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Synthesis and Mechanical Properties of C/PLA 3D Printing Composites Based on Waste Rice Noodles

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Abstract:
Spherical carbon particles were prepared by using waste Guilin rice noodles as raw materials. By blending the rice noodles based carbon (RC) powders with polylactic acid (PLA), A series of black RC/PLA 3D printing composites were synthesized and characterized. The mechanical testing result shows that the RC/PLA 3D printing composites display better mechanical properties than that of pure PLA. Moreover, the composite with carbon treated with high temperature carbonization has better impact strength.
Keywords: *Waste Guilin rice noodles; RC/PLA 3D printing composite*

1 Introduction

Polylactic acid (PLA) has become one of the most commonly used raw materials for fused deposition molding (FDM) 3D printing, for it has many desirable characteristics such as excellent degradability, good biocompatibility, low warpage, and low shrinkage during FDM molding [1]. However, the printed products of pure PLA usually display high brittleness and low impact strength, which will limit their wider application [2-3]. Composite modification with carbon materials is an effective method for improving mechanical properties of PLA, and it has been reported that carbon fibers or carbon nanotubes can substantially enhance the tensile strength, modulus and impact strength of PLA printed products [4-7]. However, the high prices of carbon fibers or carbon nanotubes (higher than 1000RMB/kg) will increase the cost of carbon/PLA 3D printing composites and are not easy to be popularized.

On the other hand, Guilin rice noodle is a traditional snack and locals' daily diet in Guilin city, China. A large number (about 70 tons) of surplus waste rice noodles are produced every day. Nevertheless, most of the waste rice noodles are used as the raw material of low-margin fermented feed, and the others are abandoned directly, which may cause potential and long-term pollution to the soil and aqueous environment. Thus, it is necessary to explore an effective way for recycling waste rice noodles.

It has been proved that hydrothermal carbonization of many organic wastes can provide cheap biomass carbon materials [8-10]. The main constituent of waste Guilin rice noodles is starch (more than 70% at dry weight), which means that waste Guilin rice noodles may be carbonized in hydrothermal conditions and form carbon particles with specific morphology. Thus, to recover waste rice noodles effectively and lower the price of carbon/PLA 3D printing composites, we prepared a kind of carbon material with waste Guilin rice noodles as raw materials (RC) and blended it with PLA, synthesizing a series of low-priced RC/PLA 3D printing composites. The mechanical properties of the resulted RC/PLA 3D printing composites are also investigated in this paper.

2 Experimental section

2.1 Materials.

The polylactic Acid (PLA, 6202D, powder) with a density of 1.24 g/cm^3 and a molecular weight of 300000 ～ 320000 g/mol was purchased from NatureWorks LLC, USA. The waste Guilin rice noodle (main organic constituent: Starch 21.36g/100g; Protein 1.91 g/100g; Fat 0.4 g/100g; Water 76.33 g/100g) was collected from the canteen of Guilin University of Technology.

2.2 Preparation of carbon spheres based on waste Guilin rice noodles

10 g waste Guilin rice noodles were grinded to a smooth paste in a mortar, and then mixed with 20 g

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deionized water. The resulted mixture was transferred to a 50ml sealed Teflon-lined autoclave, and then heated at 200 ℃ for 10 hours. After natural cooling, vacuum filtration and drying at 60 ℃ for 24 hours, black hydrothermal carbon material (RC1) was obtained. The resulted C1 carbon material was high-temperature calcinated at 800 ℃ for 1 hour under N2 protection, forming carbon material with full carbonization (RC2).

2.3 Preparation and 3D printing of C/PLA 3D printing composite

3D printing wires of C/PLA composite were produced by a XinShuo WSJXT-12 miniature single screw extruder. At first, carbon powders (RC1 or RC2) were mixed with PLA powder with a certain mass ratio (1:99, 2:98, 3:97, 4:96 or 5:95) and then dried at 60 ℃ for 12 hours, respectively. The resulted RC/PLA mixture was added to the miniature single screw extruder with a feed inlet temperature of 70 ℃, a screw area temperature of 160 ℃ and a discharge outlet temperature of 80 ℃. The semi-molten RC/PLA composite was extruded from the discharge port, drawn through the circulating water cooling tank and then introduced into the silk collector. When the screw speed, traction speed and silk collector speed were adjusted to 42 rpm, 170 rpm and 7 rpm respectively, black 3D printing wires of RC/PLA composite with a diameter of 1.75 mm ±0.1 mm were obtained (See Figure 1a).

