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Preparation of starch/acrylonitrile-butadiene-styrene copolymers (ABS) biomass alloys and their feasible evaluation for 3D printing applications

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ABSTRACT

With debranching and plasticization of starches, we have successfully prepared thermoplastic starches (TPS) with high processibility by a twin-screw extruder. Afterwards, the TPS have blended with appropriate amounts of acrylonitrile-butadiene-styrene copolymers (ABS), compatibilizers, impact modifiers, and pigments to compound in a twin-screw extruder, manufacturing TPS/ABS biomass alloys. Finally, we have prepared white and black filaments, whose diameters of 1.75 mm, for additive manufacturing (AM) with TPS/ABS biomass alloys by a single-screw extruder as well as proper mold and also executed their measurement of physical properties. In addition, their feasible evaluation for 3D printing applications has also been made. Experimental results reveal that physical properties of lab-made white and black filaments (i.e. mechanical properties, thermal resistance, flowability, and emissions of volatile organic compounds (VOCs)) are superior to those of commercial ABS filaments and the shaping samples for 3D printing have also been successfully fabricated, preliminarily demonstrating that they are potential biomass polymeric materials with excellent physical performances and high processibility for 3D printing utilizations.

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1. Introduction

In recent years, the 3D printing technology has become an attractive research topic for industrial [1], mechanical [2], and biomedical [3–5] applications owing to its advantages of easy operation, low cost, and fast manufacturability. Nowadays, there are some 3D printing techniques for metals, ceramics, papers, and polymers such as selective laser sintering (SLS) [6], stereolithography (SLA) [7], plaster-based 3D printing (PP) [8], fused deposition modeling (FDM) [9], etc., rendering the researcher and engineer alternative forming procedures. Among these 3D printing techniques, FDM is the most conspicuous additive manufacturing (AM) method with the melt and extrusion of thermoplastic polymers because of its high processibility, low cost, and facile manipulation [10]. Although several commercial polymeric materials (e.g. acrylonitrile-butadiene-styrene copolymers (ABS), polylactic acid (PLA), and so on) have been developed for 3D printing utilizations, they still cannot fully meet the practical requirements since ABS exhibits excellent mechanical properties with high processing stinks [11–13] and PLA is an environment-friendly polymers with poor mechanical performances [14–16]. Therefore, preparation of eco-friendly polymeric materials with good physical properties for FDM has become a crucial issue.

Starches, whose basic repeating units are represented with D-glucose molecules connected by glycosidic bonds [17], are eco-friendly polymeric materials consisting of two structural classes (i.e. linear amylose and chain-branched amylopectin). However, the chain-branched amylopectin exhibits poor processibiliy [18]. Although Kaseem et al. [19] have prepared thermoplastic starch (TPS)/ABS blend with glycerol as a plasticizer, the rheological results indicate that the blend is pseudo plastic and its processibiliy is insufficient. In order to give the TPS/ABS blends qualified processibiliy, we have firstly debranched the starch with α-isoamylase as an enzyme and plasticized the starch with glycerol/water as a plasticizer to produce the TPS. Afterwards, the TPS/ABS biomass

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alloys for 3D printing with competent physical performances such as high melt indices, good mechanical properties, moderate processing stinks, and high thermal stabilities have been manufactured by the introduction of ABS, compatibilizers, impact modifiers, and pigments into TPS. Eventually, the feasible evaluation of lab-made TPS/ABS biomass alloys for 3D printing applications has also successfully been executed, proving that they are potential AM polymeric materials with high processibility and excellent physical properties. Moreover, the researches about the 3D printing applications of TPS/ABS biomass alloys are never reported.

2. Experimental

2.1. Materials and instruments

Starch, plasticizers (i.e. glycerol and water), enzyme (i.e. α-isoamylase), ABS (U200B), compatibilizer (i.e. styrene-maleic anhydride copolymer; SMA; Fig. 1), impact modifier (i.e. methylmethacrylate butadiene styrene; MBS; Fig. 2), and pigments (i.e. titanium white (TiO_2) and carbon black (CB)) were purchased from Aldrich Co., Echo Chem. Co., Union Chem. Co., Mitsubishi Chem. Co., Acros Organics Co., Sartomer Co., and Janie Color Co., respectively. All the materials in this study were used without further purification. The 3D printer (i.e. da Vinci 1.0) and commercial white ABS filaments for 3D printing were acquired from XYZprinting Co.

2.2. Preparation of white and black TPS/ABS biomass alloy filaments for 3D printing and measurement of their physical properties

The manufacturing procedure of white TPS/ABS biomass alloy filaments for 3D printing was shown in Fig. 3. With α-isoamylase (100 g), glycerol (50 g), and water (50 g), the starch (5 Kg) was debranched and plasticized at 70 °C and 500 rpm by a twin-screw extruder (APV Chemical Machinery MP 2015) and then pelletized at 100 °C and 600 rpm, obtaining TPS pellets. Afterwards, TPS pellets

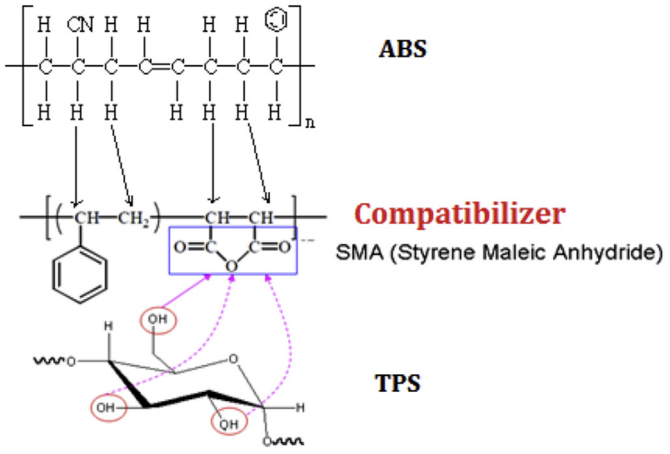

Fig. 1. Chemical structure of SMA and its compatible mechanism with TPS and ABS.

