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Research paper

Influence of flax fibre variety and year-to-year variability on composite properties

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ABSTRACT

The use of natural fibres such as flax in composite materials is an expanding market sector. Variability potentially exists at many points in the "plant-composite continuum". In this study, the influence of i) variety, ii) growth year, and iii) processing technique on the mechanical properties of composites was tested. Ten varieties of flax grown in two consecutive years were processed by the following techniques: i) twin-screw extrusion and injection moulding, ii) vacuum assisted resin transfer moulding (VARTM) and iii) pultrusion. These processes differed in the fibre length, fibre orientation and polymer used, and so differed overall in the severity of the fibre treatments.

The two repetitions of growing in two subsequent years showed a significant influence on the morphology of the raw fibre bundles and the composite properties processed with VARTM and pultrusion technique. The year effect was overlapped by the severe processing influence using twin-screw extrusion and injection moulding, where no influence of the growing year was detectable in the mechanical properties of the composites and the morphology of the fibre bundles extracted from the composites.

Although the use of different processes on the same plant material causes large differences in the mechanical properties of the composites, we could identify plant varieties that showed consistently above average and below average performance with all processing techniques and over two years. This knowledge can be used as a basis for further breeding of flax varieties optimised for composite reinforcement.

1. Introduction

Natural fibre-reinforced composites (NFC) are gaining importance in various industrial sectors. The market share for NFC in the European Union has reached 4 % of total composite production (glass, carbon, WPC, NFC) and a volume of 92 000 t (Carus et al., 2015). A recent study by Carus et al. (2015) projected strong growth, with production in 2020 to reach 130 000 t (without incentives for bio-based products) or >370 000 t (with strong incentives for bio-based products) in 2020 (Carus, 2015). The automotive industry is currently the most important market for NFCs by far, with 90 000 t consumed in 2012. Outside of the automotive sector, many other

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applications of NFCs have recently been developed, ranging from flower pots or decking components, to sporting equipment (Müssig and Haag, 2015; Pil et al., 2016). Reasons for using NFCs include their high-performance specific (density related) mechanical properties, environmental and ecological benefits and (depending on the processing and fibre quality used) their low cost (e.g. Bourmaud et al., 2013; Graupner and Müssig, 2010; Placet et al., 2012; Virk et al., 2012).

The breadth of applications of NFCs already in use demonstrates that NFCs can be produced to have a large range of mechanical properties: on the one hand fibres are rather being used as filling material than as reinforcement, on the other hand the mechanical properties can reach the demands of (semi-)structural parts. These properties depend largely on the processing technique, the fibres and the polymers that are used. In this work we focus on the influence of the flax fibre variability in the composite. Several factors that contribute to variability in fibre performance are well-described in the literature and are summarized here:

