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The effects of the injection moulding temperature on the mechanical properties and morphology of polypropylene man-made cellulose fibre composites

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ABSTRACT

Composites made of polypropylene and man-made cellulose fibres that are intended for injection moulding applications show potential for use in sustainable and light weight engineering with high energy absorption capacity. Due to the thermal sensitivity of the cellulose fibres, process parameters play an important role during the injection moulding process. A polypropylene and a man-made cellulose fibre were chosen for this investigation. Effective melt temperatures between 200 °C and 269 °C were used to process the compounds into test specimens. Tensile, impact and colorimetric tests, as well as an SEM analysis, and a measurement of the fibre length distribution were carried out in order to characterise the mechanical and optical properties of the composites. It was observed that the fibre length becomes shorter above 256 °C and elongation at break and Charpy strength (notched) of the composites already decrease at lower temperatures than tensile strength. A direct correlation between mechanical properties and discoloration was not observed. Therefore, melt temperatures up to 250 °C are suitable for these composites.

1. Introduction

At least since Henry Ford's invention of the "Soybean Car" in 1941, biocomposites have been in the focus of scientists and various industries for their potential in technical applications, especially in the mobility sector. Their lightweight potential and properties that are similar to those of their glass fibre reinforced counterparts are incentives for the use of natural fibres as reinforcement [1–4]. However, up until now, there have been very few studies that deal with the process–structure–property relations relevant when employing industrial processes, especially at higher melt temperatures, when using engineering matrix materials.

Typically, polypropylene (e.g. [2,5,6]) or polyethylene (e.g. [7–9]) are used as matrix materials for injection moulded or extruded applications. Recently, polylactic acid (e.g.[5,10–13]), and other bio-based polymers (e.g. PHB [14], PBS [15,16] or starch blends [17–19]) have gained popularity in this area because of their high level of biodegradability and the high content of renewable resources they contain. However, the thermomechanical properties of the aforementioned composites cannot compete with conventionally reinforced engineering plastics, particularly when the heat deflection temperature or impact strength are important. Different studies show that man-made cellulose fibres (rayon fibres) [20–22] possess significant potential to improve the impact properties of thermoplastic matrices. This was verified by long fibre pull-outs that resulted from a defined/tailored fibre matrix adhesion. In addition to the lightweight potential and ecological aspects, the author showed that thermoplastic biocomposites also have potential to compete with or surpass common reinforced plastics, particularly when the contain engineering plastics (e.g. polyamides [23,25] or POM [26]) as their matrix material and man-made cellulose fibres as reinforcement. In the case of matrices with a higher melting temperature (>200 °C), the thermal and mechanical stresses the fibres are subject to during the compounding and injection moulding processes have to be considered. The authors of previous studies verified that the compounding method [23] and screw configuration [25] have a significant influence on the fibre length distribution in polyamides composites. However, not only composites that contain plastics that melt at higher temperatures have this conflict. These process parameters also influence polypropylene that contains cellulosic fibres. Often times, the melt must possess enhanced flow properties to enable longer flow paths. For this reason, a higher melt temperature is

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used. The thermal degradation of treated, untreated, and modified cellulosic fibres was reported in various earlier and more recent studies (e.g. [27–31]). It was reported that natural fibres progress through three steps of degradation (including moisture) with the highest thermal stability for α-Cellulose. Especially for man-made cellulose fibres Yang and Kokot [31] reported that these fibres require a higher activation energy than natural fibres due to their stable lattice structure. However, the degradation rate of man-made cellulose fibres is higher due to the lower degree of polymerisation in the investigated man-made fibres. Hori and Wada [33] reported a geometrical change (lateral shrinkage of cell unit) of highly crystalline cellulose II and III_{II} for temperatures between RT and 250 °C due to the crystal structure and hydrogen bonds. Besides fibre degradation and structural changes, a higher melt temperature also affects the lower viscosity, and, therefore, results in less shear stress during injection. This can lead to longer fibres in the part, or to lower local temperatures.

The aim of the study is to evaluate the influence of different melt temperatures on the mechanical properties of man-made cellulose fibre reinforced composites that are based on polypropylene during the injection moulding process. These results should also provide information about the impact of the melt temperature when using engineering thermoplastic matrix materials, like polyamides.

