Plasticizer Combinations and Performance of Wood Flour–Poly(Lactic Acid) 3D Printing Filaments

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Wood flour-poly(lactic acid) 3D printing filaments were prepared *via* a melt extrusion method. Poplar wood flour and poly(lactic acid) (PLA) were used as raw materials, and different combinations of glycerol and tributyl citrate (TBC) (4% glycerol, 2% glycerol + 2% TBC, 4% TBC) were used as plasticizers. A 3D printer was used to print the filaments into standard test specimens with dimensions of 150 mm × 10 mm × 0.2 mm at the printing temperature of 220 °C. The performance of wood flour-poly(lactic acid) 3D printing filaments in terms of their interfacial compatibility, mechanical properties, melt index (MI), water absorption, and heat stability was tested under different plasticizer combinations. The results showed that under the condition of same dosage of plasticizer, the order of MI for the 3D printed filaments from high to low was 4% glycerol > 2% glycerol + 2% TBC > 4% TBC, which indicated that glycerol was more favorable for the extrusion processing of the composite filaments. However, in terms of compatibility, mechanical properties, water absorption, and thermal stability, the 3D printing filaments with 4% TBC showed better performance compared with other groups.

Keywords: 3D printing; Plasticizer; Polylactic acid; Wood plastic composites; Performance

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INTRODUCTION

Three-dimensional (3D) printing is a new type of technology that integrates digital modeling, mechanical and electrical control, information technology, materials science, chemistry, and many other fields of advanced technology, which is known as the core technology in the third industrial revolution (Du *et al.* 2014). It is currently used for rapid prototyping as well as some highly customized end-products. 3D printing is superior to traditional manufacturing technology in the aspects of structure optimization, material saving, and energy saving in the production process (Choonara *et al.* 2016). However, there are also some disadvantages associated with it. A 3D printed article consists of many droplets of polymer from the print filament that are poorly adhered, leaving voids within the article, and thus decreasing the mechanical and other properties of the article. How to improve these properties and enlarge the application of 3D printing in the fields of aerospace, biological medicine, construction, and others has become a hot research spot. Many studies have been conducted to improve the mechanical properties of 3D printed articles (Stevanovic *et al*. 2013; Slapnik *et al*. 2016). Along with the advancement and maturity of 3D printing technology, the combination of 3D printing with the traditional manufacturing process can continuously promote the transformation and upgrading of the traditional manufacturing industry.

The currently used 3D printing materials mainly include engineered plastics, photosensitive resin and rubber materials, metal materials, and ceramic materials. In addition, colored gypsum material, artificial bone powder, cell biological materials, and food material such as granulated sugar, also have been applied in 3D printing (Leong *et al.* 2003; Garcia *et al.* 2012; Wang *et al.* 2015; Tian *et al.* 2016). However, there are also some disadvantages for the current 3D printing materials in terms of performance, cost, and environmental protection.

The plastics used in traditional wood-plastic composite materials generally include petrochemical products, which are non-biodegradable materials. With the increasing awareness on environment protection, the use of renewable natural resources to develop new composite materials has become one of the hot topics in the study of composite materials. Wood flour/polylactic acid (PLA) composite-based 3D printing filaments as a new type of 3D printing filaments not only can integrate the biodegradable properties of PLA and low-cost of wood flour, but also it can facilitate full recycling of wood processing residues, thus reducing the waste of resources. Therefore, it is a new type of biodegradable 3D printing filament with broad prospects for further development. However, it is noted that although wood flour and PLA can be mixed evenly and wood flour can be covered by polylactic acid molecules, the interfacial compatibility and tenacity of the composite materials are not satisfactory (Oksman *et al.* 1998; Lai *et al.* 2003). Moreover, the addition of wood flour can limit the movement of the macromolecular chain of PLA and reduce the stereo-tacticity of PLA, which leads to a significant reduction in the crystallization ability of PLA (Liu *et al*. 2013).