* Fibre length: the reinforcement efficiency of fibres in the composite strongly depends on the ratio between their length and the critical fibre length that is necessary for effective load transfer from the matrix to the fibre. Depending on the interface properties between the fibre and the polymer, the critical fibre length can range from below 1 mm (at high adhesion) to several millimetres (see e.g. Graupner et al., 2014). In injection moulded composites the length of the fibres is close to or below the critical fibre length whereas in processes like Vacuum assisted Resin Transfer Moulding (VARTM) or Pultrusion the fibre length is significantly higher than the critical fibre length.
* Fibre orientation: classical lamination theory describes generally the dependency of composite properties on the orientation of the fibres (see e.g. Schürmann, 2005). Madsen et al. (2007) validated the influence of the loading direction on the stress-strain curves for UD hemp yarn reinforced composites. The higher the proportion of fibres that is present in the loading direction, the more dominant the fibre behaviour in the composite.
* The morphology and distribution of the fibre: for optimised composite properties, fibres have to be long, of small diameter and homogeneously dispersed without agglomerates. While the length is a key factor for strength and toughness optimisation, the fibre dispersion is relevant for stiffness (Bos et al., 2002). It is common to assume better mechanical properties of the composites if finer fibre is used, as the surface of the fibre is increased and enables more effective load transfer. However, there are exceptions (which can be attributed to overlapping effects like higher agglomeration tendency correlating with fibre fineness), as has been shown e.g. by Graupner et al. (2008) for cotton reinforced epoxy composites.
* Biochemical composition of the plant material: natural fibres themselves can be regarded as fibre-reinforced composites consisting of cell walls that are made up of (partly) crystalline cellulose fibrils embedded in a matrix of hemicelluloses and lignins. The composition as well as the orientation of the cellulose fibrils plays an important role in the mechanical properties of the plant cell (Baley, 2002). The higher the orientation of the cellulose in the cell wall layers (quantified by small Microfibril Angels) the higher the strength and stiffness of the fibre (Eder and Burgert, 2010). The plant varieties as well as the growing and retting conditions are of influence for the composition of the cell wall present in the fibre bundles (Baley et al., 2012; Lefeuvre et al., 2014).
* The convariety type: with the flax crop (*Linum usitatissimum* L.) the two main goals are fibre (mainly for textile purpose) and oil seed production, respectively (Akin, 2013). By plant breeding, different varieties were optimised for the different performance goals. Scheer-Triebel et al. (2000) determined that fibre bundles from fibre flax have higher Young's moduli than fibre bundles from oil flax varieties. They further correlate this finding with fibre bundle size and lignin content. The best fibre quality could be found in fibre varieties with small lumina and rather circular cells (Scheer-Triebel et al., 2000).
* Growing conditions: the development of the stem depends on meteorological and soil conditions that may cause variation between years or between geographical locations. The morphological differences influence the mechanical properties, so it is of interest to maximize uniformity and reproducibility of fibre morphology and mechanical properties (Pillin et al., 2011). During growth, wind and water stress can lead to the formation of dislocations that can significantly weaken the strength and stiffness performance of fibre cells (Hänninen et al., 2012; Hernandez-Estrada et al., 2016; Hughes, 2012; Norton et al., 2006) and influence the processability and the mechanical behaviour of composite structures (Hughes, 2012).

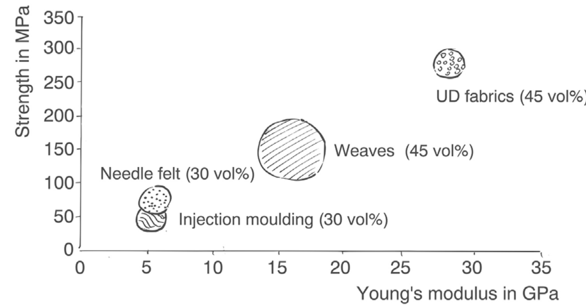

Fig. 1. Performance playground of flax composites.
Adapted from Verpoest and Baets (2012) in Müssig and Haag (2015).

The factors listed above are not independent. For example, fibre length and orientation are strongly influenced by the processing technique that is used to obtain the NFC. Verpoest and Baets (2012) developed the "flax playground" to show the range of strength and stiffness that can be achieved by different flax composites (see Fig. 1). The potential range of mechanical properties is shown in Fig. 1, but, up to now there is a lack of knowledge on the influence of the variety: the information of the suitability of different flax varieties for composite applications and their breeding potential for fibre reinforced composites processed with different methods has to the authors' best knowledge not yet been analysed. This information can be important to design new flax varieties by plant breeding for the growing market sector of flax fibre-reinforced composite materials.

Traditional composite manufacturing processes developed for non-plant based material can be adapted for natural fibres. This has been shown for example by Beaugrand and Berzin (2013) and Berzin et al. (2014): an extrusion process that is adapted to natural fibres compared to an unmodified process could improve the mechanical properties (stress at break as well as Young's modulus) of the composite by more than 30 %. At contrary, contrasting properties of the reinforcing elements not necessarily displayed functional contrasts in composites using the woody hemp core as reinforcement: elements characterized according to multiple criteria (e.g. structure, composition, mechanical properties, . . .) with theoretically different potential for reinforcement of composites (Beaugrand et al., 2014) did not display differences in NFC when processed with a non-optimised extrusion process. The processing by extrusion and injection moulding may have masked or erased some intrinsic particularities of the plant fibres, so that the promising properties of the natural fibres were not fully exploited.