ALIGNED FLAX FIBRE/POLYLACTATE COMPOSITES
–
A MATERIALS MODEL SYSTEM TO SHOW THE POTENTIAL OF BIOCOMPOSITES IN ENGINEERING APPLICATIONS

Bo Madsen [1], Hans Lilholt [1], Anders Thygesen [2], Elaine Arnold [3],
Brendon Weager [3], Roberts Joffe [4]

[1] *Materials Research Dept., Risø National Laboratory for Sustainable Energy, Technical University of Denmark,*
DK-4000 Roskilde, Denmark
[2] *Biosystems Dept., Risø National Laboratory for Sustainable Energy, Technical University of Denmark,*
DK-4000 Roskilde, Denmark
[3] *NetComposites, Tapton Park Innovation Centre,*
S41 0TZ, Chesterfield, United Kingdom
[4] *Dept. of Applied Physics and Mechanical Engineering, Luleå University of Technology,*
SE-971, 87 Luleå, Sweden

KEYWORDS: natural fibres, PLA, fibre content, porosity, tensile properties

ABSTRACT

The potential of biocomposites in engineering applications is demonstrated by using aligned flax fibre/polylactate composites as a materials model system. The failure stress of flax fibres is measured by tensile testing of single fibres and fibre bundles. For both fibre configurations, it is found that failure stress is decreased by increasing the tested fibre volume. Based on two types of flax fibre preforms: carded sliver and uniaxial non-crimp fabric, aligned flax fibre/polylactate composites were fabricated with variable fibre content. The volumetric composition and tensile properties of the composite were measured. For composites with a fibre content of 37 % by volume, stiffness is about 20 GPa and failure stress is about 180 MPa. The tensile properties of the composites are analysed with a modified rule of mixtures model, which includes the effect of porosity. The experimental results are well predicted by the model. The back-calculated effective stiffness and failure stress of the flax fibres are in the ranges 56-60 GPa and 515-730 MPa, respectively. Finally, model predictions are used to present overall guidelines for the comparisons between tensile performance of flax fibre composites and traditional fibre composites (exemplified by glass fibre composites). The stiffness per volume, per weight and per cost is compared for these two types of composites.

INTRODUCTION

Cellulose fibres, exemplified by flax fibres, constitute potentially strong and stiff fibres based on biomass. In order to evaluate and qualify such fibres for use in strong and stiff composites, the fibre characteristics need to be investigated. This is done by measuring the failure stress for single fibres and for fibre bundles. Composites with flax fibres are fabricated and tested mechanically to allow a derivation (back-calculation) of the in-situ fibre failure stress. The failure stress of flax fibres and their composites will be analysed and compared. The basis of the comparison is selected to be the test volume of the fibres. This allows a fair comparison between single fibres, fibre bundles and in-situ fibres in composites.

The characteristics of fibre composites are governed by the volumetric composition, i.e. the volume fractions of fibres, matrix and porosity. The model developed previously [1] will be used to describe the volumetric composition of flax fibre composites in a rational way. The model furthermore allows a prediction of composite stiffness as a function of the volumetric composition, represented by the fibre weight fraction of the composites.

Finally, a comparison will be made between flax fibre composites and conventional glass fibre composites, on the basis of the composite stiffness, evaluated per volume, per weight and per cost.

MATERIALS AND METHODS

Single fibre testing

Flax fibres (code F1) were supplied by Ekotex, Poland. Single fibres were manually separated from the bundles. Fibre ends were glued onto a paper frame according to the preparation procedure described in ASTM D 3379-75 Standard. During mounting the specimens were handled only by the paper frame. Fibre length inside the frame defines the gauge length, which was 10 and 20 mm. Upon clamping of the ends of the paper frame by the grips of the test machine, frame sides were carefully cut in the middle. In total, 49 and 19 fibres respectively for each length were tested. The tests were carried out on an electromechanical tensile machine. In case of 10 mm long fibres the tensile machine was equipped with mechanical grips, while 20 mm long fibres were tested with pneumatic grips. Load–displacement curves were recorded during the test. The upper grip of the machine was attached through a hinge and thus allowed to self-align. All tests were displacement controlled with the loading rate of 1 mm/min and 2 mm/min for 10 mm and 20 mm long fibres respectively (equivalent to a strain rate of $1.67\ 10^{-3}\ s^{-1}$). Although flax fibre cross-sections have a polygonal shape and fibre thickness varies somewhat along the fibre, cross-sections were treated as circular with constant diameter in order to simplify analysis. Five measurements of the diameter were performed along the length of each fibre, and the average was used to calculate failure stress of the fibres.

