite Filaments for 3D Printing

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Abstract: To achieve high-value utilization of tobacco leaf residues after aqueous and ethanol extraction, this study used tobacco leaf residue powder as filler and polylactic acid (PLA) as matrix to prepare composite filaments suitable for fused deposition modeling (FDM) via melt blending and extrusion molding. The effects of 1, 3, and 5 wt% addition contents on material properties were investigated. The results showed that the tensile strength of the composites decreased significantly compared with neat PLA (about 54 MPa), being approximately 21, 18, and 22 MPa for the 1%, 3%, and 5% groups, respectively, with the lowest value in the 3% group. This indicates that the mechanical properties are jointly affected by physical structural factors such as filler dispersion, interfacial bonding, and interlayer fusion during printing. FTIR spectra showed that the main characteristic peaks before and after compositing were basically consistent, with no obvious new peaks or significant shifts, indicating that the tobacco leaf residue and PLA were mainly combined by physical blending. TGA results indicated that the material had good thermal stability below 300 °C and could meet the FDM processing window of 180-220°C; moreover, 1–5 wt% addition had no significant effect on the thermal degradation behavior. In bending tests, the 5% group exhibited better ductility and a relatively higher ultimate load, while the 1% and 3% groups underwent brittle fracture earlier, indicating a non-monotonic relationship between addition content and mechanical properties. In summary, the tobacco leaf residue/PLA composite filaments possess printability and thermal processing stability and are currently suitable for non-load-bearing, low-load, decorative, and cultural-creative 3D-printed products. Their comprehensive performance can be further improved through interfacial modification, powder dispersion, and printing process optimization.

Keywords: Tobacco Leaf Residue; Polylactic Acid (PLA); FDM 3D Printing; Composite Filament; High-Value Utilization

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1 Introduction

Fused deposition modeling (FDM) is currently one of the most widely used additive manufacturing technologies. Polylactic acid (PLA), owing to its biodegradability, good processing performance, and relatively high printing accuracy, has become the most commonly used thermoplastic matrix material in FDM printing [1]. However, PLA itself

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has inherent shortcomings such as high brittleness and limited mechanical strength. Therefore, modifying PLA with natural biomass fillers has become an important research direction in the field of green 3D printing materials [1, 2]. Previous studies have shown that lignocellulosic fillers, under appropriate addition amounts and dispersion states, can improve the tensile properties of PLA composites. For example, Morinda citrifolia bark particles at a 6 vol% addition increased the tensile strength of PLA by 15%, and the flexural strength and modulus by 25% and 34%, respectively [1]. However, the introduction of natural fillers may also lead to a decrease in material strength, and the effect depends on the synergistic action of filler type, addition amount, interfacial bonding state, and printing process parameters [3].

Meanwhile, large amounts of tobacco leaf residues generated during tobacco processing, including residues after aqueous and ethanol extraction as well as tobacco stems and other by-products, have long been disposed of by stockpiling or incineration, resulting not only in resource waste but also in certain environmental pressure [4]. China is the world's largest tobacco-growing country, with an annual tobacco leaf output of 2.188×10^6 t in 2022. The high-value utilization of tobacco waste is crucial for the green and sustainable development of China's tobacco industry [5]. Existing studies have explored resource utilization pathways for tobacco waste in directions such as biochar materials, biofuels, and organic fertilizers, but reports on using it as a filler for FDM 3D printing composite filaments are still very limited [6]. Exploratory studies on PLA and tobacco stem composite filaments have shown that tobacco powder with an average particle size of 50 μm can maintain chemical and physical integrity at an extrusion temperature of 180 °C and may act as a nucleating agent to increase PLA crystallinity, but the filament interior still suffers from defects such as particle agglomeration, bubbles, and pores. Foreign studies have also confirmed the feasibility of tobacco stem/PLA composite filaments for the FFF process and pointed out that adding plasticizers or compatibilizers to improve the interfacial bonding between PLA and tobacco particles is an important future research direction [7]. Introducing tobacco leaf residues as a filler into the PLA matrix to prepare composite filaments suitable for FDM is expected to simultaneously achieve resource utilization of agricultural by-products and cost reduction of 3D printing consumables [8]. This study addresses the problems of low utilization rate of tobacco leaf residues after extraction and low added value of traditional disposal methods. Using PLA as a biodegradable thermoplastic matrix and tobacco leaf residue powder as a filler, composite filaments suitable for FDM were prepared through melt blending and extrusion molding, exploring a new path for high-value utilization of tobacco processing by-products.