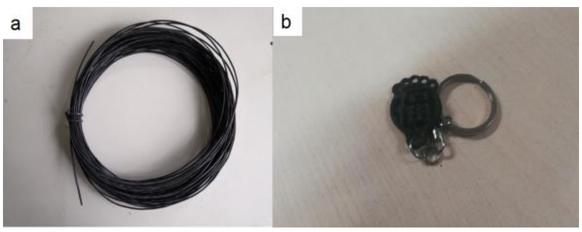

Figure 1 Photos of 3D printing wires (a) and 3D printed key chain (b) of RC/PLA composite

3D printing of RC/PLA 3D printing composite was carried out in a JG-Maker A3 FDM 3D printer, with an extrusion head temperature of 200 ℃, a bed temperature of 50 ℃, and a printing speed of 50 mm/s. The 3D printed product was a kind of black solid (See Figure 1b).

2.4 Characterization

The powder X-ray diffraction (PXRD) patterns were obtained with a PANalytical X 'pert PRO X-ray diffractometer with Cu Ka radiation (λ = 0.15418 Å) at 40 kV and 40 mA and a scan speed of 4°min^{-1} (2θ). The Infrared (IR) spectrum was recorded as KBr pellets at a range of 400-4000cm^{-1} on a Nicolet 5700 FT-IR spectrometer with a spectral resolution of 4.00 cm^{-1}. The morphology of the resulted carbon powders were characterized by using an S-4800 field emission scanning electron microscope (SEM), The IZOD impact strength of RC/PLA 3D printing composite was measured by a DR-802b cantilever beam impact testing machine with a impact energy of 1J. The tensile properties of RC/PLA 3D printing composite were tested by a UTM4503 electronic universal testing machine with a drawing speed of 1mm/min. All mechanical results presented in this paper represent the average measurement of four samples having the same composition.

3 Result and discussion

3.1 Characterization of carbon materials based on waste Guilin rice noodles

Figure 2a shows the PXRD diagram of carbon materials RC1 and RC2 based on waste Guilin rice noodles. The diffraction peaks around 26° and 44° could be assigned to (002) and (100) planes of graphite [11], respectively, which indicated that the resulted carbon materials were partially graphitized. Compared with RC1, the diffraction peaks of (002) planes in RC2 shifted to a higher angle, and the peak of (100) planes was evident, which indicated that calcination at 800 ℃ could enhance the graphitization of the materials.

The IR spectra of carbon materials RC1 and RC2 were basically similar (See Figure 2b). There were hydrophilic groups like hydroxyl and amino groups on the surface of these carbon particles. The wide absorption peak at about 3420 cm^{-1} corresponded to the stretching vibration of O-H and N-H bonds, while the bending vibrations of these function groups were located at about 1600cm^{-1}.

Most carbon particles based on waste Guilin rice noodles showed a spherical morphology with a diameter range of 4-30 μm. The surface of RC1 was smooth, while RC2's surface was rough with many small protuberances (See Figure 2c and 2d). The rough surface in RC2 particles could improve the adhesion between the carbon particles and matrix PLA materials and might be advantageous in enhancing the mechanical property.

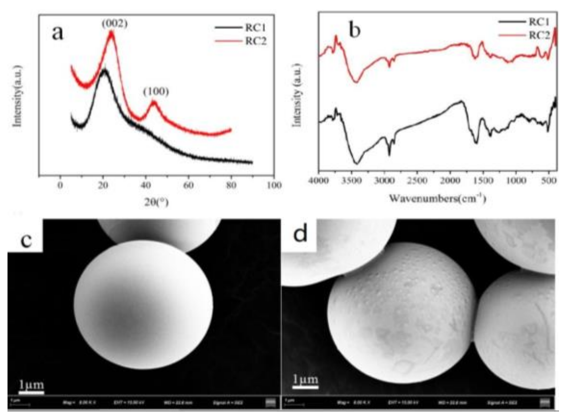

Figure 2 (a) PXRD pattens of carbon materials RC1 and RC2; (b) IR spectra of carbons materials RC1 and RC2; (c)-(d): SEM images of carbon materials RC1(c) and RC2(d)

3.2 Mechanical properties of RC/PLA composites

Figure 3 shows the IZOD impact strength diagram of RC/PLA 3D printing composites. The pure PLA displayed a weak IZOD impact strength of 1.28 kJ/m^2, which could be improved remarkably by the introduction of RC. The IZOD impact strength of RC1/PLA composites increased with the increasing of RC1 content when the mass fraction of RC1 was not more than 4wt%, while decreased when exceeding this limit. The impact strength of RC1/PLA composite with 4 wt% RC1 was 1.88 kJ/m^2, which was 47% higher than that of pure PLA (See Figure 3a).