Fibre bundle testing

Flax fibres (code F1) were supplied by Ekotex, Poland. Tensile tests of fibre bundles with length of 14.8 mm were done with a gauge length of 3.0 mm and a displacement rate of 0.03 mm/min (equivalent to a strain rate of $0.167\ 10^{-3}\ s^{-1}$) using an Instron 5566 with pressley clamps (Stelometer 654 from Zellweger Uster) [2]. Following fracture of a fibre bundle, the two fibre bundle pieces were weighed (w_f). The cross-sectional area of the fibre bundle (S) was determined by the equation:

$$S\left[\mathrm{mm}^2\right]=\frac{w_f\left[\mathrm{mg}\right]}{\rho_f\left[\mathrm{g/cm}^3\right]\times l_f\left[\mathrm{mm}\right]} \qquad \text{Eq. 1}$$

where l_f is length of the fibre bundle (=14.8 mm), and ρ_f is fibre density, which was measured to be 1.56 g/cm^3 for the flax fibres. The failure stress of fibre bundles was calculated as F/S, based on the measured failure force (F) and the calculated cross sectional area (S). Fibre bundles with variable cross sectional areas within the range 0.004-0.800 mm^2 were tested.

Composite fabrication and testing

Two types of aligned flax fibre preforms were used for fabrication of composites: (i) carded sliver of aligned flax fibres (code F5) from Ekotex, Poland, and (ii) uniaxial non-crimp flax yarn fabric (code KOM784) from Engtex, Sweden. Two types of polylactate (PLA) films were used for matrix: (i) films made by extrusion of polymer granulates, which were supplied by Biomer, Germany, and (ii) films supplied by Sidaplax, Belgium. The composites were fabricated by film-stacking of flax fibre preforms and PLA films. The stacked fibre/matrix assemblies were consolidated by using either (i) press moulding or (ii) vacuum moulding. The two types of composites will be denoted type I (carded sliver, Biomer PLA, and press moulding) and type II (uniaxial non-crimp fabric, Sidaplax PLA, and vacuum moulding). The composites were made with variable fibre contents in the range 20-50 % by weight. Tensile properties were measured for all the composites, and the failure stress data and the stiffness data were used in the present analysis. The volumetric composition was measured for only type I composites (see description of method in the study by Madsen [3]).

RESULTS AND DISCUSSION

Failure stress of fibres and composites.

The failure stress of flax fibres and their composites will be analysed and compared. The basis of the comparison is selected to be the test volume of the fibres. This allows a fair comparison between single fibres, fibre bundles and in-situ fibres in composites.

The results of the tensile testing of single fibres (with gauge length of 10 and 20 mm) and fibre bundles (with gauge length of 3 mm) are presented in Figure 1, which is a plot of measured failure stress versus fibre volume. The fibre volumes were calculated as the product of measured cross-sectional area and gauge length. The tested flax fibres were of the same type (code F1). It can be observed in Figure 1 that there is a considerable spread of the volumes of the single fibres even within a single gauge length, and that the range of fibre volumes are overlapping between the two gauge lengths of 10 and 20 mm with means of 0.0042 and 0.0078 mm^3, respectively. The results for the fibre bundles show that the fibre volumes are ranging between 0.014 and 2.168 mm^3, and that there is some overlap with the volumes of the single fibres.

In Figure 1, for both single fibres and fibre bundles, the failure stress is clearly decreased when the fibre volume is increased. This is well in line with the normal thinking firstly introduced by Griffith [4] that the failure stress of materials is reduced for increasing test volume due to the increased probability of having critical defects. Moreover, the results show that for *a given materials volume*, the failure stress is larger for fibre bundles that for single fibres. Qualitatively, this can be explained by the serial and parallel arrangement of fibre elements in single fibres and fibre bundles, respectively. In single fibres, the breakage of a weak fibre element will unavoidably lead to failure of the fibre, and thereby low failure stress. In fibre bundles, the breakage of the weak fibre element does not necessarily lead to failure of the bundle, since the stronger fibre elements in neighbouring fibres can take up the extra load, and thereby the fibre bundle will have larger failure stress.