2 Materials and Methods

2.1 Raw Materials and Pretreatment

Tobacco leaf residues after aqueous and ethanol extraction were used as raw materials. Through drying, crushing, and sieving, tobacco leaf residue powder with a suitable particle size distribution was obtained, with an average particle size controlled at about 50 μm. PLA was used as the biodegradable thermoplastic polymer matrix. During pretreatment, the focus was on controlling the powder moisture content and particle size distribution to reduce bubbles, agglomeration, and uneven discharge during subsequent melt blending and extrusion.

2.2 Preparation of Composite Filaments

Tobacco leaf residue powder was melt-blended with PLA at addition amounts of 1 wt%, 3 wt%, and 5 wt%, with pure PLA as a control. Composite filaments were prepared by extrusion molding.

2.3 Preparation of FDM Printed Specimens

The prepared pure PLA filaments and tobacco leaf residue/PLA composite filaments were used for FDM 3D printing to print tensile and flexural test specimens. During printing, discharge uniformity, interlayer bonding state, nozzle clogging, filament breakage, warping, and surface quality were recorded to evaluate the FDM printing adaptability of the composite filaments. Pure PLA specimens were prepared under the same printing parameters as the performance comparison benchmark.

2.4 Performance Testing and Characterization

(1) Tensile performance test: A universal testing machine was used to test the tensile strength of pure PLA and 1%, 3%, and 5% tobacco leaf residue/PLA composites. Parallel

sample results for each group were recorded, averages were calculated, and comparisons with pure PLA were made to analyze the effect of addition amount on tensile performance.

(2) Infrared spectroscopy test: A Fourier transform infrared spectrometer was used to collect infrared spectra of pure PLA and the composites in the range of 4000–400 cm^{-1}. The positions, intensities, and shapes of the main absorption peaks were compared to determine whether the addition of tobacco leaf residues caused obvious changes in the main chemical structure of PLA and whether the compositing process was mainly physical blending.

(3) Thermogravimetric test: A thermogravimetric analyzer was used to test the thermal stability of the materials. The mass retention before 300 °C, main decomposition stage, initial decomposition temperature, and final residual mass were recorded to evaluate the thermal stability of the composites within the FDM processing window of 180–220 °C and to compare the effects of different addition amounts on thermal degradation behavior.

(4) Flexural performance test: The maximum flexural load, force-displacement curves, and fracture displacement of pure PLA and the composites were tested to evaluate their flexural load-bearing capacity and ductility. The effects of interfacial debonding, powder agglomeration, decreased FDM interlayer bonding, and increased internal defects on flexural performance were analyzed.

(5) Structure and defect analysis: Combined with the filament extrusion state, printed specimen appearance, and fracture characteristics, the effects of filler dispersion, agglomeration, pores, moisture content, and interlayer fusion on composite performance were analyzed.

2.5 Data Processing and Analysis

Parallel sample statistics were performed for the test results of each group, and the performance differences between pure PLA and the 1%, 3%, and 5% addition groups were compared. Combined with the tensile, flexural, FTIR, and TGA results, the non-monotonic relationship between tobacco leaf residue addition amount and composite performance was comprehensively discussed. The contributions of physical structural defects and interfacial interactions to mechanical performance were clarified, providing a basis for subsequent interfacial modification, dispersion optimization, and printing process regulation.

3 Results

3.1 Tensile Performance

Figure 1 showed the comparison of tensile strength between pure PLA and 1%, 3%, and 5% tobacco powder/PLA composites. The pure PLA specimen had the highest tensile strength, about 54 MPa; the tensile strengths of the 1%, 3%, and 5% addition composites were about 21, 18, and 22 MPa, respectively, decreases of about 61%, 67%, and 59% compared with pure PLA. Among them, the 3% addition group had the lowest strength, and the 5% addition group showed a slight recovery compared with the 3% group, indicating that under the current preparation conditions, the relationship between tobacco leaf residue addition amount and tensile strength is not simply linear but exhibits non-monotonic variation.

