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Role of flax cell wall components on the microstructure and transverse mechanical behaviour of flax fabrics reinforced epoxy biocomposites

J. Acera Fernández[a,b], N. Le Moigne[a,*], A.S. Caro-Bretelle[a], R. El Hage[a,1], A. Le Duc[c], M. Lozachmeur[c], P. Bono[c], A. Bergeret[a,**]

[a] *Centre des Matériaux des Mines d'Alès (C2MA), Ecole des Mines d'Alès, 6 Avenue de Clavières, 30319 Alès Cedex, France*[2]
[b] *French Environment and Energy Management Agency (ADEME), 20 Avenue du Grésillé, 49000 Angers, France*
[c] *Fibres Recherche Développement (FRD), Technopole de l'Aube en Champagne, Hôtel des Bureaux 2, 2 rue Gustave Eiffel, CS 90601, 10901 Troyes Cedex 9, France*

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ABSTRACT

Mechanical and chemical processes used in the extraction of flax fibres for the production of technical flax fabrics and other flax products have a significant effect on their biochemical composition, structure and properties. In this work, we investigated the effect of different chemical extraction treatments on the biochemical composition and physical chemical properties of flax fabrics and their influence on the microstructure and mechanical properties of thermo-compressed flax fabrics reinforced epoxy composites. A unidirectional (UD) flax tow woven fabric with minimal processing was chosen in order to retain as much of the original flax cell wall structure as possible. The flax fabric was treated by various aqueous and organic solvents with increasing solvation capacity, so as to gradually extract cell wall components from the fibres. The treated flax fibre fabrics were characterised in terms of biochemical composition, wettability and dimensional characteristics. The influence of chemical extraction treatments and the role of cell wall components on the microstructural and mechanical properties of UD flax/epoxy biocomposites were investigated and discussed by means of Scanning Electron Microscopy (SEM), image analysis, Differential Scanning Calorimetry (DSC) and transverse tensile tests. Our results demonstrate that non-cellulosic cell wall components of flax fibres play a key role in the dispersion of flax yarns within the epoxy matrix, and in the mechanical behaviour of biocomposites.

1. Introduction

Natural fibres are a promising alternative to synthetic fibres in technical textiles and composites applications due to their advantageous specific mechanical properties, their interesting viscoelastic and acoustic damping performances, as well as their lower environmental impact during their production and use phases, and their end of life (Dissanayake et al., 2009; Duflou et al., 2014; Joshi et al., 2004; Le Duigou et al., 2011a,b). Mechanical performances of natural fibre based composites are strongly influenced by the intrinsic mechanical properties of the fibres and matrices used and also by the fibre/matrix interfacial adhesion. Among all the natural fibres, flax fibres are nowadays the most advanced natural technical fibres (Holbery and Houston, 2006; Sliseris et al., 2016; Yan et al., 2014a,b, 2015). They indeed possess high modulus and tensile strength compared to other natural fibres (Baley, 2002; Faruk et al., 2012). They can be used as short fibres in thermoplastics for injection moulding applications, and can also be transformed in structured technical fabrics that are impregnated and processed with thermosets and thermoplastics intended to be used in structural applications.

In contrast to synthetic fibres, usually made of a mono-component in the bulk (glass, carbon, etc.), natural fibres such as flax fibres, present a complex hierarchical and layered structure made of several biopolymers constituting the core structure of the cell walls. As the main component, cellulose macromolecules crystallize in microfibrils which are reasonably oriented along the fibre axis with an angle of roughly 5 to 10° for flax fibres (Bledzki et al., 1996; Bledzki and Gassan, 1999; Dittenber and Gangarao, 2012; Mohanty et al., 2000), and embedded in a matrix of non-

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* Corresponding author. Fax: +33 466785365.
** Corresponding author.
E-mail addresses: nicolas.le-moigne@mines-ales.fr (N. Le Moigne), anne.bergeret@mines-ales.fr (A. Bergeret).
[1] Current address: Laboratoire de Chimie-physique des matériaux (LCPM), Université Libanaise, Faculté des Sciences II, Fanar, Lebanon.
[2] C2MA is member of the European Polysaccharide Network of Excellence (EPNOE), www.epnoe.eu.

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cellulosic components such as hemicellulose, lignin, pectins and proteins. In general, the cell walls in higher plants are made of an outer layer, the primary (P) wall and successively deposited concentric inner layers constituting the secondary (S1) and (S2) walls, in which the different biopolymers are distributed and organised, thus forming a multi-component and tri-dimensional fibrillar structure (Klemm et al., 2005, 1998; Krässig, 1993). These fibrillar cells, also called elementary fibres, are usually gathered in fibre bundles, also called technical fibres, within the stems of higher plants. A combined method using visible photomicrograph, associated with a 3-D mid-infrared transmission, allowed localizing the distribution of the different components across flax stem sections (Morvan et al., 2003). The authors observed that flax stem surfaces in the epidermal region are mainly composed of pectins and lipophilic components, such as waxes. Underneath the epidermis, the internal zone rich in bast fibres, where cellulose is predominant, showed substantial amounts of pectins and some acetylated non-cellulosic polysaccharides, whereas lignin and non-cellulosic polysaccharides are more concentrated in the inner core tissues of the stem. These observations showed that, in the case of flax fibres, cohesion of the bundles in bast fibres is not insured by lignin because it is present in very low amount. On the other hand, pectins are the major components accumulated in the primary cell wall and cell junctions, i.e. middle lamellae, and act as adhesives that ensure the cohesion of flax fibre bundles. Furthermore, the content and the nature of fibre surface components depend on the natural fibre considered. Marques et al. (2010) have analysed by gas chromatography the lipophilic extractives of four natural fibres (flax, hemp, sisal and abaca) and identified several components especially fatty alcohols and acids, aldehydes and ester waxes, in the case of flax fibres. Considering this complex multi-component microstructure, several kinds of interfaces should thus be considered when incorporating flax fibres in a polymer matrix: (i) the interface between the individualised elementary fibres and the polymer matrix, (ii) and the interface in between the elementary fibres within the fibre bundles whose cohesion is mainly ensured by pectic cements.