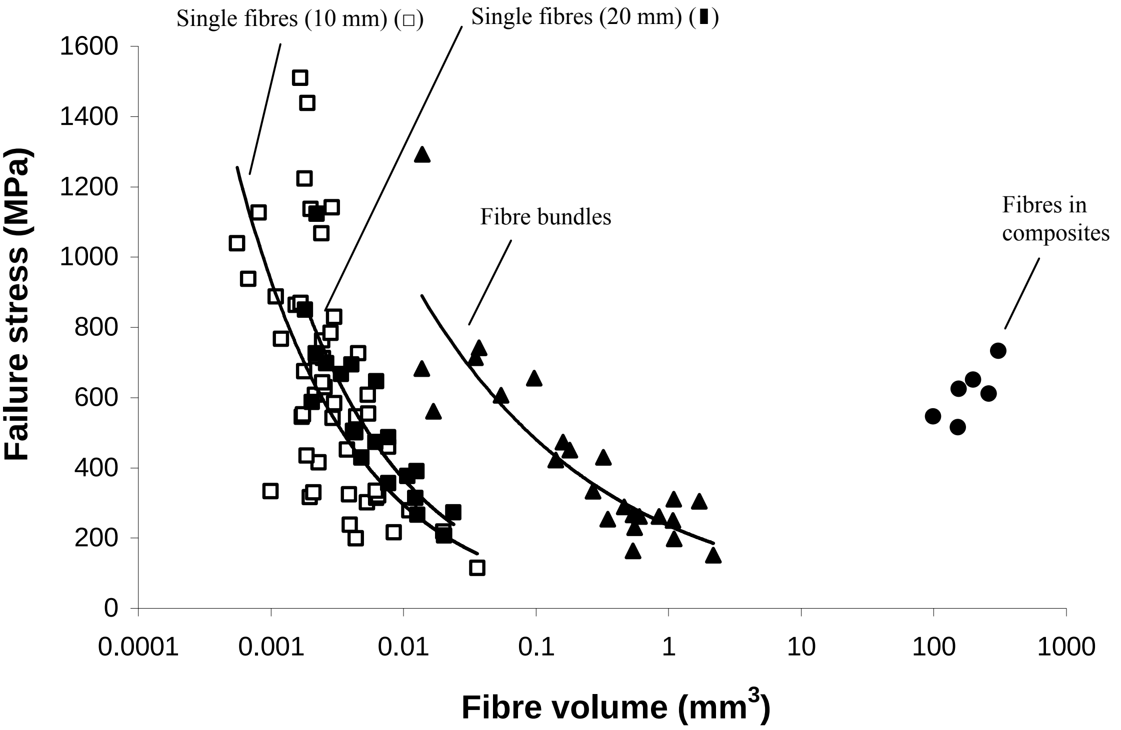

Figure 1. *Plot of measured fibre failure stress versus volume of flax fibres; single fibres (code F1, 10 and 20 mm gauge length), fibre bundles (code F1), and fibres in aligned flax fibre/PLA composites (code F5, see text for details). Lines are fitted power law regression lines. The scale of the fibre volume axis is logarithmic.*

Figure 1 shows also results for failure stress of flax fibres in type I aligned flax fibre/PLA composites (carded sliver, Biomer PLA, and press moulding). The failure stress of the fibres is back-calculated from the measured failure stress of the composites using a “modified rule of mixtures” model [5]:

$$\sigma_c=\left(V_f\ \sigma_f+V_m\ \sigma_{m*}\right)\left(1-V_p\right)^2 \quad \Rightarrow \quad \sigma_f=\frac{\sigma_c\left(1-V_p\right)^{-2}-V_m\ \sigma_{m*}}{V_f} \qquad \text{Eq. 2}$$

where V_f, V_m and V_p are volume fractions of fibres, matrix and porosity, respectively, σ_c is the failure stress of the composites, σ_f is the effective failure stress of the fibres, and σ_{m*} is the effective failure stress of the matrix at the failure strain of the composites. The failure stress of the composites was measured to be in the range from 124 to 177 MPa in composites with fibre volume fractions in the range 0.22 to 0.37. As shown in the figure, the back-calculated failure stress of the flax fibres in the composites is in the range 515 to 730 MPa. The related fibre volumes in the figure correspond to the volume of fibres in the tensile test specimens with a gauge section of length 30 mm, width 15 mm, and thickness in the range 1.00 to 2.05 mm. Thus, in Figure 1, the results for fibres in composites are believed to represent large bundles of impregnated fibres. It is demonstrated that failure stress of impregnated fibre bundles are larger that could be expected from the extrapolated regression line of the un-impregnated fibre bundles. This can be explained by using the same qualitative analogy as above. In an impregnated fibre bundle, the breakage of a weak fibre element will not necessarily lead to failure of the impregnated fibre bundle, since the stronger fibre elements *both in the same fibre* *and in the neighbouring fibres* can take up the extra load, and thereby the impregnated fibre bundle will demonstrate even larger failure stress.

