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X-ray micro-computed tomography investigation of fibre length degradation during the processing steps of short-fibre composites

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ABSTRACT

The mechanical properties of composites in the fibre direction are mainly attributed to the fibre slenderness, or aspect ratio. A trade-off between performance and processability is usually required, and dependent on the intended application. If the fibre length could be retained or not severely degraded during various processing steps towards the injection-moulded component, a stiffer and stronger composite product could be obtained. The processing steps for injection moulded wood-fibre composites here include: pulping, commingling, extrusion, pelletizing, and injection moulding. To tune the processing parameters systematically for retained fibre length, it would be useful to investigate the degradation of the original fibre length distribution throughout the processing chain. The fibre length degradation has been monitored by X-ray micro-computed tomography through the processing steps in wood pulp-fibre reinforced polylactide. A significant fibre-length degradation was found. In particular, the extrusion step was found to result in a drastic fibre length reduction.

1. Introduction

Wood pulp fibres are readily available as a renewable reinforcement for composite materials. The fibre length (individual tracheid cell length) is about 1–4 mm for softwood fibres (see e.g. [1–3]). The wood fibres are still sufficiently short to be used in melt processing of thermoplastic-matrix composites, and result in considerable less wear of tools and dies than e.g. short glass fibres. With a diameter of about 25 μm, the relatively high aspect ratio makes these fibres interesting from a reinforcement perspective compared to wood flour, where the reinforcing particles have much lower aspect ratio [4]. However, during the manufacturing processes, the fibres are worn down to very small aspect ratios; sometimes even to particle size. With a particle-like geometry the wood fibres are acting as a filler rather than as a reinforcement. Strength could even be lower than that of the neat matrix if the cellulose fibre segments are too short [5]. For sufficiently long fibre segments, i.e. a couple of times longer than the ineffective length of stress-build up at the end of an oriented embedded fibre, the desired reinforcement can be achieved. In a complicated melt processing procedure, there are several steps that individually contribute to the degradation of the initial fibre length, and hence also to the inferior mechanical properties, as compared e.g. with resin infusion methods, where the fibres in the fibre mat are basically intact after impregnation and curing. In this study, we focus on the manufacturing of injection-moulded wood pulp-fibre reinforced polylactide (PLA), i.e. an entirely renewable and biodegradable composite material. The processing steps for these injection-moulded wood-fibre composites are pulping, commingling, extrusion, pelletizing and injection moulding. The fibre length distribution is quantified step-by-step along the processing line. If the step which results in the most severe fibre degradation could be identified, measures could be taken to mitigate and treat the fibres more gently at this step to retain some of its slenderness. Subsequently, this procedure could be repeated to identify a stage further downstream that is liable for the most serious fibre degradation in the updated processing chain. Of course, there is a balance between processability and mechanical performance, and some fibre length reduction must be acceptable in order to keep the high production rate of injection-moulded composite products leading to low manufacturing costs. The quantification of fibre length distributions in composites has traditionally been done by chemically dissolving the matrix and studying a dilute fibre suspension by optical microscopy, see e.g. [6]. The fibre length distribution can then be quantified either manually by measuring the fibre lengths one by one, or by using two-dimensional image analysis. The dissolution

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and optical microscopy procedures are direct, but rather time consuming. Fibre lengths in water suspensions could be more swiftly characterised by on-line systems where the projected dimensions of individual fibres are measured digitally as they flow in a controlled manner, one by one, through a capillary tube. The Innventia FiberMaster [7,8] and Kajaani Fiberlab [9] are examples of such equipment. X-ray microcomputed tomography (XμCT) is a non-destructive method to assess the three-dimensional structure of a sample from two-dimensional X-ray projection images. After computational processing of the projection images, a three-dimensional array of X-ray attenuation coefficients is obtained. For many materials, especially non-diffractive ones, the attenuation coefficient correlates with local density of the material. Thus, the method essentially produces a density map of the sample. Recent development of XμCT methods has conveyed the composite research community with a non-destructive technique capable of characterising the geometry of fibres inside the composite material sample. With the aid of 3D image analysis, it is possible to quantify fibre length, diameter and orientation distributions from the reconstructed 3D images, as shown by Luengo Hendriks [10], Miettinen et al. [11,12] and Joffre et al. [13]. Some interesting previous studies on this general topic include the work by Alemdar et al. [14], who showed that XμCT can be used to quantify the dimensions of wood and flax fibre in polypropylene matrix. In that case the Image-Pro Plus software was used to identify the Weibull parameters of the size distributions of the fibres.