To improve the compatibility of wood flour/PLA plastic composites, it is necessary to modify the formulation. The common method used is to add plasticizer, elastomer, or silane coupling agent into the mixture of wood flour/PLA (Li *et al*. 2011). Several studies have been conducted to analyze the performance of wood-plastic composites with different plasticizers or coupling agents. For example, Yang *et al*. (2014) prepared wood flour/polylactic acid composites using ethylene glycol, glycerol, and polyethylene glycol 400 (PEG400) as plasticizers and analyzed the effect of different plasticizers on the properties of composite materials. Faludi *et al*. (2013) applied two reactive coupling agents, N,N-(1,3-phenylene dimaleimide) (BMI) and 1,1-(methylenedi-4,1-phenylene) bismaleimide (DBMI) to improve interfacial adhesion in PLA/wood composites. It is noted that most studies in wood-plastic composite have been mainly focused on the material itself and its formulation. However, there has been a lack of studies on the performance of wood flour-poly(lactic acid) 3D printing filaments with plasticizers.

Therefore, the purpose of this study is to determine the optimal plasticizer by analyzing the performance variations of wood flour-poly (lactic acid) 3D printing filaments in terms of interfacial compatibility, mechanical properties, melt index (MI), water absorption, and heat stability under different plasticizer combinations.

EXPERIMENTAL

Materials

Materials used were poplar wood flour (the wood flour was sieved into 140-mesh to 160-mesh size powder) and PLA. Experimental reagents used were glycerol (Shangqiu Liangfeng Health Products Co., Ltd., Shangqiu, China), tributyl citrate (TBC) (Shanghai Macklin Biochemical Co., Ltd., Shanghai, China), and distilled water. A total of 270 g of

wood flour and 630 g of PLA were used to prepare the filaments *via* three kinds of solution, and the specific steps are described below.

Preparation of plasticised composites

The wood flour and PLA were evenly divided into three groups. In the first group 4% glycerol (based on its total weight) was added, in the second group 2% glycerol and 2% TBC (based on its total weight) were added, and in the third group 4% TBC of its total weight was added. These mixtures were then sealed in plastic bags and kept for 18 h to ensure the plasticizer effectively dispersed in the mixture (Zuo *et al*. 2015).

Filaments preparation

A twin-screw extruder (SHJ-20, Nanjing Giant Machinery, Nanjing, China) was used for pelleting, and the extrusion temperature for the sections numbered from material inlet (I) to outlet (V) was set at 135 °C, 150 °C, 170 °C, 170 °C, and 135 °C, respectively. Then, a small single-screw extruder (Kunshan Huanxinyang Electrical Equipment Co., Ltd., Kunshan, China) was used for extrusion molding, to acquire the samples of 3D printer filaments with a diameter of 1.75 mm at an extrusion temperature of 170 °C (Fig. 1(a)).

Standard specimen

A 3D printer (MakerBot Replicator 2, Makerbot Company, New York, USA) was used to print the prepared filaments into standard test specimens with dimensions of 150 mm × 10 mm × 0.2 mm at the printing temperature of 220 °C (Fig. 1(b)).

(a) (b)

Fig. 1. Images of 3D printing filaments (a) and a standard specimen (b)

Methods

Structure assessment

The D/max-220 X-ray diffractometer made by Rigaku (Tokyo, Japan) was used in XRD testing, and the testing conditions were set as follows: 40 kV voltage, 30 Ma current, 5° starting angle, 40° termination angle, and 0.02° gradual scanning step width. By analyzing the X-ray diffraction (XRD) pattern and material composition, the crystallinity changes of the specimens were evaluated. The compatibility between wood flour and PLA was primarily compared among three groups of specimens.

The Quanta-200 SEM (FEI, Hillsboro, Oregon, USA) was used to observe the section of the filaments. Brittle failure was conducted on the filaments in liquid nitrogen, and then the section was sprayed with metal first and tested later. The amplification factor for filaments was 500 and the 3D printed test specimens were amplified by 1000. By observing the highly amplified section profile, further comparisons of the different compatibility among the three groups were made.

Mechanical properties

The tensile property of wood flour/PLA printed test specimens were tested as per the national standard GB/T 1040-92 (1992), using a CMT-5504 omnipotent mechanical tester (Shenzhen Suns Technology Stock LLC., Shenzhen, China). Each test specimen was tested 5 times and its average was applied. T

Through comparison of the tensile strengths and breaking elongations of the standard test specimens, the solution with the best mechanical performance among the three groups was identified.

Melt index (MI)

The MI was measured according to the national standard GB/T 3682-2000 (2000), under the conditions of 170 °C and 2.16 kg, using a RLS-400 melt index meter (Chengde Kecheng Testing Machine LLC., Chengde, China). By comparing the liquidity among the three groups, the level of difficulty in extruding the wood-plastic composite 3D printing filaments can be obtained.

