COMPRESSION MOULDING OF FLAX FIBRE REINFORCED COMPOSITE MATERIALS

P.J.J Dumont[1], L. Orgéas[2], M. Hubert[2], B. Vermeulen[3], P. Vroman[3], S. Rolland du Roscoat[2], J.-F. Bloch[1]

[1] *Laboratoire de Génie des Procédés Papetiers (LGP2), CNRS / Institut polytechnique de Grenoble (Grenoble INP), BP 65, 38402 Saint-Martin-d'Hères cedex, France:* pierre.dumont@grenoble-inp.fr

[2] *Laboratoire Sols-Solides-Structures-Risques (3S-R), CNRS / Université de Grenoble (UJF & Grenoble INP), BP 53, 38041Grenoble cedex 9, France.*

[3] *École Nationale Supérieure des Arts et Industries Textiles (ENSAIT), 2 allée Louise et Victor Champier, BP 30329, 59056 Roubaix cedex 1, France.*

ABSTRACT: The rheological properties of flax fibre reinforced composites processed by compression moulding are investigated in this work. For that purpose, a model composite made up of needled flax fibre mats impregnated by a viscous paraffin gel was used. Lubricated simple compression tests were performed using a large size rheometer. Results reveal that two flow regimes can occur depending on the test conditions: a relevant one phase flow regime at low temperature and/or high strain rates, an undesirable two-phase flow regime at high temperature and/or low strain rates.

KEYWORDS: flax fibres, rheology, concentrated fibrous suspension, X-ray microtomography.

INTRODUCTION

Polymer composites reinforced with natural fibres are increasingly used in many industrial applications, for instance in the automotive industry [1]. Flax fibre mats can be used to reinforce polymer matrices to obtain compounds moulded by compression. Due to the interesting physical properties of flax fibres, these materials offer a suitable alternative to classical glass-fibre compounds such as Sheet Moulding Compounds (SMC), Bulk Moulding Compounds (BMC) or Glass Mat Thermoplastics (GMT) for producing semi-structural parts. But it is well known that these latter classical materials exhibit rather complex flow phenomena such as fibre orientation, fibre-matrix separation and smooth polymeric skin-layer development [2-4]. These phenomena depend on materials parameters such as the microstructure of the fibrous reinforcement, the matrix rheology and the content of mineral fillers, as well as on the process conditions such as the strain rates, the temperature of the mould and of the compounds and the quality of the contact between the mould surfaces and the compound.
There are structural analogies between flax-fibre mats and glass-fibre mats, e.g. consolidation of the fibrous mat by needle-punching. Thus, this study aims at determining the rheological phenomena occurring during the compression moulding of flax-fibre reinforced compounds. It will allow a better understanding of the analogies

between compounds with plant-fibres and glass-fibre compounds, as well as to reveal specific phenomena.

MATERIALS AND RHEOLOGICAL TESTS

A mixture of 90 % wt of slender bundles (mean length ≈ 45 mm, mean diameter ≈ 25 µm) of flax fibres and 10 % wt of slender polypropylene (PP) fibres (mean length ≈ 45 mm, mean diameter ≈ 15 µm) was used as raw material for producing non woven fibrous mats. These mats were produced by carding at the CENT (Centre Européen du Non Tissé) (Roubaix, France). A cross-lapper was used to form webs of required basis weights (240 g m^2). Then these mats were consolidated: i.e. they were needle-punched twice to obtain the reference mats. The microstructure is revealed in figure 1 where an image obtained by X-ray microtomography is shown. This image reveals the complex and entangled structure of the fibrous network of the flax fibre mats. It shows also that fibres are mainly aligned in-plane. After suitable filtering and thresholding operations, this image could also be used to calculate the volume fraction of fibres, which is low and equal to 5%. Some of these mats were also thermo-linked at 180°C in order to melt PP fibres to increase the bonding degree between fibres.

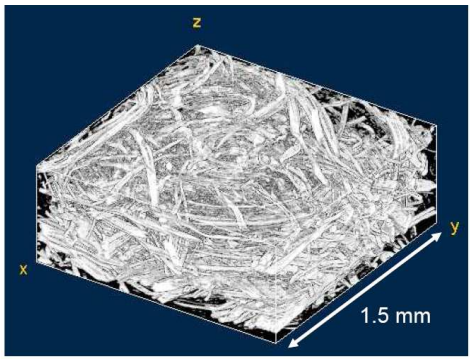

Fig. 1 Image obtained by microtomography of the fibrous network of the flax fibre mat (voxel size of 5×5×5 μm^3, ESRF, ID19 beamline).