In conclusion, the data in Figure 1 illustrates well some fundamental relations between failure stress of single fibres and fibre bundles (un-impregnated and impregnated). Future work needs to be addressed to develop a more quantitatively based relation between the different fibre configurations. The figure also shows that there is a large spread of failure stress of single flax fibres, but the variation is considerable reduced when the fibres are used as reinforcement in composites. The good reinforcement capacity of flax fibres in composites is demonstrated by the estimated effective failure stress in the range 515 to 730 MPa.

Volumetric composition of composites

The volumetric composition (i.e. V_f, V_m and V_p) and the fibre weight fraction (W_f) were measured for the type I composites (carded sliver, Biomer PLA, and press moulding). The results are presented in Figure 2A which shows a plot of the volumetric composition as a function of the fibre weight fraction. Predictions of the experimental data are made by a model developed in recent study by Madsen et al. [1]. The model equations will be not presented here.

The model lines in Figure 2A predict two cases of composite volumetric interaction: Case A and B. In Case A, where the fibre weight fraction is below a transition value, $W_{f\ trans}$, the volume fractions of fibres, matrix and porosity are governed by the density of fibres and matrix, and a number of fibre and matrix correlated porosity constants that can be evaluated from the microstructure of the composites. In Figure 2A, $W_{f\ trans}$ is equal to 0.46, and the model predictions are made using a fibre density of 1.56 g/cm^3, a matrix density of 1.25 g/cm^3, and a fibre correlated porosity constant of 0.25 (the matrix correlated porosity constant is assumed to be zero). In Case B of the model, the fibre weight fraction is above $W_{f\ trans}$. The fibre *assembly* is fully compacted to its minimum volume (under the operating process conditions), which means that the volumetric interaction is constrained by a maximum obtainable fibre volume fraction, $V_{f\ max}$. In Figure 2A, $V_{f\ max}$ is equal to 0.37. Altogether, it is shown in the figure that when W_f is increased from zero towards $W_{f\ trans}$, the porosity is relative small, and is only modestly increased. However, as W_f is increased above $W_{f\ trans}$, the porosity starts to increase dramatically. This is due to the situation where the fibre assembly is compacted to its minimum volume (i.e. V_f has reached $V_{f\ max}$), and the available matrix volume is insufficient to fill the free space between the fibres.

In conclusion, it can be observed in Figure 2A that the model is well capable of predicting the volumetric composition of composites as a function of the fibre weight fraction. More examples of the good agreement between model predictions and experimental data of plant fibre composites are shown in the study by Madsen et al. [1].

Stiffness of composites: aligned flax fibre/PLA composites

Figure 2B shows the measured stiffness of the type I aligned flax fibre/PLA composites (carded sliver, Biomer PLA, and press moulding) as a function of fibre weight fraction. Maximum stiffness of about 20 GPa is obtained experimentally for composites with a fibre weight fraction of 0.47, which corresponds to a fibre volume fraction of 0.37 ($\approx V_{f\ max}$ in Figure 2A). The model line in Figure 2B is made by the "modified rule of