On the other hand, the IZOD impact strength of RC2/PLA composites also increased greatly with the increasing of RC2 addition, while the improvement effect of RC2 on the impact strength of PLA based printing products was much higher than that of RC1. The impact strength of RC2/PLA composites with 5 wt% RC2 was up to 9.68 kJ/m^2 and 7.5 times more than that of pure PLA (See Figure 3b). It might be due to the higher graphitization degree and rough surface of RC2, which could absorb the impact energy and combine with the PLA matrix more effectively.

The introduction of RC could also improve the tensile properties of PLA material. As shown in Figure 4a, with the increasing of RC1, both tensile strength and elongation at break of RC1/PLA composites increased firstly and then decreased. The RC1/PLA composite with 2 wt% RC1 displayed the highest tensile strength of 30.0 MPa and good elongation at break of 18.9%, which were 71% and 154% higher than that of pure PLA, respectively. A similar phenomenon was observed in the RC2/PLA system. However, though with high IZOD impact strength, the RC2/PLA composite contained 5 wt% RC2 displayed relative weak tensile properties (Tensile strength: 19.86MPa; Elongation at break: 10.76%), which was only slightly higher than that of pure PLA.

4 Conclusion

In this paper, we converted Guilin rice noodles to spherical carbon particles (RC), and synthesized a series of black RC/PLA 3D printing composites. The result RC/PLA 3D printing composites have better mechanical properties than pure PLA. In particular, calcinated carbons (RC2) in RC/PLA composites could effectively improve the low impact resistance of the PLA matrix. The RC2/PLA composite containing 5wt% RC2 has outstanding impact strength (9.68 kJ/m^2, 7.56 times of pure PLA), and its tensile strength (19.86MPa) and elongation at break (10.76%) are also improved compared with that of pure PLA. Such RC/PLA 3D printing composites have a higher added value than that of fermented feed, which could be advantageous for recycling waste rice noodles. On the other hand, due to low-cost carbon particles from waste Guilin rice noodles, the resulted RC/PLA 3D printing composites could also be a new way for low-cost 3D printing.

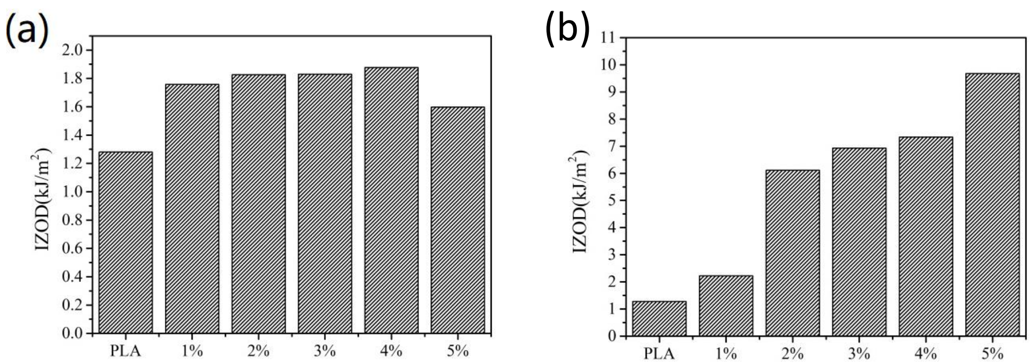

Figure 3 Izod impact strengths of RC/ PLA composites with different amount of RC1 (a) and RC2 (b)

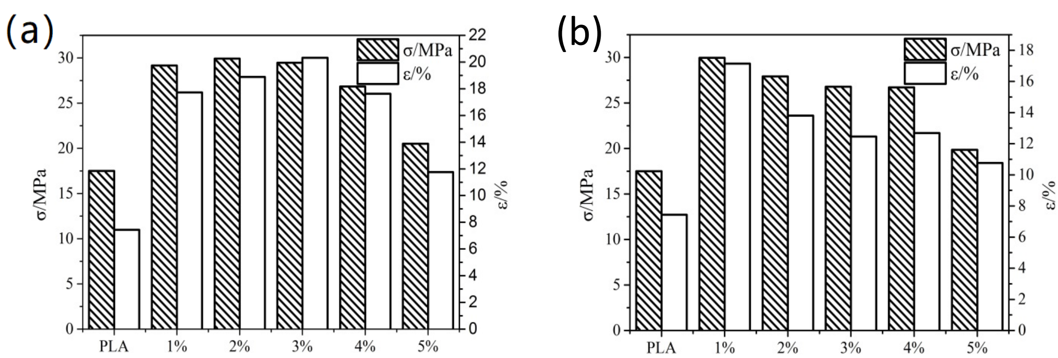

Figure 4 Tensile properties of RC / PLA 3D printing Composites with different amount amount of RC1 (a) and RC2 (b)

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