These mats were impregnated with a paraffin gel (Versagel®, Penreco). It is a transparent material, which can be easily handled at room temperature. Its viscosity is close to viscosities of opaque industrial suspending polymers, when heated at moderate temperatures (≈ 10^3–10^4 Pa s at 1 s^{-1} and 50°C or 1 Pa s at 1 s^{-1} at 120°C).
Four steps are needed to make the prepreg. In the first step, the paraffin gel is melted in a mould (160 mm × 80 mm) at a temperature of 130°C. In a second step, a stack of two flax fibre mats is inserted into the mould. The mats are impregnated by capillary action for 30 min. In a third step, the mould is closed to compress and consolidate for 20 min the assembly. For this operation, the temperature of the mould is set at 100°C to increase the viscosity of the paraffin gel in order to avoid leakage. Finally, the sample (160 mm × 80 mm × 5 mm) is extracted from the mould.
Square samples (length L_0 = 80 mm) were compressed between cylindrical compression plates (diameter of 400 mm) heated at 50 ± 1°C or 90°C. This rheometer was mounted on a mechanical testing device (MTS 4M, load cell 2 kN, maximum cross-head velocity 8 mm s^{-1}). Prior to the tests, the surfaces of the sample were coated with a silicone grease layer so that the homogeneity of the sample deformation was ensured during the compression. The current height h of the sample and the current axial compression force F ($\mathbf{e}_3$ direction) were simultaneously recorded during the tests. This allows the axial

strain $\varepsilon = \ln(h/h_0)$, as well as the axial stress $\sigma = |F|/L_0^2$ to be determined. All tests were performed at constant axial strain rate $D = \dot{h}/h$ varying from 3 10^{-3} to 0.3 s^{-1}.

RHEOMETRY RESULTS AND DISCUSSION

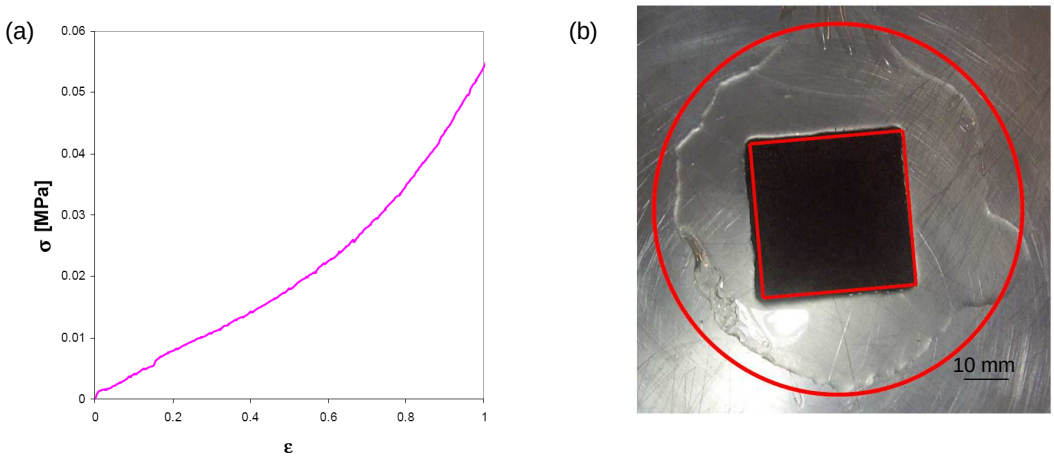

Fig. 2 (a) Stress-strain compression curve (composite made up of non thermo-linked mat, T = 90°C, D = 0.1 s^{-1}), (b) Picture of the composite after compression.