Water absorption

According to the requirements of GB/T 1034-98 (1998), a test specimen was cut into dimensions of 50 mm × 10 mm × 0.2 mm and then vacuum-dried for 24 h at 50 °C, and subsequently weighed (recorded as m_1, g). Under room temperature (25 °C), the test specimen was submerged into distilled water and soaked for 24 h. The specimen was then removed, wiped by filter paper, and weighed again (recorded as m_2, g). The water absorption rate was calculated according to Eq. 1.

$$\text{Water absorption (\%)} = (m_2 - m_1) / m_1 \times 100 \quad (1)$$

The comparison of results on the degree of water absorption expansion among the three testing groups can reflect the degree of compatibility between the wood flour and PLA.

Heat stability

A thermogravimetric analyzer (TGA) 209-F3 (Netzsch, Free State of Bavaria, Germany) was applied to conduct the heat stability test on the composite filaments.

The temperature was increased from room temperature to 600 °C at the speed of 10 °C /min, the nitrogen flow was set at 30 mL/min, and the sample-feeding volume was kept at 5 mg.

By analyzing the TGA and the derivative thermogravimetric (DTG) images, one can compare the thermal stability of the filaments in the three groups.

RESULTS AND DISCUSSION

Effect of Various Plasticizers on the Compatibility of Composite Filaments and Specimens

The use of a plasticizer in a composite can reduce the interfacial energy, promote the dispersion of wood flour, prevent the agglomeration of dispersion phase, and improve the interphase adhesion (Liu *et al.* 2013). Thus it would increase the interfacial compatibility of the two phases. The XRD diffraction diagrams of wood flour/PLA 3D printed specimens produced by using various plasticizers are shown in Fig. 2.

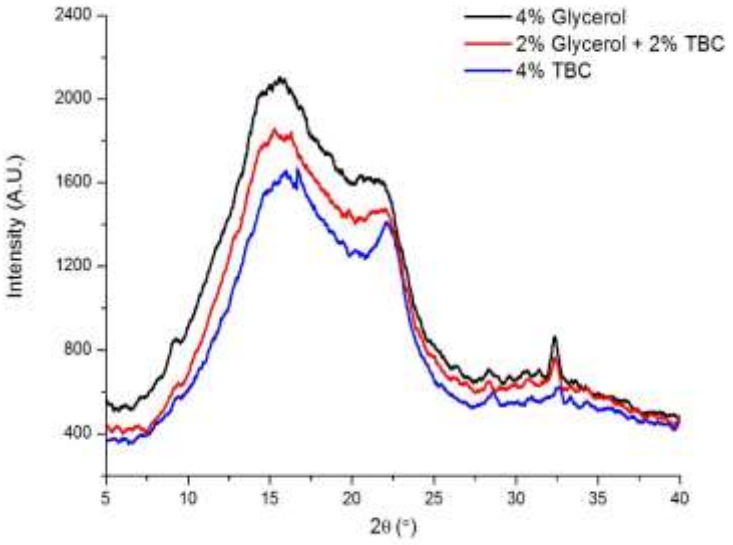


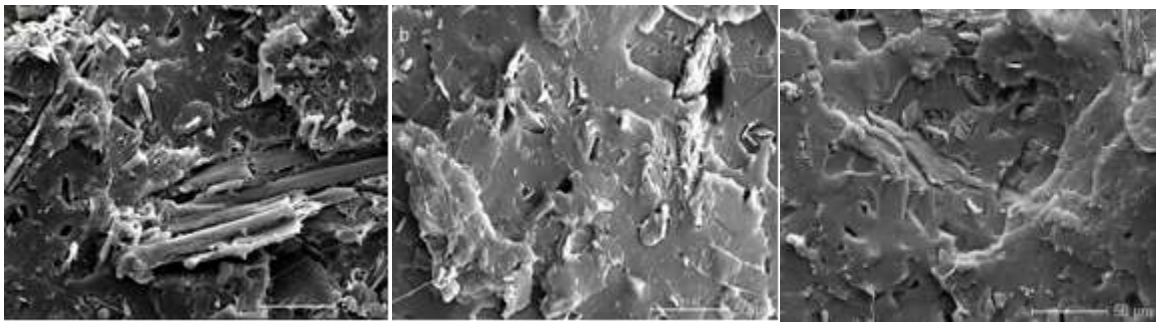
Fig. 2. XRD diffraction patterns of specimens with various plasticizers