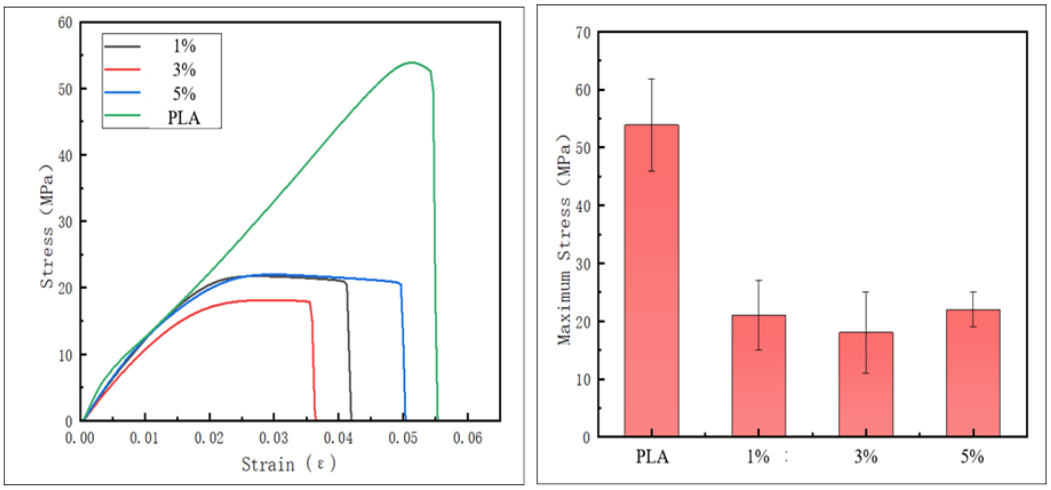


Figure 1 Effect of 1%,3%, and 5% tobacco powder and PLA composites on tensile strength

3.2 Infrared Spectroscopy

Figure 2 showed the FTIR spectra of pure PLA and 1%, 3%, and 5% tobacco powder/PLA composites. The positions of the main absorption peaks of all samples were generally consistent: the peak at about 1750 cm^{-1} is assigned to the stretching vibration of the ester carbonyl (C=O) in PLA, the peak near 1180–1080 cm^{-1} is mainly related to the C–O–C bond vibration in the ester group, and the peak near 1450–1360 cm^{-1} is related to the vibration of the —CH_3 group. With increasing tobacco powder addition, some peak intensities and baselines changed to a certain extent, but no obvious new absorption peaks or significant shifts of characteristic peaks were observed, indicating that the tobacco leaf residues and PLA were mainly combined by physical blending and particle interlocking, without causing obvious changes in the main chemical structure of PLA.

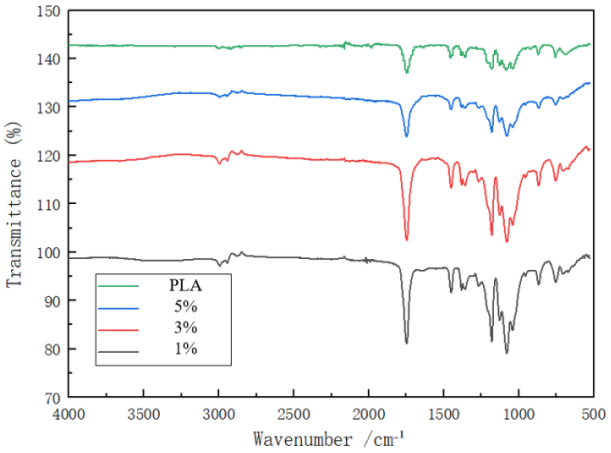


Figure 2 Differences in infrared spectra of 1%,3%, and 5% tobacco powder composites with PLA

