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Procedural influences on compression and injection moulded cellulose fibre-reinforced polylactide (PLA) composites: Influence of fibre loading, fibre length, fibre orientation and voids

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ABSTRACT

The influence of fibre loading (20, 30, 40 mass%), fibre fineness, and the processing procedure (compression moulding – CM and injection moulding – IM) on the tensile and impact strength of lyocell/PLA composites was examined. The results revealed a significantly higher tensile and impact strength for CM composites compared to IM composites. An increase in strength up to a fibre loading of 40% was determined for CM composites, while for IM composites the highest values were measured at a fibre loading of 30%. Composites were investigated for their void content, fibre orientation, fibre length and process-induced fibre damage. A better fibre/matrix adhesion and compaction of IM composites was found while fibre orientation as well as mechanical properties of extracted fibres show no significant differences between CM and IM composites. The different mechanical characteristics of CM and IM samples are attributed predominantly to the fibre aspect ratio and the distribution of voids.

1. Introduction

Apart from perfectly aligned (unidirectional) man-made fibre-reinforced composites with a good compaction (a good compaction means a low volume of voids) and adhesion between fibre and matrix, composite properties are in general difficult to compare. Characteristics of fibres and matrix itself [1–3], fibre loading [4–6], fibre/matrix adhesion [7–9], fibre orientation [10,11], fibre length [1,12,13], compaction [14] and the production process as well as the procedural settings affect the composite properties clearly. It is well known that compression moulded (CM) long glass fibre-reinforced composites display higher strength and impact strength values than short glass fibre-reinforced injection moulded (IM) composites [4,5,10].

For a relatively new composite system like cellulose fibre-reinforced bio-based plastics it is very important to investigate the influences of CM and IM on the resulting composite characteristics systematically. It is crucially to understand the relationship between properties and structures of fibres, matrix and the resulting composite. Mechanical properties were investigated as a function of different fibre loadings, fibre fineness values, compaction, fibre orientation, fibre length distribution and potential damage of the fibres by the different processes (CM vs. IM). Due to the lower variation and the reproducing quality compared to natural fibres, regenerated cellulose fibres (lyocell) were used as reinforcement in a polylactide (PLA) matrix. The analyses are guided by the following working hypotheses:

1. A better wetting of the reinforcing fibres with the PLA matrix takes place during IM compared to CM due to a more effictive mixing of fibres and matrix.
2. In composites with good compaction an increasing fibre loading leads to improved mechanical properties.
3. Since the specific fibre surface becomes smaller an enlarging fibre diameter results in a decrease of the mechanical composite properties due to the lower bonding surface between fibre and matrix.
4. Fibres in CM composites are better oriented in a preferred direction than in IM composites. The fibre orientation in CM specimens is not affected by the fibre loading and the fibre fineness. In IM samples an increasing fibre loading and the use of finer fibres leads to a reduced preferred orientation

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because finer fibres are less aligned in a preferred direction due to the lower bending stiffness compared to coarser fibres.

5. Fibre shortening during IM is not affected by the fibre diameter. But an increasing fibre loading leads to a stronger fibre shortening and fibre damage compared to the CM process due to frictional effects.

2. Materials & methods

2.1. Fibres & matrix

Tencel® lyocell fibres were supplied by Lenzing AG (Lenzing, AT) with fibre fineness values of 1.3, 3.3, 6.7, 12.0 and 15.0 dtex. The length of the staple fibres was in the range between 38 and 100 mm.

Polylactide (PLA) was used as a matrix and was supplied in fibre form (Ingeo fibres type SLN 2660 D; Eastern Textile Ltd., Taipei, TW) with a fineness of 6.7 dtex and a staple fibre length of 64 mm. Fibres were produced from a NatureWorks™ 6202D PLA with a density of 1.24 g/cm^3, a melting temperature of 160–170 °C and a glass transition temperature of 60–65 °C.

2.2. Fibre characterisation

Prior to the investigations fibres were conditioned according to DIN EN ISO 139 at 20 °C and 65% relative humidity for at least 18 h.

Tensile characteristics. Tensile characteristics of the fibres were determined with a Fafegraph M testing machine (Textechno, Mönchengladbach, DE) working with a pneumatic clamping system (PVC/ PMMA clamps). 70–80 fibres were investigated with a load cell of 100 cN at a gauge length of 3.2 mm and a testing speed of 2 mm/min. For the removement of the fibre crimp a clamping mass of 200 mg was used for fibres with a fineness of 1.3 to 6.7 dtex and of 350 mg for fibres with a fineness of 12.0 and 15.0 dtex.

Fibres having a fineness of 1.3, 6.7 and 15.0 dtex were additionally investigated at gauge lengths of 1, 2, 5 and 10 mm.