It is known that the peak at $2\theta = 17°$ is the characteristic peak of PLA (Zhang 2012). As indicated in Fig. 2, at $2\theta = 17°$, the order of the intensity of crystallization peak of the 3D printing standard specimens with different plasticizer combinations from strong to weak was: 4% glycerol> 2% glycerol + 2% TBC> 4% TBC. Based on the data in Fig. 2, the degree of crystallinity was computed, and the results are shown in Table 1. In comparison with the other two group specimens, the degree of crystallinity for the group with 4% TBC as plasticizer was relatively lower, which indicated that the interfacial compatibility of the wood flour-poly(lactic acid) composite was better. In this case, the crystal structure of PLA was damaged by TBC and wood flour can enter into the crystalline region, so the degree of crystallinity was diminished. In addition, the surface of wood flour was covered by PLA, which hindered the molecular nucleation and also improved the compatibility of the two phases.

Table 1. Degree of Crystallinity of Composites with Different Plasticizers

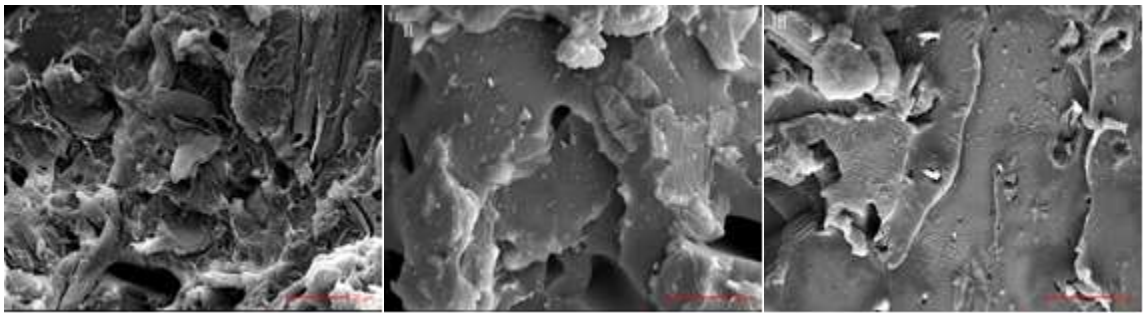
Plasticizers	4% glycerol	2% glycerol + 2%TBC	4% TBC
Degree of crystallinity / %	40.29	39.14	38.43

To further analyze the effects of the three kinds of plasticizers on the interfacial compatibility of wood flour and PLA, SEM was used to test the wood flour/PLA composite filaments and standard 3D printing specimens, and the results are shown in Figs. 3 and 4.



(a) b) (c)

Fig. 3. SEM for filaments (Magn 500x): (a) 4% glycerol; (b) 2% glycerol + 2% TBC; and (c) 4% TBC



(a) (b) (c)

Fig. 4. SEM images for 3D printing specimens (Magn 1000x): (a) 4% glycerol; (b) 2% glycerol + 2% TBC; and (c) 4% TBC

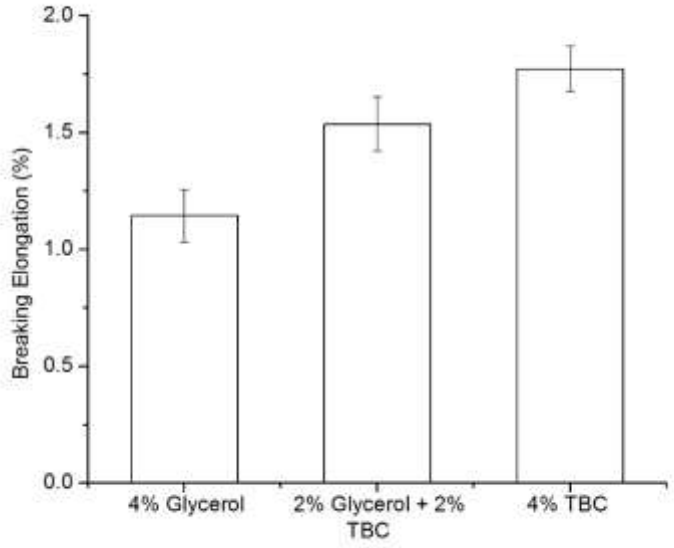
The interaction between hydrophilic wood flour and hydrophobic PLA was generally inferior, as they belong to two incompatible systems. It was shown from Figs. 3 and 4 that there was a very clear interface between the wood flour and PLA substrate on the section images of groups 1 and 2. The interface appeared uneven, and the uneven phenomenon in group 1 was more severe than that in the second group, which indicated that there was poor compatibility between the wood flour and PLA, and that the combination effect between the wood flour fiber and PLA particles was not satisfied. The SEM images of group 3 (4% TBC) showed that there was no clear boundary between the wood flour and PLA, and no clear prominent wood and PLA particles were found, which suggested that the adhesive strength of the two materials was strong, and their interdependency was good. Therefore, the order of the compatibility between the wood flour/PLA composite for the filaments and the 3D printed specimens from high to low were: 4% TBC > 2% glycerol + 2% TBC > 4% glycerol.