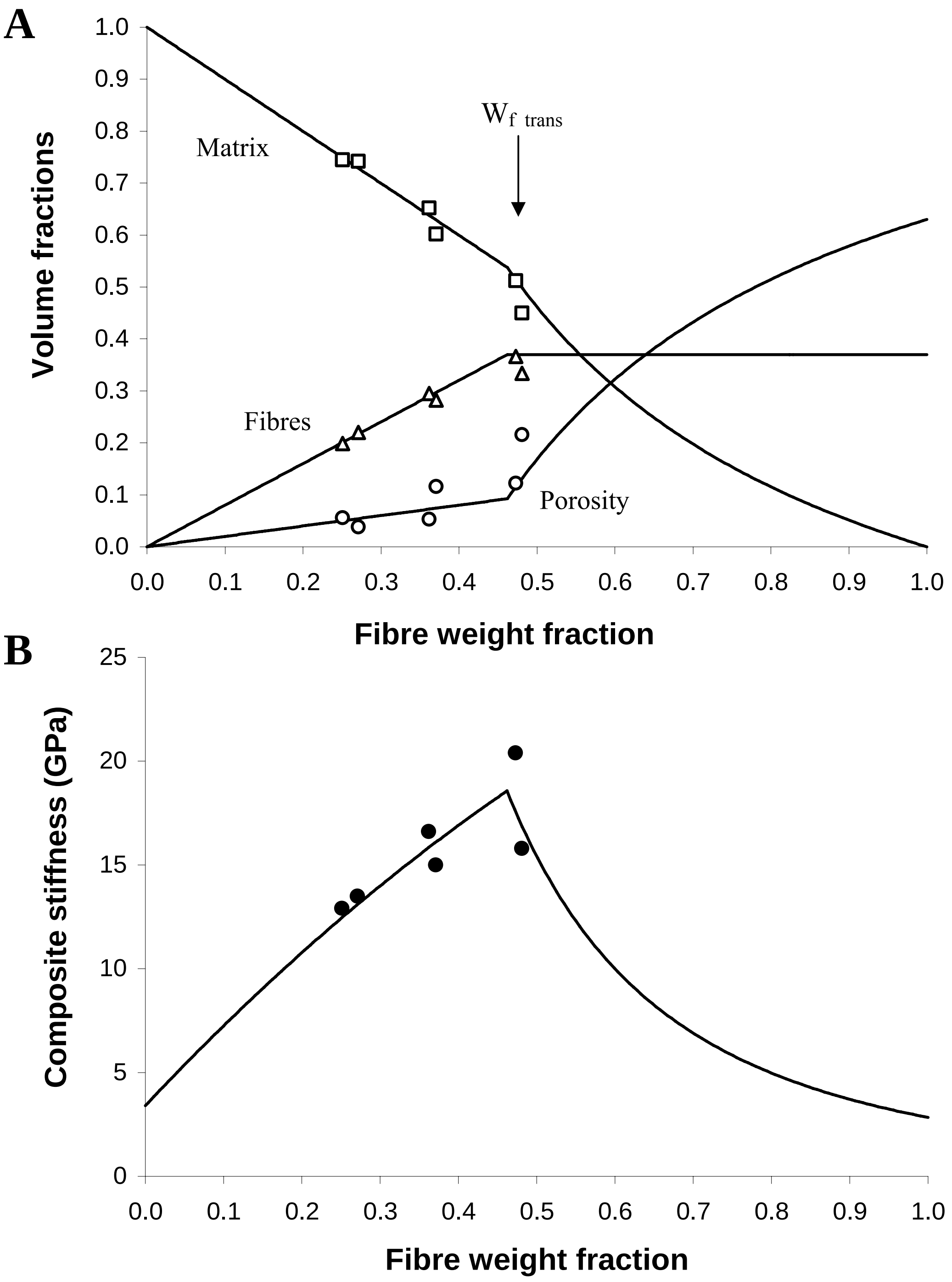

Figure 2. *(**A**) Volumetric composition and (**B**) stiffness as a function of fibre weight fraction of type I aligned flax fibre/PLA composites (carded sliver, Biomer PLA, and press moulding). Symbols are experimental data and lines are model predictions. See text for details.*

mixtures" model (Eq. 2) by replacing failure stress (σ) with stiffness (E) and by using the model predictions of Figure 2A to convert volume fraction of fibres, matrix and porosity (V_f, V_m and V_p) to fibre weight fractions. The model line is calculated by using a measured matrix stiffness (E_m) of 3.4 GPa and a fitted fibre stiffness (E_f) of 56 GPa. The model line is in good agreement with the experimental data. The line predicts that the stiffness increases non-linearly up to a maximum value at the transition fibre weight fraction (the same as in Figure 2A), and stiffness is thereafter rapidly decreased due to the increase in porosity as shown in Figure 2A.

The volumetric composition was not measured for the type II aligned flax fibre/PLA composites (uniaxial non-crimp fabric, Sidaplax PLA, and vacuum moulding). For these composites, the fibre weight fraction was only assessed from the weight ratio of fibres and matrix used to fabricate the composites. In many cases, this is the typical way to estimate the fibre content of composites. However, model predictions of mechanical properties require knowledge of fibre content by volume, and not by weight. Accordingly, a model is needed to convert weight fractions into volume fractions, and as shown above, this can be done by the model presented in Figure 2A.