3.3 Thermogravimetric Performance

Figure 3 showed the thermogravimetric curves of pure PLA and 1%, 3%, and 5% tobacco powder/PLA composites. All composites maintained high thermal stability below 300 °C, with no obvious rapid weight loss, and could meet the FDM printing processing window of 180–220°C. After 300°C, the main thermal decomposition stage began. The weight loss process of the composites was slightly earlier than that of blank PLA, among which the 3% addition group had a relatively lower initial decomposition temperature, while the curves of the 1% and 5% addition groups were relatively close. The TGA curves of the 1%, 3%, and 5% addition samples almost completely overlapped over the entire temperature range, with a residual mass difference of no more than 2% at the same temperature, and the final residual rate at 500 °C was 38% for all. This indicates that within the low filling range of 1%–5%, the tobacco leaf residue addition amount had no significant effect on thermal degradation behavior, and thermal stability was mainly dominated by the intrinsic characteristics of the PLA matrix.

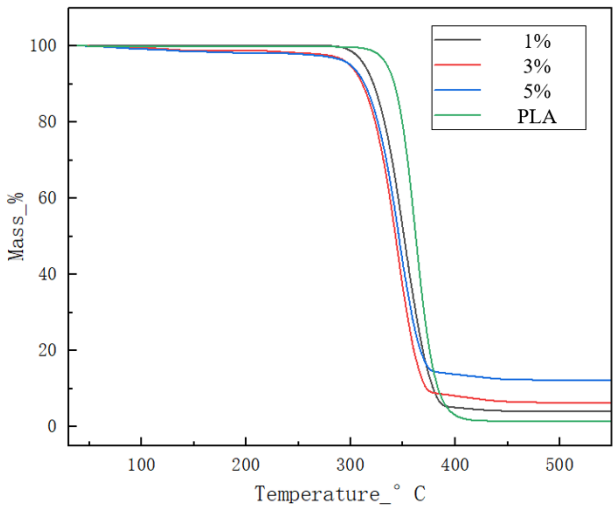


Figure 3 Thermogravimetric difference of tobacco powder (1%, 3%, 5%) and PLA composites

3.4 Flexural Performance

Figure 4 showed the flexural test results of pure PLA and 1%, 3%, and 5% tobacco powder/PLA composites. Pure PLA had the highest maximum flexural load, indicating the best flexural load-bearing capacity. After adding tobacco powder, the maximum flexural load of the composites decreased significantly, with the 3% and 5% addition groups being relatively lower. However, the 5% addition group had a maximum load (220 N) slightly higher than that of pure PLA (210 N), with a fracture displacement exceeding 20 mm and a gentle load decrease, showing good ductility; while the 1% and 3% addition groups underwent brittle fracture within 15 mm, with obviously insufficient load-bearing capacity. Flexural performance also showed non-monotonic variation, indicating that the effect of addition amount on mechanical performance is jointly controlled by interfacial chemical interactions and physical structural characteristics.

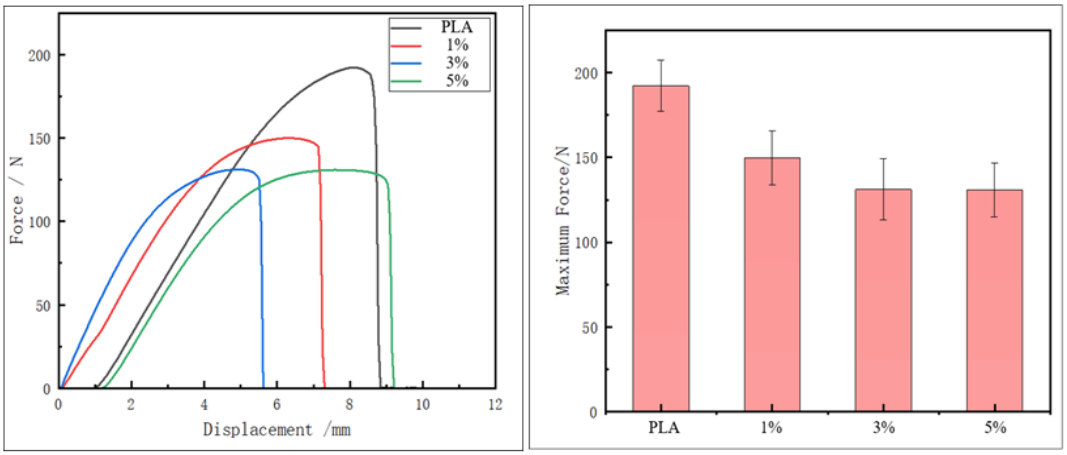


Figure 4 Comparison of bending performance between 1%,3%, and 5% tobacco powder and PLA composites