Effect of Various Plasticizers on the Mechanical Properties of 3D Printed Specimens

The mechanical properties of the composites were closely related to the morphological structure of wood flour in the PLA substrate, while its dispersion state was dependent on the plasticizer (Liu *et al*. 2005). On one hand, the plasticizer reacted with the

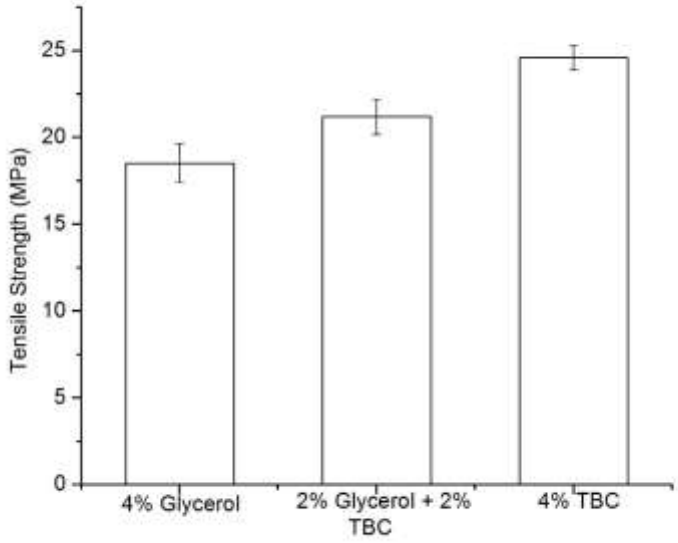
hydroxyl groups present on the surface of wood flour to lower the polarity of the surface of wood flour; on the other hand, the plasticizer allowed the immersional wetting of the PLA substrate on the surface of wood flour and increased the compatibility of both. In addition, it was favorable for the formation of a mechanical interlock structure, which increased the interfacial interaction between the PLA substrate and wood flour.

Figure 5 illustrates the effect of different plasticizers on the mechanical properties of wood flour/PLA 3D printed specimens. As shown in Fig. 5, the tensile strengths of the three groups were 18.53 MPa, 21.18 MPa, and 24.60 MPa, respectively; while the breaking elongations were 1.14%, 1.54%, and 1.77%, respectively. The order of the mechanical properties' effect on the 3D printed specimens from high to low was 4% TBC > 2% glycerol + 2% TBC > 4% glycerol. The tensile strength was mainly affected by the interfacial compatibility of composite specimens. When under stress, stress concentration will occur at the interface between wood flour and PLA. If the compatibility is better, the interdependence between wood flour and PLA would be stronger, which can afford greater stress. The XRD patterns and SEM images of the three groups indicated that the compatibility of the composite specimens with 4% TBC was the best, so the tensile strength was the largest. The breaking elongation was affected by the degree of crystallinity. The group with 4% TBC had the smallest degree of crystallinity, so the tenacity of the composite specimens was the best and the breaking elongation was the largest.



(a)

Fig. 5(a). Mechanical properties of 3D printed specimens with different plasticizers; (a) Breaking elongation, (b) Tensile strength



(b)

Fig. 5(b). Mechanical properties of 3D printed specimens with different plasticizers; (a) Breaking elongation, (b) Tensile strength

Effect of Various Plasticizers on MI of Filaments

The wood flour/PLA composite filaments were prepared *via* a melt extrusion method under the condition of melt flow. Therefore, it was required that the processed material had proper flowability. If the flowability was too small, it was unfavorable for mould filling and the extrusion would become difficult. However, if the flowability was too large, enough extrusion pressure was hard to form; it is also unfavorable for the product shaping and leads to strength defects in final products (Maiti and Hassan 1989). The molecular chain of wood flour contains a large amount of hydroxyl groups and hence its surface can show powerful polarity. When wood flour was combined with nonpolar PLA, the composite filaments formed under the state of melt belonged to a complex rheological system. Therefore, it was necessary to evaluate the melt flowability of wood flour/PLA composite filaments prepared by using various plasticizers.