The production of flax fabrics from bast fibres in textile and composite applications requires several transformation steps involving mechanical processes, such as scutching and hackling, as well as chemical treatments such as desizing, scouring, alkali extraction, kiering or bleaching. Their main objectives are to remove shives, separate and refine fibre bundles and clean it by removing impurities such as waxes and proteins to obtain yarns that are further arranged to form woven or non-woven fabrics (Lacasse and Baumann, 2004; Müssig and Hughes, 2012). Coroller et al. (2013) investigated the microstructure and mechanical properties of unidirectional flax/epoxy composites. The authors used three varieties of flax fibres, i.e. Hermes, Andrea and Marylin, and found that the mechanical extraction process, especially the hackling step, has a strong influence on the fibre dispersion within the matrix. A significant improvement of the longitudinal tensile strength of the composite was obtained with hackled fibres due to their greater ability of individualization into elementary fibres that produces a more homogeneous composites microstructure. Bourmaud et al. (2010) investigated the effect of two “soft” water-washing treatments (72 h at 23 °C and 1 h at 100 °C) on technical flax fibres, and analysed sugar loss by thermogravimetric analysis (TGA) and the resulting separation and tensile properties of the fibres. They found that water-washing treatment removed non-cellulosic polysaccharides from the middle-lamellae and observed a much more drastic extraction of cell wall components at 100 °C during 1 h, because of the hydrolysis of pectin chains from the walls. The authors also noticed a decrease in fibre diameter scattering and a slight increase of average tensile properties for the treatment at 23 °C for 72 h. Stronger washings by alkali treatments were used by several authors to improve the mechanical properties of flax fibres reinforced epoxy composites. Van de Weyenberg et al. (2006) treated flax slivers with NaOH solutions at 1, 2 and 3% for 20 min at room temperature prior to their alignment, stacking with adhesive films of epoxy resin and autoclaving. The authors observed a slight increase in the longitudinal flexural strength and modulus of the composites with the increment of NaOH concentration. Transverse flexural strength and modulus were also improved by the treatments, especially with a NaOH solution at 1%. This was explained by the removal of impurities and waxes and the creation of a rougher fibre surface which should favour the mechanical interlocking and the chemical bonding at the fibre/matrix interface. Besides, a decrease of the fibre strength was reported after alkalisation, supporting the hypothesis that the increase in composites mechanical properties was mainly due to a significant improvement of interface quality. Yan et al. (2012) carried out an alkali treatment, with 5% w/w at 20 °C for 30 min, on flax, linen and bamboo woven fabrics to improve the mechanical properties of epoxy based composites manufactured by a vacuum bagging technique. The authors found a negative effect of the alkali treatment on the tensile strength and modulus of flax, linen and bamboo single-strand yarns extracted from the fabrics. On the other hand, an increase in longitudinal tensile and flexural properties was measured for all treated composites. These results supported that alkali treatments degrades the fibre properties but can significantly improve the fibre/matrix adhesion, and hence the composites mechanical performances. The different mechanical and chemical processes used for the extraction of flax fibres in the production of flax fabrics and other products thus have a significant effect on the biochemical composition, structure and properties of flax fibres, and hence should strongly influence the resulting microstructural and mechanical properties of biocomposites.

The aim of the present work is to investigate the effect of different chemical extraction treatments on the biochemical composition and physical chemical properties of flax fabrics and their influence on the microstructure and mechanical properties of flax fabrics reinforced epoxy composites. In order to keep as much as possible the original flax cell wall structure, a unidirectional (UD) flax tow woven fabrics that has undergone minimal processing, was selected. It was treated by various aqueous and organic solvents with increasing solvation capacity, so as to extract gradually cell wall components from the fibres. Untreated and treated flax fabrics were then processed by thermo-compression with a low viscosity epoxy resin to obtain UD flax/epoxy biocomposites. The treated flax fibre fabrics are characterised in terms of biochemical composition, wettability and dimensional characteristics. The influence of chemical extraction treatments and the role of cell wall components on the microstructural and transverse mechanical properties of UD flax/epoxy biocomposites are investigated and discussed.

2. Material and experimental methods

2.1. Flax fabrics, epoxy resin and chemicals

UD flax tow woven fabrics (FRD-UD41) with an areal density of 218 g/m^2 was provided by Fibre Recherche Développement (FRD, France). These flax woven fabrics (Fig. 1) are weaved in weft direction with flax tows, which are spinned from retted and hackled fibres. Flax tows were not washed, treated or oiled for the production of woven fabrics in order to minimise the chemical and mechanical degradation of fibre cell walls and/or the addition of chemical products. The dry linear density and insertion density of flax yarns within the untreated fabrics are 88.8 ± 5.8 tex and 19 picks/cm, and 25.2 ± 0.8 tex and 12.8 end/cm in the weft and warp direction, respectively. Accordingly, the relative amounts of flax yarns in the weft and warp direction for the untreated fabrics