4 Discussion

The above results indicate that the mechanical performance of tobacco leaf residue/PLA composites does not decrease monotonically with increasing filler addition amount, but is jointly controlled by various physical structural factors such as interfacial compatibility, powder dispersion state, internal pores in the filament, moisture content, and FDM interlayer fusion [3, 9]. FTIR results showed that the main characteristic peaks of PLA before and after compositing were basically consistent, indicating that no obvious chemical bonding was formed between the tobacco leaf residues and PLA, and the interfacial interaction was mainly physical blending [10]. Due to the limited interfacial compatibility between tobacco powder and PLA, local defects may form around the particles and induce stress concentration during tensile testing, leading to a significant decrease in tensile strength compared with pure PLA [3]. The lowest strength in the 3% addition group may be related to local agglomeration or increased pores under this formulation; the recovery in the 5% group may be related to improved interlayer bonding and enhanced physical structural load-bearing capacity due to melt frictional heat compensation under high filling conditions [9]. TGA results further showed that the thermal degradation behaviors of the 1%, 3%, and 5% groups were almost completely equivalent, indicating that the abnormally low tensile strength of the 3% group was not caused by thermal degradation or differences in thermal stability, but more likely originated from physical structural defects such as uneven filler agglomeration and insufficient FDM interlayer fusion. In flexural tests, the 5% group showed good ductility and relatively high ultimate load, while the 1% and 3% groups underwent brittle fracture earlier, which corroborates the tensile results and indicates that the effect of addition amount on mechanical performance is a non-monotonic evolution process jointly controlled by interfacial chemical interactions and physical structural characteristics [10]. Overall, although the flexural load-bearing capacity of the current composites is lower than that of pure PLA, they still maintain good overall formability and certain deformation accommodation capacity. Subsequent improvements through compatibilizer or plasticizer modification, powder dispersion optimization, moisture content control, and interlayer bonding improvement can further enhance their comprehensive performance [3].

5 Conclusions

In this study, composite filaments suitable for FDM 3D printing were prepared by melt blending and extrusion molding using tobacco leaf residues after aqueous and ethanol extraction as filler and PLA as matrix. The effects of 1%, 3%, and 5% addition amounts on material performance were systematically evaluated. The results show that the composites exhibited excellent thermal stability below 300 °C , could meet the FDM processing window of 180–220 °C, and the 1%–5% addition amounts had no significant effect on thermal degradation behavior. FTIR analysis confirmed that the tobacco leaf residues and PLA were mainly composited by physical blending, without causing obvious changes in the main chemical structure of PLA. In terms of mechanical properties, the tensile strength of the composites decreased significantly compared with pure PLA, with the lowest value in the 3% group and a certain recovery in the 5% group ; the flexural performance also

showed non-monotonic variation, and the 5% group exhibited better ductility and a relatively higher ultimate load . Overall, the tobacco leaf residue/PLA composite filaments possess printability and thermal processing stability, but their current mechanical properties are not yet sufficient to replace pure PLA in load-bearing structural parts. At this stage, they are more suitable for cigarette pack models, display pieces, decorative parts, personalized cultural and creative products, and other low-load 3D printed products. Future research should focus on the interfacial compatibility between tobacco leaf residues and PLA, powder dispersion state, moisture content control, melt flow, and interlayer bonding during printing. On the premise of ensuring controllable raw material pretreatment and processing costs, the continuous preparation capability and FDM printing stability of the composite filaments should be improved, providing a feasible material pathway for the high-value utilization of tobacco processing by-products.

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