As shown in Fig. 6, the MI of the three groups were 14.70 g/min, 9.70 g/min, and 7.09 g/min, respectively, which can meet the processing requirements. The order for the MI of the 3D printed filaments from high to low was 4% glycerol > 2% glycerol + 2% TBC > 4% TBC. The influence of glycerol on the MI values was larger than that of TBC, which indicated that glycerol was more favorable for the extrusion processing of the composite filaments. The melting point of wood flour with high crystallization was higher than its degradation temperature, making it difficult to be processed. To increase the flowability of the melted wood flour, the plasticizer needs to be added. Under the action of the heat and shearing force of the extruder, glycerol can effectively destroy the hydrogen bonds and crystallinity of wood flour, and lower its intermolecular force. Therefore, in comparison with TBC, glycerol can more easily plasticize wood flour. When the plasticized wood flour was mixed with PLA, the melt flowability of the composite was enhanced.

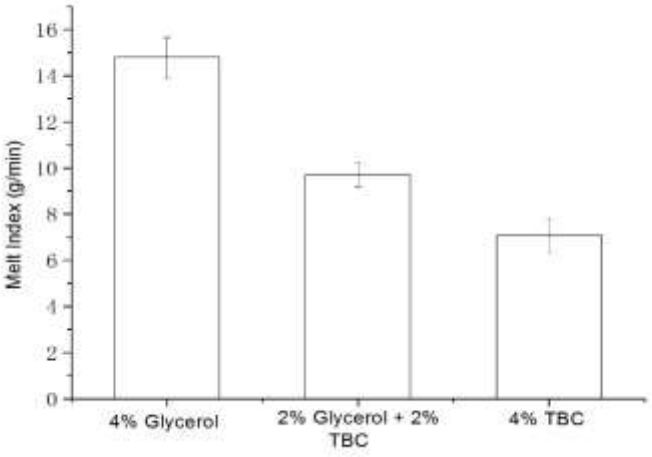


Fig. 6. Melt index of composite filaments with various plasticizers

Effect of Various Plasticizers on Water Absorption of 3D Printed Specimens

When the wood flour and PLA were mixed directly, the interfacial bonding force was inferior, and the product was more sensitive to moisture. As the water absorption of the 3D printing material could be directly affected by its service environment, it was necessary to measure the water absorption of 3D printed specimens, and the result is shown in Fig. 7.

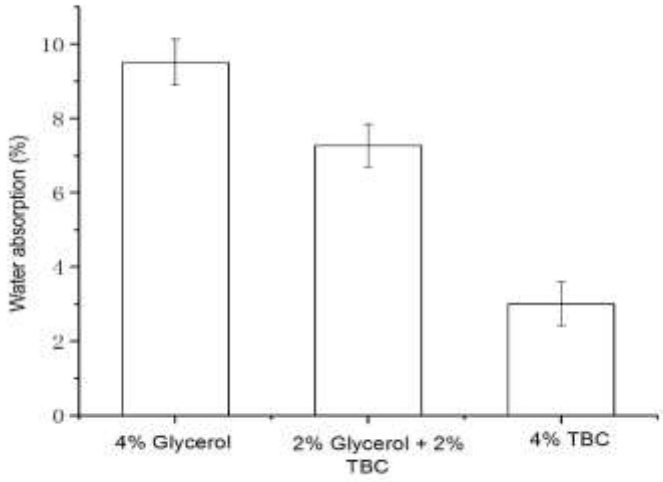


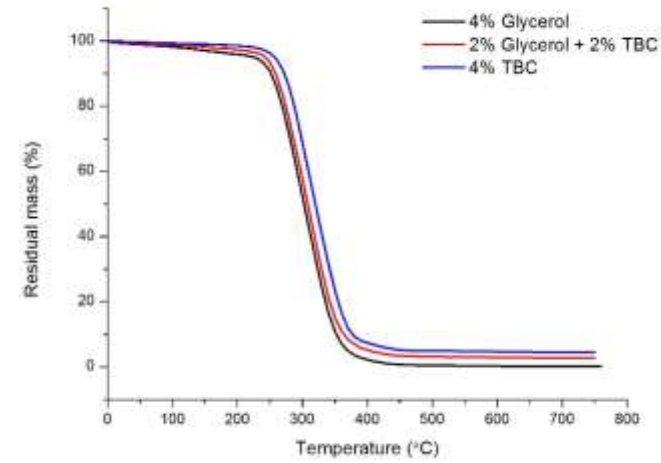
Fig. 7. Water absorption of 3D printed specimens with various plasticizers

