NOVEL BIODEGRADABLE WOOD FIBRE POLYLACTIC ACID FOAM SANDWICH COMPOSITES

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1 Introduction

SustainComp, a project sponsored by the Seventh Research Framework Programme (FP7) of the European Union (EU), has as one of its main goals to develop advanced cellular composites based on renewable resources, for application in the fields of packaging, display and core materials [1,2]. Fibre reinforced foams have the potential of being lighter, stiffer and stronger than conventional foams.

Until now wood fibre reinforced polylactic acid (PLA) composite foams have been successfully produced using supercritical carbon dioxide. Upon addition of wood fibres the stiffness properties of the foams in compression improve. A significant increase of specific stiffness was achieved by adding 5-10 wt% wood fibres [1].

The aim of this particular work has been to develop a process to produce sandwich materials where the faces are composites of PLA and wood fibres and the core is a foam structure of PLA, which might also be reinforced with wood fibres.

Sandwich structures represent a key component of composites structural design technology. They provide the structural efficiency of very lightweight material (core) "sandwiched" between higher stiffness and strength laminates (skins) in order to carry tension, compression and shear loads imposed upon the resultant structure. The primary properties of interest for the core material are typically low density and high compression and shear stiffness and strength as well as good bonding with the skins. PLA foams, both neat and reinforced with wood fibres, are investigated as a core material for sandwich structures intended for packaging applications.

2 Materials and Methods

2.1 Preform Manufacturing

Stratified preforms of PLA fibres (PLA01, N.I. Teijin Shoji Co. Ltd., Japan) and wood fibres (fully bleached birch, Innventia AB, Sweden) were manufactured using a wet commingling technique similar to slurry processing in papermaking [1]. A black colouring agent (Cartasol, Sandoz AG, Switzerland) was used to dye the wood fibres. The colouring reduces the dewatering time during preform fabrication [3]. Only wood fibres used to reinforce the core material were died since preliminary results showed a very poor adhesion between the PLA and coloured wood fibre skins. It has been shown in another work that treatment of the wood fibres with a surfactant can contribute to increase the foam expansion attributable to reduced wood fibre network forming ability [4]. However, due to worse fibre-matrix adhesion of treated fibres, foams with inferior strength properties were obtained.

Each preform weighed 2 g and consisted of a fibre mat composed of three layers, i.e. the two wood fibre skins and the PLA core in between. The wood fibre content in the core was changed from 0 to 10 wt%. The preforms were dried in a ventilated oven at 55°C for more than 24 hours before being consolidated by compression moulding at 120°C and 200 bar for 10 min. Additional drying before any further processing was done at 55°C for at least 24 hours to avoid matrix degradation.

2.2 Foaming with Supercritical Carbon Dioxide

Foaming was carried out in a custom made autoclave (SITEC Sieber Engineering AG, Switzerland) with supercritical CO_2 (PanGas AG, Switzerland). The saturation temperature and pressure were chosen as 165°C and 200 bar, respectively. The depressurization rate was set to 5 bar/s in order to ensure good foam homogeneity [2]. Rectangular strips (20×5 mm) cut out from the consolidated compounds were either expanded freely or confined in custom designed moulds.
The moulds were designed and built in order to satisfy several requirements, i.e. during foaming samples must be kept horizontal so that the expansion can occur in the thickness direction of the sample (*z*-axis) and the elongated shape of samples shall match the small diameter of the autoclave chamber. Moulds were constituted by three assembled parts (i) a plate which supports the samples and allows moulds to be stacked in a rack, (ii) a frame that imposes the maximum possible foaming expansion depending on its thickness (3 mm in this case) and (iii) a thin perforated cover that confines the expansion along *z*-axis, allowing CO_2 to enter during pressurization and to leak during depressurization.

2.3 Experimental

Scanning electron microscopy (SEM) was used to observe the microstructure of the obtained sandwich materials. Samples were cut using a razor blade and stuck onto aluminium pin-type studs with double sided adhesive carbon discs. Furthermore, gold coating was made with a BioRad sputter coater (Polaron Instruments Inc., USA) in order to obtain an electrically conductive thin coating to improve secondary electron emission. Samples were observed with a microscope (FEI XLF30-FEG, FEI Company, