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Characterization of interfacial stress transfer ability in acetylation-treated wood fibre composites using X-ray microtomography

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ABSTRACT

The properties of the fibre/matrix interface contribute to stiffness, strength and fracture behaviour of fibre-reinforced composites. In cellulosic composites, the limited affinity between the hydrophilic fibres and the hydrophobic thermoplastic matrix remains a challenge, and the reinforcing capability of the fibres is hence not fully utilized. A direct characterisation of the stress transfer ability through pull-out tests on single fibres is extremely cumbersome due to the small dimension of the wood fibres. Here a novel approach is proposed: the length distribution of the fibres sticking out of the matrix at the fracture surface is approximated using X-ray microtomography and is used as an estimate of the adhesion between the fibres and the matrix. When a crack grows in the material, the fibres will either break or be pulled-out of the matrix depending on their adhesion to the matrix: good adhesion between the fibres and the matrix should result in more fibre breakage and less pull-out of the fibres than poor adhesion. The effect of acetylation on the adhesion between the wood fibres and the PLA matrix was evaluated at different moisture contents using the proposed method. By using an acetylation treatment of the fibres it was possible to improve the strength of the composite samples soaked in the water by more than 30%.

1. Introduction

Wood fibres offer several potential advantages over synthetic fibres, such as glass or carbon fibres, when used as reinforcement of composite materials: they are light, from a renewable resource, and already available at relatively low cost and in industrial quantities (Almgren et al., 2009b; Huda et al., 2006a). Injection-moulded short-fibre composites are used in high-volume applications with short processing cycles and lower demands on the load carrying capacity. However, if the mechanical properties could be improved, injection-moulded composites could be used in mechanically more demanding applications. It is well known that an Achilles' heel of wood fibre composites is the poor affinity between the hydrophilic fibres and the hydrophobic polymeric matrices such as polypropylene (PP) or polyethylene (PE), resulting in both agglomeration of the fibres (Balasuriya et al., 2001; Joffre et al. 2014b) and lower stress transfer ability at the fibre-matrix interface (Almgren, 2010; Bledzki et al., 2005). Both phenomena drastically decrease the mechanical performance of the material. To improve the affinity between the matrix and the fibres, the use of less hydrophobic polymeric matrices, such as polylactic acid (PLA) has been found to result in better mechanical performances (Huda et al., 2006b; Mamun and Bledzki, 2013). However, due to the reduced hydrophobicity of the PLA, moisture is able to diffuse into the material resulting in dimensional changes of the reinforcing fibres (Almgren et al., 2009a). Even though the hygroexpansion of cellulosic fibres has also been used as inspiration of prototypes for moisture-controlled devices when developing biomimetic systems for mechanosensing and actuation (Burgert and Fratzl, 2009; Le Duigou and Castro, 2015; Le Duigou et al., 2016), dimensional instability is usually regarded as an inconvenience when designing engineering materials: swelling and shrinkage of the fibres inside the composite results in dimensional changes, induces stresses and

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loss of mechanical performance (Almgren et al., 2009a). Moreover due to the helical structure of wood fibres any dimensional variation induced by a moisture change is coupled with a twist of the fibre (Burgert et al., 2007, 2005; Joffre et al., 2016), which in a composite might lead to debonding and consequently lower the stress transfer ability of the fibre matrix interface. Hence, a trade-off is needed: using a completely hydrophobic matrix will protect the fibres from the moisture but will result in a low stress transfer and difficulties in blending matrix and fibres, whereas a less hydrophobic matrix will result in a better stress transfer but the mechanical performance will be inevitably dependent on the moisture uptake of the sample.

To overcome these issues different strategies have been developed during the last twenty years. One path consists in modifying the fibres to improve their adhesion to polymeric matrices (Dányádi et al., 2010; Nenkova et al., 2006; Schirp et al., 2014) and/or to add a compatiblilizer to the mixture aiming to improve the stress transfer from the matrix to the fibres (Hu and Guo, 2015; Karmarkar et al., 2007; Marais et al., 2012; Ndlovu et al., 2013; Sudár et al., 2016). However, there is a need to better understand the mechanisms behind the change in mechanical properties due to these modifications, and particularly to quantify the stress transfer ability at the fibre/matrix interface resulting from these modifications. This quantification is difficult mainly due to the small dimension of the fibres, making direct pull-out tests extremely cumbersome as pointed out by (Desarmot and Favre, 1991; Favre and Merienne, 1981). It is necessary to develop more accessible methods.