As shown in Fig. 7, the water absorption rates of the three groups were 9.51%, 7.27%, and 3.01%, respectively. The water absorption of the 3D printed specimens was larger when 4% glycerol was added than the mixture of glycerol and TBC, or TBC itself. This is due to the fact that glycerol contained more hydrophilic hydroxyl groups than TBC. In addition, the exposed hydroxyl groups in glycerol increased the water absorption rate of 3D printed specimens greatly. Therefore, the main factors that affected the water absorption of 3D printed specimens were the effect of plasticizer on the compatibility of

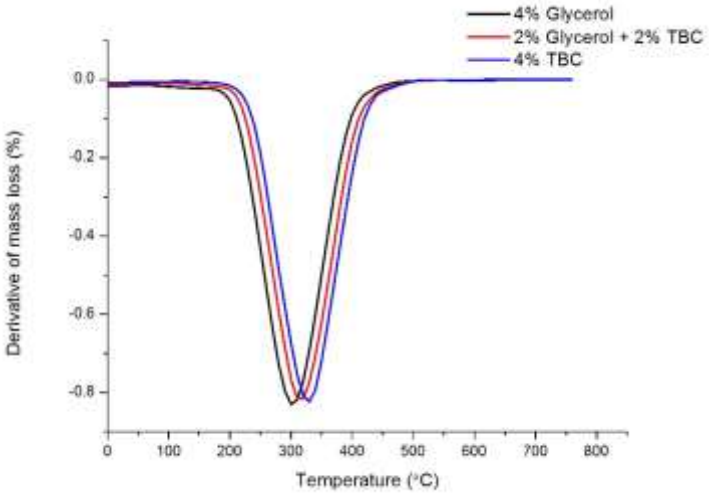
wood flour/PLA and the quantity of hydrophilic hydroxyl groups in the plasticizer molecule, *i.e.* the more the hydroxyl groups on the surface the larger the water absorption rate became.

Effect of Various Plasticizers on Heat Stability of Filaments

The thermal performance of 3D printing filaments is an important parameter for assessing the processing temperature of the material and the scope of operational temperature. In addition, it also reflects the heat stability of the material. Therefore, it has great influence in analyzing the thermal properties of composite filaments. Figure 8 shows the TGA and DTG curves of the wood flour/PLA composite filaments with different plasticizer combinations.



(a)



(b)

Fig. 8. The TGA and DTG curves of the wood flour/PLA filaments with various plasticizers: (a) TGA curves; and (b) DTG curves

As shown in Fig. 8(a), the 3D filaments with different plasticizer combinations started to degrade at 250 °C and the entire process was a first-order degradation occurring between 250 °C and 500 °C. The heat stability of the three groups was different due to the

different initial decomposition temperature and the order from high to low was: 4%TBC>2%TBC+2% glycerol>4% glycerol. The DTG curves in Fig. 8(b) showed that the temperatures corresponding to the maximum thermal decomposition rates of the composite filaments in the three groups with different plasticizer combinations were different, and the order from high to low was: 4%TBC>2%TBC+2% glycerol>4% glycerol. These findings indicated that 4%TBC can effectively improve the heat stability of composite filaments compared with the other two groups. TBC can enhance the interfacial adhesion between wood flour and PLA, so more energy will be needed in order to damage the adhesion during the process of filament degradation, which led to the increase of heat stability.

CONCLUSIONS

1. The dosage of 4% TBC can effectively destroy the crystallization structure of PLA and improve the interfacial compatibility of wood flour-poly(lactic acid) 3D printing filaments compared with other two plasticizer combinations (4% glycerol, 2% glycerol + 2% TBC).

2. In terms of compatibility, mechanical properties, water absorption, and thermal stability, the 3D printing filaments with 4% TBC showed better performance compared with other two plasticizer combinations; however, the MI index was relatively lower. Therefore, further research should be conducted in order to improve the melt flowability of the wood flour-poly(lactic acid) 3D printing filaments.

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