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Influence of the process parameters on the mechanical properties of engineering biocomposites using a twin-screw extruder

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ABSTRACT

The utilization of bio-based engineering polymers as a matrix material for cellulosic fiber reinforced composites has become an important focus in materials research. This is due to a rising demand for sustainable materials from renewable resources. In addition to this aspect, the bio-based materials provide an advantage for lightweight applications with their lower density. In this investigation, the completely bio-based polyamide 10.10, with a melting point above 200 °C, was used as a polymer matrix. Chopped man-made cellulose fibers (Cordenka CR-Type) were investigated as reinforcement for use in injection molded applications. A co-rotating twin-screw extruder with a screw-diameter of 18 mm was used for compounding. It was verified that reinforcing polyamide 10.10 with 20 wt% and 30 wt% cellulosic fibers is possible, resulting in an increase of impact and tensile properties. Furthermore, it was shown that the temperatures and screw-configurations of the twin-screw extruder only result in different fiber length distributions but in minor differences of the morphological structure and mechanical properties of PA 10.10 with 20 wt% fibers. Compounds with 30 wt% cellulose fibers show significant higher impact properties that those with 30 wt% glass fibers.

1. Introduction

Over the past decade, natural fiber composites have risen in their importance for technical parts [1]. Especially in the automobile industry, natural fibers were used as reinforcement in thermoplastic matrix materials for interior parts [2]. Due to their thermal sensitivity, natural fibers are mainly used as reinforcement in polyolefins [2] and bio-based polyesters [3], because of their lower melt temperatures. Man-made cellulose fibers have been used as reinforcement in previous studies and showed a significant increase in the mechanical properties, especially in the case of the notched impact strength [4–8].

For the last few years, engineering thermoplastic resins reinforced with cellulosic fibers, such as wood pulp or man-made cellulose fibers, have been becoming increasingly the focus of research [9], mainly with polyamides [10–13,17–19]. The reasons for using engineering matrix materials are the higher thermo-mechanical properties, especially the higher heat-distortion temperature in comparison to polyolefin matrix materials. For this reason applications "under the hood" [12,13] are possible. The use of cellulosic reinforcement fibers also offers significant potential for lightweight applications in comparison to common glass fiber reinforcement [14–16].

One of the first publications by Klason et al. [17] about polyamides with cellulosic reinforcement describes compounding of PA 6 and PA 12 based on crude oil with cellulose fibers. They found a significant increase of the tensile strength and the Young's modulus for the bio-composites. Sears et al. [18] applied for a patent in 2000 which describes a method, so called "low temperature compounding technique" to reinforce petro-based PA 6 and PA 6.6 using cellulose fibers. This compounding method is based on a twin-screw extruder and an adapted temperature profile for gentle processing of the thermal-sensitive fibers.

Using bio-based polyamides as matrix material for a composite offers the possibility to achieve a bio-content of 100%. The raw material for bio-based polyamides is castor oil, which is obtained from the fruit of the castor plant [20–22]. Nowadays, the most popular types of bio-based polyamides are PA 6.10, PA 10.10 and PA 4.10. However, only the PA 10.10 is completely based on renewable raw materials. Feldmann [10,11] shows that man-made cellulose fibers with bio-based polyamides (PA 6.10 and PA 10.10) achieve higher strength and impact properties than lignocellulose fibers, such as abaca fibers. Moreover the man-made fibers show a higher thermal-resistance than the abaca fiber. The properties of the completely bio-based engineering composites with 30 wt%

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man-made cellulose fibers are comparable with the properties of common PA 6 with 30 wt% glass fibers [10,11]. For these studies special pultrusion methods, a single-step and a two-step process, were used.

Because of the difficulties in feeding man-made cellulosic fibers, different processes like compression molding [23], pultrusion [11,24,23,25] and kneading [26,8] were used for preparing the composites. In the literature, no studies about compounding engineering polymers with cellulosic fibers by using a conventional twin-screw extruder and a metering feeder system are described. It is necessary to use this conventional setup to bring up these composites to an industrial scale. For this reason, the influence of the screw configuration and the process temperature of a twin-screw extruder relating to the mechanical properties was investigated in this study, expecting that a gentle process configuration of the sensitive cellulose fibers results in higher mechanical properties.

2. Experimental

2.1. Materials

PA 10.10. (Vestamid Terra DS) was provided by the company Evonik Industries and used as a matrix polymer. The raw material of the completely bio-based copolyamide is castor oil which is obtained from the fruits of the castor-plant. The monomers are hexamethylenediamine and decandiacid. The Melt Flow Index (MFI) of the matrix polymer is 90 g/10 min (230 °C/5 kg)/130 g/10 min (250 °C/5 kg) according to ISO 1133.