2. Experimental

2.1. Materials

The polypropylene PP 575P (homopolymer, injection moulding grade) from the company SABIC was used as the polymer matrix. Man-made cellulose (viscose) fibres Cordenka 700 Super 3 (T0, 1350 filaments per roving) with a filament diameter of approx. 12 μm and a spin finish were used as reinforcement. The properties of the materials are shown in Table 1.

2.2. Composites preparation

A composite based on polypropylene with a fibre content of 30 wt% and without any coupling agent was prepared and investigated in this study.

2.2.1. Compounding

The composites were compounded using a special compounding technique, as reported by the author in [23,24]. This pultrusion process is very gentle on the fibres, even at higher temperatures, which leads to better mechanical properties in the final composite. Finally, the strand was pelletised into granules with a length of 3 mm, and the material was ready for injection moulding. The melt temperature was set at 200 °C in the extrusion nozzle, and the haul off speed was 5 m/min.

2.2.2. Injection moulding

Before injection moulding, the compounds were dried using a TORO-systems TR–Dry–Jet EASY 15 until a moisture content of 0.1% was left. The test specimens (Type 1A) for the tensile test were manufactured on an injection moulding machine (Klöckner Ferromatic FM85), which had a screw with a diameter of 40 mm, and a clamping force of 85 kN. The cycle time was approx. 35 s, including a cooling time of 20 s. The processing temperatures and injection pressures are shown in Table 2. Specimens with melt temperatures below 200 °C and above 270 °C could not be manufactured with the injection moulding machine, because either the pressure or the level of degradation were too high. The mould temperature was set at 40 °C, and the cavity pressure at approx. 500 bar ± 10 bar.

2.3. Characterisation

All composites were characterised in dry state, and while they were at room temperature.

2.3.1. Colorimetry

The cellulose fibre reinforced materials discoloured due to thermal and mechanical stresses during processing. In order to measure this discolouration of the specimen, the colorimetry UltraScan Pro provided by Hunterlab was used. This colorimetry uses the Lab color space system.

2.3.2. Thermogravimetric Analysis (TGA)

The mass loss caused by the degradation of the man-made cellulose fibres at higher temperatures was evaluated using the Q500 produced by TA instruments. Nitrogen was used as a flushing gas, and a first step for drying the fibres was integrated at 110 °C. When the weight remained steady, the temperature was set to the actual level (200 °C, 220 °C, 240 °C, 260 °C and 280 °C) needed to measure the mass loss induced by the injection moulding parameters that took place at higher temperatures. The heating rate was 40 K/min and the evaluation time was 15 min after reaching the actual temperature. For reference purposes, the polypropylene was also measured to assess the amount of matrix degradation.

2.3.3. Scanning Electron Microscopy (SEM)

The morphology of all composites was investigated using the scanning electron microscope Zeiss Ultra 55 plus. Fractured notched Charpy specimens were coated with gold, and images were taken with a low level of magnification (250×) to obtain an overview of the fibre distribution and the fibre pull-outs. In order to gain a closer look on the micro level, images with a higher magnification were prepared (1000×).

2.3.4. Fibre length distribution

A dynamic image analyser (QICPIC) with a wet dispersing unit MIXCEL from the company Sympatec was used to measure the fibre length distribution in the test specimens. For this purpose, a piece of a test specimen with a length of 10 mm was cut out for

Table 1
Selected properties of the matrix material and of the reinforcement fibres [34].

Matrix material	Density (g/cm^3)	Tensile strength @ yield (MPa)	Tensile modulus (MPa)	Notched impact strength (kJ/m^2)	Melting temperature (°C)	MFI (5 kg) 200/220/240/260 °C (g/10 min)
PP 575P (Sabic)	0.91	34.1	1523	3.45	166 °C	27/41/61/86
Fibres	Density (g/cm^3)	Diameter (μm)	Tensile strength (MPa)	Tensile modulus (GPa)	Elongation at break (%)	Titre (dtex)
Cordenka 700 Super 3	1.5	12	825	22	13	2440