Figure 3 shows the measured stiffness of the type II aligned flax fibre/PLA composites (uniaxial non-crimp fabric, Sidaplax PLA, and vacuum moulding). Maximum stiffness of about 19 GPa is obtained experimentally for composites with a fibre weight fraction of 0.50. The model line is calculated based on the same input parameters as used in Figure 2, only $V_{f\,max}$, and E_f is slightly changed to fit the data. $V_{f\,max}$ is set equal to 0.39 (corresponding to a $W_{f\,trans}$ of 0.49) and E_f is set equal to 60 GPa. It can be observed that the model line is well predicting the experimental data.

The work on integrating model predictions of volumetric interaction and mechanical properties of plant fibre composites, as exemplified in Figures 2 and 3, is ongoing [6].

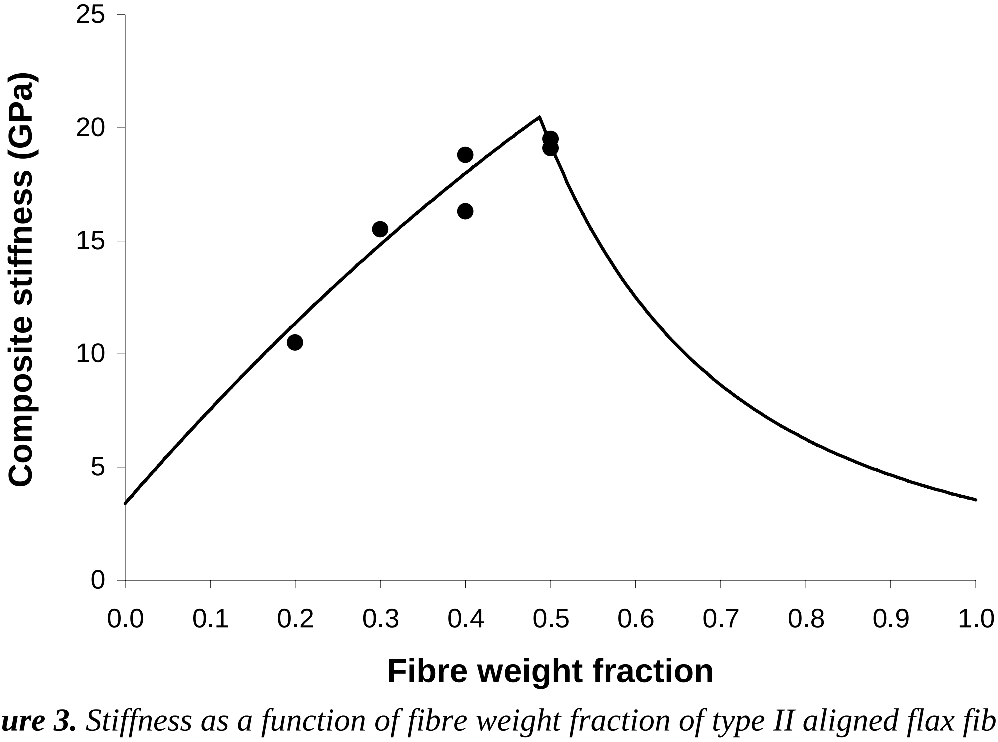

Figure 3. *Stiffness as a function of fibre weight fraction of type II aligned flax fibre/PLA composites (uniaxial non-crimp fabric, Sidaplax PLA, and vacuum moulding). Symbols are experimental data and lines are model predictions. See text for details.*

Stiffness of composites: comparison between flax fibres and glass fibres

The above presented models of volumetric composition and stiffness of composites can be used to make overall guidelines for the comparison between stiffness of flax fibre composites and traditional fibre composites. The latter ones will be exemplified by glass fibre composites. Figure 4A-C shows model predictions of stiffness per volume, per weight and per cost of aligned fibre composites with flax fibre/PLA matrix, and glass fibre/PLA matrix.