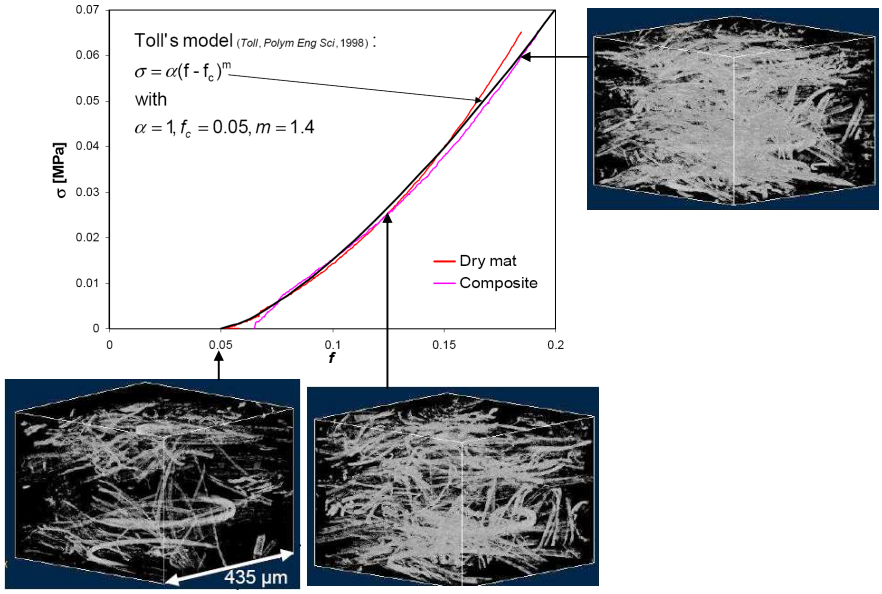

Fig. 3 Axial stress as a function of the fibre volume fraction f for the composite material deformed at 0.1 s^{-1} at a temperature of 90°C, as well as for a dry mat, together with scanned volumes observed at various compression stages (voxel size of 1,7×1,7×1,7 μm^3, ESRF, ID19 beamline).

Figure 2(a) shows the evolution of the axial stress with respect to the axial strain for a compression test performed at a temperature of 90°C and at a axial strain rate of D = 0.1 s^{-1}. This curve exhibits a typical non Newtonian rheological behaviour of the composite material. It shows also a significant strain-hardening. In figure 2(b), we have reported a picture of a sample after compression. It appears clearly that a two-phase flow occurred during the compression: there was no in-plane flow of fibres, liquid phase migration and fibrous phase consolidation. Furthermore, we have reported in figure 3 the evolution of the axial stress as a function of the fibre volume fraction f of the fibrous network of the composite material. It can be observed that the composite curve can be superimposed

with the consolidation curve of the dry mat (see also the micrographs depicting the consolidation of the fibrous mat). This figure underlines the negligible contribution of the matrix to the stress evolution for these specific flow conditions. This graph shows that the consolidation of the mat can be approximated by a phenomenological law, which is often used to describe the consolidation of fibrous media [5]. The values of α and m are respectively equal to 1 MPa and 1.4. These values are consistent with those of the literature [5].

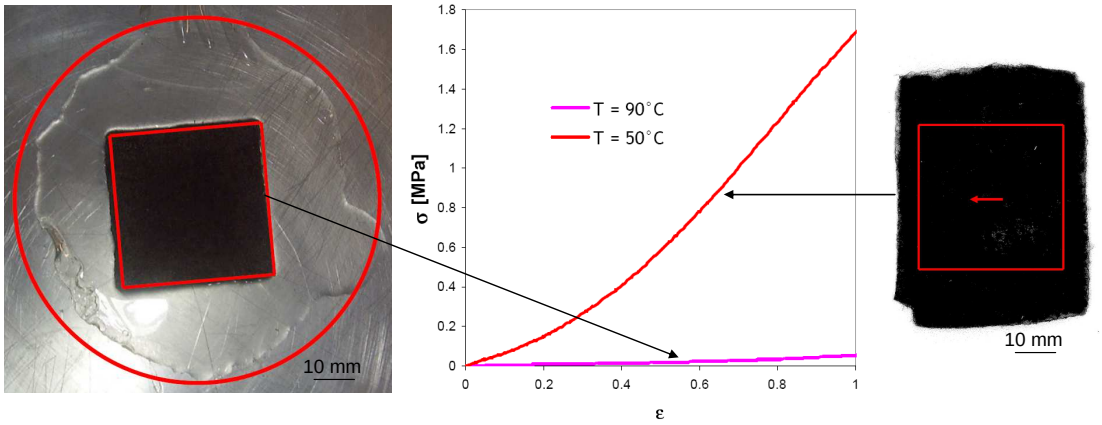

Fig. 4 Stress-strain compression curves at T = 50°C and T = 90°C (axial strain rate D = 0.1 s^{-1}) and pictures of the samples after compression.