Chopped, man-made cellulose (viscose) fibers with a filament diameter of approx. 12 μm and a length of 2 mm were used as reinforcement. Besides the spin finish, the fibers were prepared with a PPL sizing (aqueous polyvinyl alcohol solution) of approx. 10 wt% for use in a gravimetric feeding system. Also, glass fibers (Lanxess CS 7928) with a length of 4.5 mm and a sizing suitable for polyamides were investigated for reference purposes (Table 1).

2.2. Composites preparation

Commercial glass fiber reinforced thermoplastic compounds commonly contain 30 wt% glass fibers. According to a similar fiber volume of the cellulose fibers in the prepared compounds, an amount of 20 wt% of cellulosic fibers was chosen for this investigation. Additionally one compound with 30 wt% cellulosic fibers was prepared to compare the same fiber weight content.

2.2.1. Compounding

The composites were compounded with a ZSE 18 HPE (Leistritz) twin screw extruder with a screw-diameter of 18 mm and a process length of 40 D. The polyamide was dried below 0.1% moisture content in a dry air drier (TORO-systems TR–Dry–Jet EASY 15) and the cellulosic fibers below 1% moisture content in an air convection oven before compounding. The polyamide granules and the chopped fibers were fed into the side feeder of the compounder at Section 3 by a gravimetric feeding system (see Fig. 1). The screw speed of the extruder was optimized in preliminary tests to 200 rpm and a throughput of 4 kg/h. Afterwards, the strand was cooled down on a discharge conveyer and was pelletized to a length of 3 mm by a strand pelletizer (Scheer SGS 25-E).

Two different screw configurations (Fig. 1) were used in this investigation in order to determine the mechanical properties of the compounds relating to the exposure to shear strain. The screw configuration (C1) includes kneading discs and mixing elements to mix and distribute the fibers after they have passed through the feeding zone. The configuration (C2) is totally different and only consists of conveying elements after the fiber feeding zone. The measured torque of the twin-screw extruder showed mean values of 69.5% with screw configuration C1 and 58.2% with configuration C2. That indicates a lower application of mechanical energy into the material in consequence of the used conveying elements. The influence of the processing temperature was investigated using two different temperature settings (see Table 2).

In order to minimize the mechanical stresses induced by shear strain, a single-screw extruder (*S*) with a standard three-section screw was used for reference purposes. The screw-diameter of the single-screw extruder, Schwabenthan Polytest 30P, was 30 mm, the process length was 25 D and the overall throughput was approx. 4 kg/h. The temperatures of the three sections were set at 200 °C/205 °C/210 °C and the die temperature was 230 °C, based on T1.

2.2.2. Injection molding

Prior to the injection molding process, the cellulose fiber reinforced compounds were dried using an air dryer until a moisture content of 0.1% was achieved. The test specimens for the tensile test were manufactured according to DIN EN ISO 527-1A using an injection molding machine (Klöckner Ferromatik FM85) with a screw diameter of 40 mm, and a clamping force of 85 kN. The cycle time was approx. 40 s, including a cooling time of 20 s. The processing temperatures are shown in Table 3.

2.3. Characterization

All composites were characterized under dry state conditions and at room temperature.

2.3.1. Colorimetry

The cellulose fiber reinforced materials discolor to brown due to thermal and mechanical stresses during processing. To measure this discoloration, the colorimetry UltraScan Pro provided by Hunterlab was used. This colorimetry uses the L, A, B color space, wherein the L-Value represents the lightness of the color. As such, this value of the compounds was evaluated. To measure the brightness of the granules, a petri dish was filled with each type of compound and closed by another petri dish. Afterwards, the fully filled

Table 1
Properties of the matrix material and properties of the reinforcement fibers [10,11,22].

Matrix material	Density (g/cm^3)	Tensile strength (MPa)	Tensile modulus (MPa)	Notched impact strength (kJ/m^2)	Melting temperature (°C)
PA 10.10-Vestmaid Terra Ds14 (Evonik)	1.04	38	1802	2.75	200°C
Fibers	Density (g/cm^3)	Diameter (μm)	Tensile strength (MPa)	Tensile modulus (MPa)	Elongation at break (%)
Man made cellouse Cordenka CR type	1.5	12	825	22	13
Glass fibers Lanxess CS 7928	2.6	11	2600	73	3.5

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