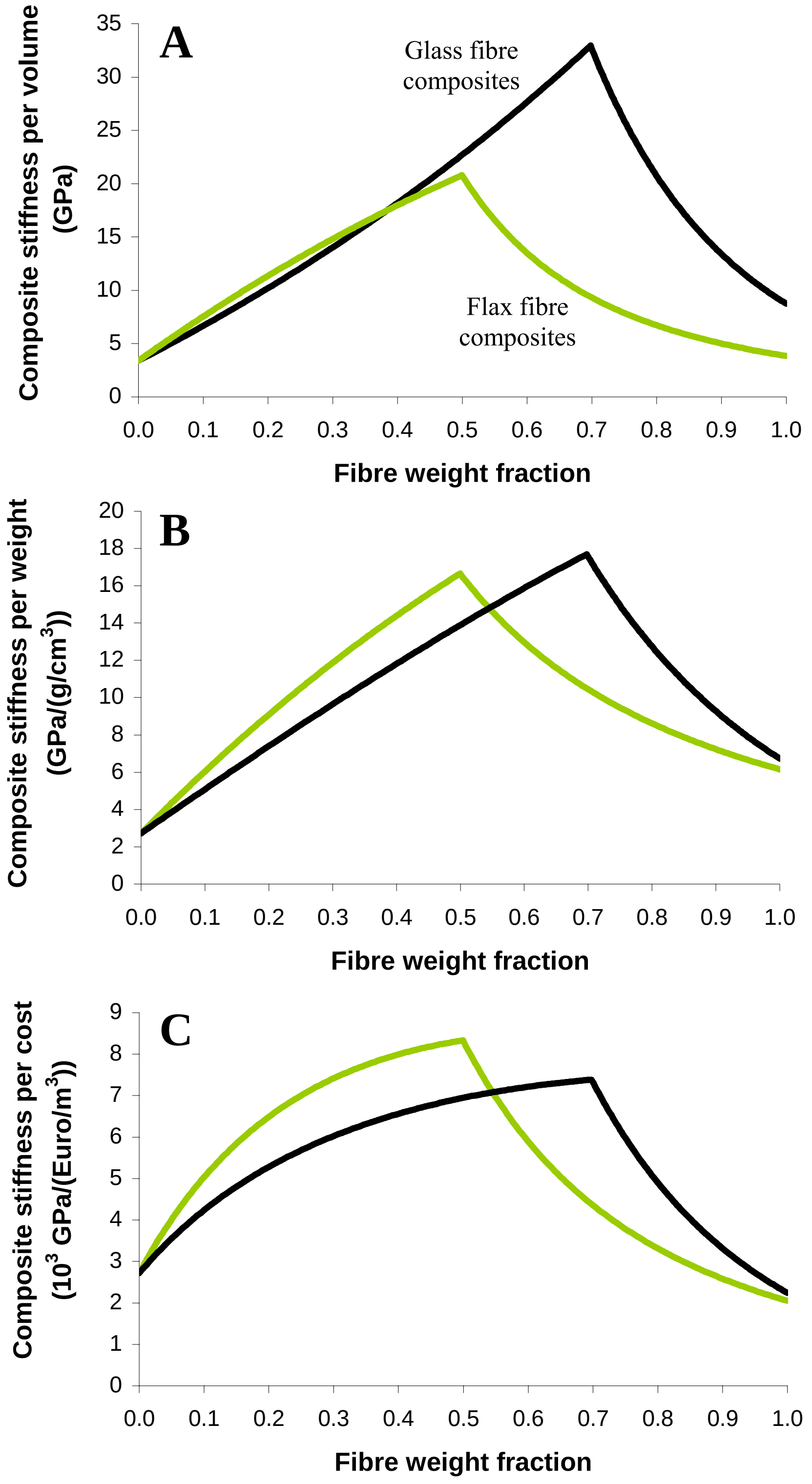

Figure 4. *Guidelines for comparison between aligned flax fibre and glass fibre composites with a PLA matrix: (**A**) stiffness per volume, (**B**) stiffness per weight, and (**C**) stiffness per cost. See text for details of model parameters.*

The input parameters used in the model predictions for flax fibres and glass fibres are following: 1.56 vs. 2.60 g/cm^3 for the fibre density, 0.25 vs. 0.10 for the fibre correlated porosity constant (i.e. it is assumed that glass fibre composites contain less porosity), 0.40 vs. 0.50 for the maximum obtainable fibre volume fraction (i.e. it is assumed that glass fibre assemblies have larger packing ability [7]), 60 vs. 70 GPa for the fibre stiffness, 3.0 Euro/kg of both flax and glass fibre preforms (i.e. it is conservatively assumed that the cost of the two fibre types is the same). The cost of PLA matrix is assumed to be 1.0 Euro/kg.