In this study, wood pulp fibres were acetylation-treated and mixed with PLA aiming to produce 100% renewable composites with improved mechanical performance in the presence of water. It has already been shown that bodification by acetylation has a positive effect on the equilibrium moisture content (EMC) of wood and lignin (Gordobil et al., 2015): acetylated fibres will absorb less moisture in the wet state (Tserki et al., 2005) which should limit the swelling of the fibres and hence possibly improve the dimensional stability and the mechanical performance of the composite. To confirm this assumption, tensile test and notch sensitivity tests are here performed on wood fibre and plastic composites manufactured from acetylated and unmodified fibres at different relative humidities (RH) and using different ratios of treated/untreated fibres. The fracture surfaces were analysed with X-ray microtomography (X-μCT) and the length of the fibres sticking out of the matrix are estimated using automatic digital image processing. The underlying assumption is that a good stress transfer at the matrix-fibre interface will result in fibre breakage whereas a poor bonding will result in a pull-out of the fibre. Thus, for a given manufacturing process, the length of the fibre segments sticking out of the matrix should be an estimate of the stress transfer ability of the composite. Based on these measurements the effect of the acetylation treatment on the mechanical properties of the composite was investigated at different levels of relative humidity.

2. Experimental procedures

2.1. Materials and manufacturing

Softwood CTMP fibres (CS770, Rottneros, Sweden) were used in the wood-fibre plastic composites. The acetylation of the fibres was performed in 50 g batches, the fibres were oven dried and then boiled in acetic anhydride in a 1-L glass reactor for 4 h. The treated fibres were washed and oven dried, and its weight gain percentage (WPG) was calculated. The average WPG of the eight performed batches was 23%. All the acetylated batches were mixed together prior to the composite manufacturing. Unmodified and acetylated wood plastic composites were compounded with PLA (NatureWorks Ingeo 3251D) on a co-rotating twin screw extruder (Coperion ZSK 26 K, 10.6, Coperion, Germany) and granulated. The granules were then injection moulded (Engel ES200/110 HL-V) into plates measuring $1 \times 100 \times 100\ mm^3$. The fibre/polymer ratio was held constant at 20/80 for all the materials. 5 different samples were realized by changing the ratio between modified/unmodified fibres from 0 to 1 by steps of 0.2.

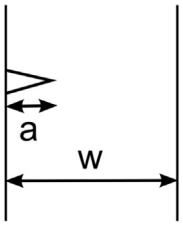

Fig. 1. Crack cut in the sample (a – length of crack, w – width of the specimen).

2.2. Mechanical testing

Rectangular samples of 15 mm width were cut using a band saw. Notches were made in some samples using a razor blade. The razor blade had a mark of the desired depth in order to control the insertion and thus the length of the crack (see Fig. 1). Two different crack length ratios (a/w) were cut: 0.1 and 0.3. Prior to the test the samples were stored in three relative humidity (RH) conditions: 22% RH, 65% RH, or fully soaked in water. The sample were conditioned during two weeks in order to ensure equilibrium conditions. The temperature was kept constant at 23 °C (±2 °C) during the conditioning. The samples were tested just after being removed from the controlled environment. The mechanical tests were performed using a Shimadzu AGs-X at a cross head speed of 5 mm/min. Each measurement was repeated 4 times. Samples breaking in the clamps were discarded and the experiment was repeated. All samples were cut in the injection direction. Since the fibre orientation distrubution might vary within a plate, for each experiment the samples were taken from all the different parts of a single plate aiming to limit any possible bias related to a local variation of fibre orientation.

2.3. X-ray microcomputed tomography

In order to acquire information about the fracture surface X-μCT scans were performed using a Bruker Skyscan 1172 at constant room temperature of 23 °C (±2 °C) and a relative humidity of 30% (±2%). 1200 projections were acquired and the resulting images had 2 000 × 2 664 pixels of size 2.1 μm. 3D images with voxel size 2.1 μm were reconstructed from the images using NRecon 1.6.9 (Bruker, Belgium). A resolution of about 2.1 μm was sufficient to estimate the length of the fibre even if the fibre broke during the mechanical test, since a small part of the fibre was always visible outside the matrix. However the resolution was not sufficient to directly assess the interface, and voids at the fibre matrix interface smaller than 2 μm were not detected.

2.4. Image analysis

The steps involved in the estimation of the fibre length are illustrated in Fig. 2. The process to estimate the fibre length of the fibres sticking out the matrix interface consists of the following steps: (a) A ring artefact reduction algorithm is applied to the reconstructed images (Wernersson et al., 2013) and the images are automatically thresholded using the chord method (Zack et al., 1977). The use of an automatic threshold avoids any manual choice which could interfere with the results, and was thus preferred in this study. (b)