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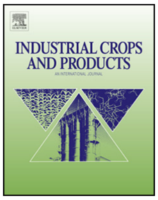

Elementary flax fibre tensile properties: Correlation between stress–strain behaviour and fibre composition

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ABSTRACT

Flax fibres represent an eco-friendly alternative to glass fibres in composite materials. Natural fibres do not have a unique standard response to tensile testing. To increase understanding of the response to tensile testing, six batches of flax fibres (*Linum usitatissimum*) were selected for their differences in tensile properties: four batches consisted of Marylin-variety cultures grown in the same area but in different years, and two were reference-samples from Oliver and Hermes varieties. Their tensile values were either moderate ($E \approx 45$–55 GPa, $\sigma \approx 800$–1000 MPa) or high ($E > 55$ GPa, $\sigma > 1000$ MPa). Three major types of stress–strain behaviour were observed, but in different proportions in each sample. The first one consisted of a linear tensile behaviour; the second one was composed of two linear distinct sections, and the third one displayed a non-linear section at the beginning of the loading stage up to a threshold point, followed by a section where the tangent modulus increased up to failure. The samples exhibiting a large proportion of the third type of behaviour were characterised by high tensile properties. The extent of the non-linear section highly depended on the variety. Within the Marylin variety, the tensile properties were higher when the non-linear section was smaller. Considering the fibre as a composite *per se*, reinforced by cellulose microfibrils coated with hemicelluloses embedded in a matrix of incrusting pectins, we found some correlation between tensile behaviours and the cell-wall composition that highlighted the importance of the hemicelluloses and hemicelluloses/pectins ratio.

1. Introduction

Since 2000, numerous studies have been carried out on the use of plant fibres in composite applications (Biagiotti et al., 2004; Dittenber and GangaRao, 2012; La Mantia and Morreale, 2011; Mohanty et al., 2002). Among plant fibres, flax fibres appears to globally represent an eco-friendly alternative to glass fibres in composites (Joshi et al., 2004). When flax fibres are compared to glass fibres by Life Cycle Assessment, several environmental indicators used such as climate change, acidification or non-renewable energy consumption are in favour of flax fibres. Nevertheless, it has been pointed out that the use of fertilizers and the hackling are two steps which could be optimise as they have an high impact on the eutrophication indicator and the energy consumption (Le Duigou et al., 2011). Depending on the variety (Sharma et al., 1999), the environment, and the agro-industrial practices (Chemikosova et al., 2006; Easson et al., 1994; Norton et al., 2006), they might display good tensile properties close to those of glass fibres, ranging from 46 to 72 GPa in stiffness and from 741 to 1454 MPa in strength (Baley, 2002; Bourmaud et al., 2010; Charlet et al., 2010; Pillin et al., 2011).

At maturity, flax cellulosic fibres are composed of a thin primary cell-wall and a thick secondary cell-wall containing three layers (S1, S2, S3) made essentially of cellulose and a few non-cellulosic polysaccharides (Fig. 1) (Gorshkova and Morvan, 2005; Gorshkova et al., 2003; Hearle, 1963).

Opposite to lignocellulosic fibres, flax fibres contain a few amount, around 2%, of lignin (Day et al., 2005; Love et al., 1994). The S2 layer, which represents up to 80% of the fibre cross-section, is reinforced by longitudinally oriented microfibrils of cellulose at about a 10° angle from the fibre axis, and is mainly responsible for the fibre tensile properties (Bourmaud et al., 2013a,b). Microfibrils were described as strongly interacting with hemicelluloses enriched in glucomannan moieties, and being embedded in

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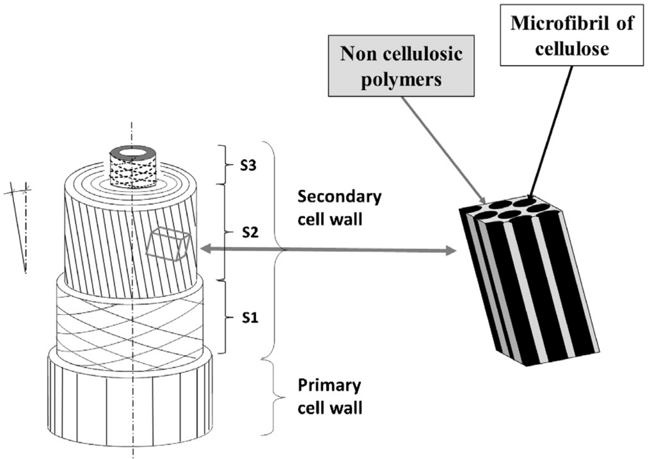

Fig. 1. Schematic representation of a fibre cell-wall organisation.

a matrix of incrusting pectins (Fig. 2) (Bourmaud et al., 2013a,b; Gorshkova and Morvan, 2005; Mcdougall, 1992; Morvan et al., 2003).

Altogether, a flax fibre was assimilated to a complex composite multilayer structure; its responses to tensile stress are governed by interactions and interfaces between the structural cell wall layers. Consequently, the stress–strain curves of cellulosic fibres were not expected to be simple (Baley, 2002).