In comparison to glass fibres, flax fibres have typically lower stiffness, and lower fibre packing ability [7] which lead to lower maximum obtainable fibre volume fraction in the composites. In addition, flax fibre composites typically also have larger porosity content. Accordingly, as shown in Figure 4A, flax fibre composites will typically have lower maximum obtainable stiffness (per volume). The figure shows maximum stiffnesses of 20 and 33 GPa for flax and glass fibre composites, respectively.

A key technical advantage of flax fibres (and of plant fibres in general) is their relatively low density of about 1.5 g/cm^3, which results in good weight based mechanical properties (also denoted specific properties). This is demonstrated in Figure 4B showing that maximum stiffness per weight of flax and glass fibre composites is almost the same: 16.3 and 17.2 GPa/(g/cm^3), respectively.

Materials cost is of critical importance in most industrial products. Even though flax fibres are derived from low-cost biomass resources, the required manufacturing steps (i) to grow the plants, (ii) to extract fibres from plants, and (iii) to process the fibres into suitable preforms for composites, are all adding costs to the use of flax fibre as reinforcement in composites. However, based on information from fibre suppliers it can be assumed that the cost of flax fibre preforms is the same (or potentially less) that the cost of glass fibre preforms [3]. As demonstrated in Figure 4C, in the case of the same cost per weight of fibre preforms, flax fibre composites typically will have larger stiffness per cost than glass fibre composites. The figure shows maximum stiffness per cost of 8.2 and 7.3 $10^3 \cdot$GPa/(Euro/m^3) for flax and glass fibre composites, respectively.

CONCLUSIONS

The flax fibre failure stress was evaluated relative to the test volumes, for single fibres, for fibre bundles, and for in-situ fibres in composites. These represent individual fibres, un-impregnated bundles, and impregnated bundles.

For individual fibres the failure stress decreases with increasing test volume.

These also holds for fibre bundles, and for a given materials volume the failure stress is larger for bundles than for individual fibres.

For in-situ fibres in composites the relatively few data represent fibre failure stress at larger test volumes. Such impregnated fibre bundles show larger failure stresses than would be expected for un-impregnated fibre bundles at a similar materials volume.

For individual fibres, un-impregnated fibre bundles and impregnated fibre bundles, the average failure stresses are comparable in the range 430 to 630 MPa.

The volumetric composition model is predicting well the experimental data for volume fractions of fibres, matrix and porosity for the type I aligned flax fibre/PLA composites.

The model also predicts well the experimental stiffness values, with a predicted maximum stiffness for type I aligned flax fibre/PLA composites of 19 GPa at a transition fibre weight fraction of 0.46.

The model also predicts the curves for stiffness without detailed knowledge of the volumetric composition, as exemplified for the type II aligned flax fibre/PLA composites.

The comparison between flax fibre composites and glass fibre composites shows that on volumetric basis glass fibre composites have higher maximum stiffness, on weight basis stiffnesses are very similar, and on cost basis flax fibre composites have higher maximum stiffness (for same cost per kg for fibre preforms).

ACKNOWLEDGEMENTS

The work was partly financed by the EU sixth framework programme project “New classes of engineering composite materials from renewable resources”, BIOCOMP, IP 515769-2.

REFERENCES

1. Madsen B., Thygesen A. and Lilholt H., “Plant fibre composites – porosity and volumetric interaction”, *Composites Science and Technology*, 2007; 67: 1584-1600.
2. Thygesen A., Daniel G., Lilholt H. and Thomsen A. B., “Hemp fiber microstructure and use of fungal defibration to obtain fibers for composite materials”, *Journal of Natural fibres*, 2005; 2(4): 19-37.
3. Madsen B., “Properties of plant fibre yarn polymer composites – An experimental study”, Ph.D.-thesis, Technical University of Denmark, Denmark, 2004, Series R, no 82.
4. Griffith A. A., *Phil. Trans. R. Soc.*, 1920; A221: 163.
5. Madsen B., Hoffmeyer P. and Lilholt H., “Hemp yarn reinforced composites – II. Tensile properties”, *Composites Part A*, 2007; 38: 2204-2215.
6. Madsen B., Thygesen A. and Lilholt H., “Plant fibre composites – porosity and tensile stiffness”, 2008, In preparation.
7. Madsen B. and Lilholt H., “Compaction of plant fibre assemblies”, In: Proceedings of the 23rd Risø International Symposium on Materials Science: “Sustainable Natural and Polymeric Composites – Science and Technology”, Risø National Laboratory, Roskilde, Denmark, 2002: 239-250.