2. Experimental procedures

2.1. Materials

The natural fibres used for making the composites were bleached sulphite pulp fibres extracted from Norway spruce (*Picea Abies*) delivered from Innventia AB. The matrix material was polylactic acid (PLA). PLA is thermoplastic aliphatic polyester manufactured from starch-rich renewable resources such as maize, sugar beets or wheat. For a biopolymer, PLA has decent physical and mechanical properties, making it a candidate for substitution of certain petrochemical thermoplastics. Due to the good adhesion to cellulose fibres, it has been proven to be suitable to use PLA as matrix material in pulp-fibre composites [15,16]. The PLA fibres used here were PLA 01 quality delivered from Unitika (1.7 dtex, 1 mm length). The polymer fibres were first run through a pulp screen (Wennbergsil) and dried at room temperature in order to facilitate filament release.

2.2. Manufacturing

Commingled wood fibre/PLA fibre mats were manufactured using a laboratory-scale dynamic-sheet former. The polymer and wood fibres were evenly distributed, limiting the fibres to aggregate into larger bundles, which has been observed to be the case when PLA pellets and pulp fibres are introduced directly in the extruder [17]. The extrusion experiments were run on a 40 mm co-rotating twin screw extruder, Werner & Pfleiderer ZSK40. The material was run through the extruder either once or twice, as shown in Fig. 1. Multiple extrusion runs will reduce porosity and improve compounding and surface finish, but most likely reduce the average fibre length and thus impair mechanical properties. The extrudates and pellets obtained after two extrusions were less rough and thus easier to process (see Fig. 1). The extrudates were cooled in water and led to a chopper to produce pellets. After pelletizing the material was dried in an oven at 50 °C for several hours. Extrudates were also broken into smaller pieces using a hammer mill. The weight fraction of wood fibres, w_f, was kept constant at 30%. The weight fraction is more convenient to measure during the manufacturing, whereas the volume fraction, V_f, is used as input in micromechanical models and can be tracked by binarization of XμCT images. The volume fraction can be estimated from measurable quantities, using the relation

$$V_f = \frac{w_f \rho_f}{w_f \rho_f + w_m \rho_m} \tag{1}$$

where ρ_f is the density of the fibres, 1.5 g/cm^3 [18], and ρ_m is the density of the PLA, 1.25 g/cm^3 [19]. Thus the volume fraction of fibre introduced here is 26.3%. After drying the pellets were fed into the injection moulding machine equipped with a multiple die containing mould profiles for dogbone shaped specimens. At each step of the process, samples have been taken off for XμCT analyses.

2.3. X-ray computed microtomography

To prepare the composite samples for the XμCT imaging, a rotary tool was used to carve the sample into a cylinder with radius of approximately 1 mm. The cylinder was then glued to the top of a carbon fibre tube that served as a sample holder. To scan pulp fibres and commingled fibres, no specific sample preparation was needed; a bundle of fibres was simply glued on top of a glass tube acting as a sample holder. The XμCT imaging was done using Xradia MicroCT-400 tomograph. For composite materials a resolution of approximately 1 μm was chosen, resulting in field of view of approximately 1 mm^3. Pulp and commingled fibres were longer, so field of view of approximately 8 mm^3 was used with 2 μm resolution. A typical cross-sectional slice through a sample is visualised in Fig. 2a and some three-dimensional visualisations are drawn in Fig. 4. For measuring the void content, it was necessary to use large samples to have sufficiently large and representative volumes. In this case, entire pellets or machined pieces of the injection-moulded sample were scanned using a Skyscan 1172 tomograph. The larger samples resulted in lower resolution, about 3 μm, which is still sufficiently high to detect the vast majority of the typical voids.

2.4. Image analysis

To separate the fibres from the matrix in the three-dimensional images, a variance weighted mean filter was first applied in order to attenuate imaging noise. The images were then thresholded. Objects having volume smaller than 5000 voxels, corresponding to a sphere whose radius is less than 10 pixels, were considered as noise and removed. A typical result of these operations is presented in Fig. 2. The determination of fibre length and cross-sectional area was done as in [11,12]. The method is based on estimation of the local orientation of the fibres that facilitates extraction of cross-sectional slices. The cross-sectional area of fibres is determined from the slices. The fibre length is estimated using the constrained path-opening method as in [10,11], whose results are then correlated to the cross-sectional properties. The fibres are sampled statistically such that the results are expressed as length and cross-sectional area distributions of fibre volume. A brief description of this method is given in Appendix A.

3. Results and discussion

Softwood tracheids from species like Norwegian spruce and Scots pine have a native length of up to 4 mm [1]. These fibres are inevitably shortened during each of the following processing steps towards the wood-fibre composite. The wood is chipped, pulp cooked, commingled with the thermoplastic fibres, extruded once or twice, pelletized, and finally injection moulded to form a