The stress–strain curves of cellulosic fibres including flax (Andersons et al., 2005; Aslan et al., 2011; Baley, 2002; Charlet et al., 2009) and hemp (Duval et al., 2011; Pickering et al., 2007; Placet et al., 2013) have been found to be quite complex by being nearly elastic or displaying more or less non-linear regions. Part of the complexity comes from the presence of both crystalline and amorphous celluloses (Northolt, 1985). In the case of hemp, cellulosic fibres have been reported to display three types of tensile behaviour in response to tensile testing (Duval et al., 2011; Pickering et al., 2007; Placet et al., 2012). Type I (TI) exhibited a linear relationship similar to that observed for glass fibres. Type II (TII) was non-linear and characterised by two distinct sections with a decreasing slope in the second section. Type III (TIII) showed nonlinearities. In the case of flax, the different types of behaviour described in the literature appear less clear. Andersons et al. (2005), working on elementary fibres that were manually separated from bundles of enzyme-retted flax, have noted for most of the fibres an initial short non-linear region, followed by a main linear domain. Aslan et al. (2011) found that green (non-retted) fibres exhibited a nearly linear TI behaviour while retted cottonized flax fibres showed TI behaviour as well as a complex non-linear TIII stress–strain behaviour. These authors explained the differences by the presence of defects induced by the mechanical treatments during the scutching and cottonization processes. The complex third type behaviour has also been reported for some varieties of dew-retted long fibres such as the Hermes variety (Charlet et al., 2007) or the Ariane variety (Baley, 2002; Baley et al., 2012). The latter authors described a particular form of stress–strain curve, with an increasing tangent modulus up to failure as the strain increased. In the case of the Ariane variety, after drying of the fibres (14 h at 105 °C) with an initial moisture content at 8.3%, the percentage of TIII behaviour increased while the stress and strain at failure significantly decreased (Baley et al., 2012, 2005). From all these data, the impacts of retting and drying treatments or the mechanical stress applied on the stress–strain curves have to be questioned in term of internal structural changes.

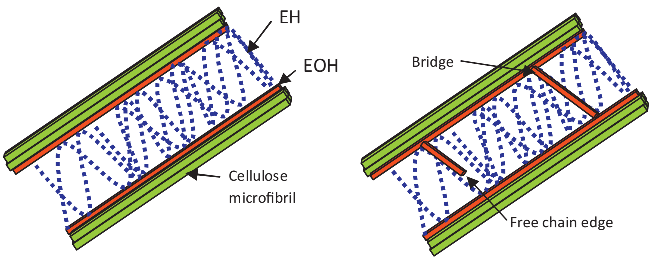

Fig. 2. Schematic representation of the chemical structures in the S2 layer of flax cellulosic fibres. In green: cellulose microfibrils; in blue: matrix of incrusting pectins that were extracted with acid (EH); in red: hemicelluloses, able or not to establish bridges between two microfibrils depending on the amount of pectic matrix that were extracted with alkali (EOH) (Bourmaud et al., 2013a,b). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of the article.)

The main mechanism could be a partial reorientation of cellulose microfibrils along the fibre axis (Aslan et al., 2011; Baley, 2002; Bourmaud et al., 2013a,b). Exceeding a certain level of shear stress might cause the break of hydrogen bonds, and a slippage of the matrix occurs (Burgert and Fratzl, 2009). The removal of shear stress would induce the microfibrils to lock into a new position by the recovery of hydrogen bonds. This phenomenon has been described as a stick-slip mechanism (Fratzl et al., 2004). The fibre stiffening, in the axial direction, might also be attributed to the partial crystallisation of the paracrystalline cellulose and the extension of cellulose microfibrils (Astley and Donald, 2003; Hearle, 1963; Placet et al., 2013). On the other hand, when working on different varieties collected in various areas and over different years, an interesting correlation was found between the average tensile properties of fibres and the content of uronic acids that can be selectively extracted either with alkali (considering as interacting with cellulose microfibrils) or with acids (considering present in the matrix in which the microfibrils are embedded) (Alix et al., 2009; Bourmaud et al., 2013a,b).

In a previous study (Lefeuvre et al., 2013), the impact of drought was studied on the average mechanical tensile properties of eight samples of the same Marylin variety, grown over a period of three years in the same area (Neubourg plateau, Normandy, France). On one hand, no impact of the cultivation year was statistically demonstrated. On the other hand, within each year, there were samples displaying high average tensile properties and others exhibiting moderate properties.

The aim of the present study is to increase understanding of the reason for differences in average tensile properties at rupture. The task was approached by the analysis of the individual stress–strain curves of about 50 fibres for each of the four Marylin variety samples. Two samples, named M1_2009 and M2_2009, were grown in 2009; the two others, named M1_2010 and M2_2010, were grown in 2010 (drought year). The samples designated as M1 were shown to display high average tensile properties ($E > 55$ GPa, $\sigma > 1000$ MPa) while M2 were characterised by moderate mechanical properties ($E \approx 45$–55 GPa, $\sigma \approx 800$–1000 MPa). Two other reference samples were chosen from the literature for their high (Hermes, a textile variety (Charlet et al., 2007)), or moderate (Oliver, an oleaginous winter variety (Alix et al., 2009)) average tensile properties. Interestingly, the difference in the average tensile properties between Hermes and Oliver were partly explained by their differences in cell-wall composition through the proportion of non-cellulosic polysaccharides which encrusted or coated the cellulosic micro/macrofibrils (Alix et al., 2009). In the first part of the results, the average tensile properties were analysed as a function of the stress–strain type; in the second part, the stress–strain type III was particularly investigated in term of deformation and tangent modulus. Finally, the data of the stress–strain curves were discussed regarding some parameters of their cell-wall