Fibre extraction. To investigate the fibre length distribution and the tensile strength of processed fibres, fibres were extracted from the composites. For this purpose the composites were conditioned according to DIN EN ISO 139. 1 g of a composite from the parallel section at the position of approximately 1/3 of the length of a tensile test specimen (type 1 A, DIN EN ISO 527-2) was weighed exactly in a Duran® filter crucible with a porosity of 3 and a volume of 30 ml (Carl Roth GmbH & Co. KG, Karlsruhe, DE) with a scale (type Kern ABT 120-5 DM, d = 0.00001 g; Kern und Sohn GmbH, Balingen, DE). Crucibles were boiled for 15 min in xylene (97%, Carl Roth GmbH & Co. KG, Karlsruhe, DE) and filtered with suction using a vacuum pump and a filter flask. The procedure was repeated four times. Then the crucibles were dried for 1.5 h at 105 °C, conditioned and weighed a second time.

Fibre length. The measurement of the fibre length was performed with the image analysis software Fibreshape 5.11 (IST AG, Vilters, CH). Fibres were prepared on a slide frame (40 × 40 mm^2, glass thickness 2 mm, company Gepe, Zug, CH) and scanned (4000 dpi resolution for lyocell 1.3 and 3.3 dtex, 2400 dpi resolution for lyocell 6.7 dtex and 2000 dpi resolution for lyocell 12.0 and 15.0 dtex). Only short fibres up to a length of approx. 8 mm extracted from IM composites were analysed. Intersecting fibres were not considered for the evaluation. 500–1700 fibres of each sample were analysed.

2.3. Composite production

Production of semi-finished products. Multilayer webs were produced with a laboratory carding machine with a working width of 200 mm (Shirley Developments Limited SDL, Stockport, UK, serial number 02895). Reinforcing fibres and PLA fibres were mixed during the carding process at environmental conditions (approx. 25 °C, 45% relative humidity) and were oriented predominantly in length direction to the production direction. The initial mass of the lyocell fibres was calculated based on the dry mass for a fibre loading of 20, 30 and 40 mass%. As reference samples multilayer webs were produced from 100% PLA fibres.

Compression moulding (CM). Multilayer webs were dried for 2 h at 105 °C in a forced air oven (Thermicon®, Heraeus GmbH, Hanau, DE). Then, the multilayer webs were CM between two Teflon sheets. At first a hot CM step was carried out at 180 °C for 5 min in a hot press (type HP-S10, Joos, Pfalzengrafenweiler, DE). Afterwards, the plates were CM in a cold press (type HP-S60, Joos, Pfalzengrafenweiler, DE) at 25 °C for 3 min to reduce the processing time. The adequate pressure for each fibre loading was determined in previous experiments. A too low pressure results in poor compaction of the composites, an excessively high pressure to flow processes and to a change in fibre orientation. Therefore it is necessary to adjust the pressure of each fibre loading individually. The pressure was set to 5 MPa for the PLA reference sample and composites with a fibre loading of 20 mass%, 12 MPa for composites with a fibre loading of 30 mass% and 17.5 MPa for composites with a fibre loading of 40 mass%. For the different composite tests, specimens were prepared from composite boards.

Injection moulding (IM). For the IM samples multilayer webs and composite boards were produced in the same manner as described above (differing: hot pressing: 3 min). Granules of a size of approximately 4 × 8 × 2 mm^3 were produced from these composite boards with a shredder (type EBA 2326C, Krug & Priester GmbH & Co. KG, Balingen, DE, compare [15]. The fibre length is limited to 8 mm. Granules were predried in a forced air oven for 2 h at 105 °C (type 2111520000200, Windaus-Labortechnik GmbH & Co. KG, Tullingen, DE). IM was carried out with an Arburg 320C Allrounder 600 – 250 IM machine (Arburg GmbH + Co KG, Loßburg, DE; Screw: ø 30 mm, circumferential speed 20 m/min; die diameter: 5 mm) at a temperature profile of 170 °C – 172 °C – 175 °C – 177 °C – 180 °C. The injector was heated to 180 °C and the tool was cooled to 25 °C. The injection pressure was set to 600 bar, the back pressure to 10 bar and the hold pressure to 480 bar, respectively. The cooling time was 30 s. The tool was in form of a standard test specimen (type 1 A according to DIN EN ISO 527-2).

2.4. Characterisation of matrix and composites

Before composite characterisation test specimens were conditioned according to DIN EN ISO 291 at 23 °C and 50% relative humidity for at least 18 h.

Tensile characteristics. 5–6 test specimens (type 1A, DIN EN ISO 527-2; thickness of CM samples: 2 mm; thickness of IM samples: 4 mm) were tested with a universal testing machine type Zwick Z 020 (Zwick/Roell, Ulm, DE) working with a load cell of 20 kN and a pneumatic clamping system (clamping pressure: 1–2 bar). The gauge length was fixed to 100 mm. A preload of 50 N was used and the test was performed with a speed of 2 mm/min.

Impact characteristics. Unnotched Charpy impact strength was determined for 5–6 test specimens with the dimensions of 80 × 10 mm^2 (thickness of CM samples: 2 mm; thickness of IM samples: 4 mm) with a pendulum impact testing machine (type 5101, Zwick, Ulm, DE) operating with a pendulum hammer of 2 J according to DIN EN ISO 179. The bearing distance was set to 40 mm. The sample was hit on the wide side.

The results of the CM and IM samples are not directly comparable due to the different sample thickness which affects the impact strength. The impact strength increases up to a certain thickness. When a critical specimen thickness is exceeded, the impact