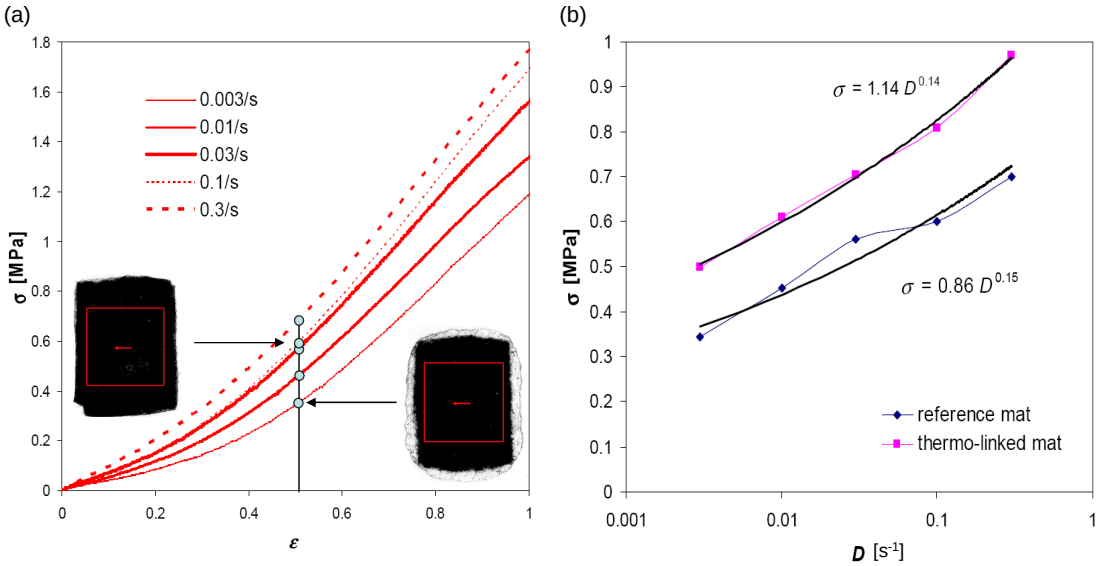

Fig. 5 (a) Stress-strain compression curves for various strain rates D, (b) influence of the strain rate D on the axial stress σ (for the composites made up of the reference mat and the thermo-linked mat).

It is clear that these flow conditions are inefficient to produce homogeneous composite parts: i.e. to disentangle the needled fibrous mat and drift the fibres. Then, some other flow conditions were tested in order to i) restrain the permeation of the liquid phase, ii) to induce and increase the pore fluid pressure, iii) to disentangle and drift the fibres during compression. Therefore it was chosen to increase the fluid viscosity by reducing the moulding temperature from 90°C to 50°C. The corresponding stress-strain curve is shown in figure 4 for a strain rate D of 0.1 s^{-1}, together with the previous experimental stress-strain curve obtained at 90°C. Moulding at 50°C increases the stress significantly

by a factor equal to 25! Nonetheless, as shown by the picture of a sample after compression (right hand side of the graph of figure 4), the flow was homogeneous and one-phase: i.e. no liquid phase migration could be observed and the fibres flowed in-plane. Furthermore the rectangular shape of this sample reveals that the flow was anisotropic and easier perpendicular to the fibre orientation as indicated by the arrow on the picture. At 50°C, the influence of the strain rate is revealed for the stress-strain curves in figure 5(a): a moderate increase of stress levels is recorded with the increase of the axial strain rate *D*. Figure 5(b) shows that the stress is a power law function of the strain rate and that the composite exhibits a shear-thinning rheology as given by the strain rate sensitivity exponent equal to 0.15. Once again two flow regimes can be identified: for *D* below 0.03 s^{-1}: a one-phase flow, for *D* over 0.03 s^{-1}: a two-phase flow. Finally, figure 5(b) shows that the thermo-linking operation increases the stress response by 35%.

CONCLUSION

Lubricated compression tests were performed using a large size rheometer to analyse the rheology of a model polymer composite reinforced with a needled non-woven flax mat. Test results revealed the influence of the testing temperature, the applied strain rate, the thermo-linking and the fibre orientation on the rheology of the composites. This behaviour exhibits strain hardening, shear thinning and anisotropy. Two flow regimes were identified: at low temperature and/or high strain rates: a one-phase flow, which is relevant for compression moulding; and at high temperature and/or low strain rates: an undesirable two-phase flow.

ACKNOWLEDGMENTS

This work was performed partially within the framework of the ESRF LTP “Heterogeneous Fibrous Materials”.

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