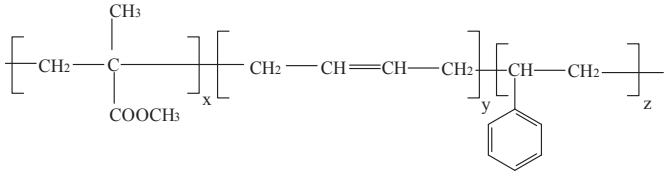

Fig. 2. Chemical structure of MBS.

were compounded with ABS, SMA, MBS, and TiO_2 at 180 °C and 600 rpm by a twin-screw extruder (APV Chemical Machinery MP 2015), procuring white TPS/ABS biomass alloys. The prescription and composition of white TPS/ABS biomass alloys were manifested in Table 1 and Fig. 4. Finally, white TPS/ABS biomass alloy filaments for 3D printing, whose diameters were 1.75 mm, were prepared with white TPS/ABS biomass alloys and a proper die at 220 °C and 1000 rpm by a single-screw extruder (Brabender PL 2100). The similar procedure was executed in the manufacture of black TPS/ABS biomass alloy filaments for 3D printing except the pigment was replaced with CB.

Furthermore, we prepared the specimens for the mechanical and thermal properties with white and black TPS/ABS biomass alloy filaments by a 3D printer (da Vinci 1.0) at 210 °C. Heat distortion temperature (HDT), flexural modulus, notched Izod impact strength, melt index (MI), tensile strength, flexural strength, and volatile organic compound (VOC) emission were measured with the standards of ASTM D648, ASTM D790, ASTM D256, ASTM D1238, ASTM D638, ASTM D790, and ASTM D3960, respectively.

3. Results and discussion

As shown in Tables 1 and 2, we have firstly manufactured TPS/ABS biomass alloy **a** with 30wt% TPS and 70wt% ABS. The HDT, flexural modulus, impact strength, MI, tensile strength, and flexural strength of TPS/ABS biomass alloy **a** are 92 °C, 21,500 Kgf *cm/cm, 8.26 J/m, 9.2 g/min, 355 Kgf/cm^2, and 626 Kgf/cm^2, respectively. Its physical properties cannot meet the requirements of polymeric materials for 3D printing while the HDT, flexural modulus, impact strength, MI, tensile strength, and flexural strength of the commercial white ABS are 100 °C, 23,000 Kgf *cm/cm, 15.00 J/m, 10.0 g/min, 400 Kgf/cm^2, and 700 Kgf/cm^2, respectively. In order to enhance the heat resistance, flowability, and mechanical properties, we have tried to blend the compatibilizer (SMA) with TPS/ABS biomass alloy **a**, obtaining TPS/ABS biomass alloy **b**. Experimental results reveal that introduction of SMA can dramatically improve the heat stability, flowability as well as mechanical performances and only HDT as well as impact strength cannot achieve the specification. This result represents SMA is an effective compatibilizer for TPS and ABS. As manifested in Fig. 1, the hydrogen bond takes place between the anhydride group of SMA and the hydroxyl group of TPS while strong van der Waals force generates between the alkyl chains of SMA and ABS. The production of hydrogen bond and strong van der Waals force may practically combine TPS and ABS to form the TPS/ABS alloy, promoting the thermal stability, flowability, and mechanical properties. Consequently, we have added the impact modifier (MBS) into TPS/ABS biomass alloy **b**, acquiring TPS/ABS biomass alloy **c**. Although introduction of MBS causes the slight decreases of HDT, flexural modulus, MI, tensile strength, and flexural strength, the impact strength of TPS/ABS biomass alloy **c** rises to 14.83 ft-lb/in.

To further heighten the impact strength, we have increased the blending amount of MBS to 2 wt% and the impact strength of TPS/ABS biomass alloy **d** achieves 18.57 ft-lb/in, surpassing the physical properties of commercial white ABS for 3D printing. The HDT, flexural modulus, MI, tensile strength, and flexural strength of TPS/ABS biomass alloy **d** are 96 °C, 24,200 Kgf *cm/cm, 11.9 g/min, 481 Kgf/cm^2, and 751 Kgf/cm^2, respectively. In case of TPS/ABS biomass alloy **d**, merely the HDT cannot fit the benchmark. Eventually, we have blended the white pigment (TiO_2) with TPS/ABS biomass alloy **d**, manufacturing TPS/ABS biomass alloy **e**. Experimental results manifest that addition of TiO_2 can further raise the thermal resistance and the HDT, flexural modulus, impact strength, MI, tensile strength, and flexural strength of TPS/ABS biomass alloy **e** are

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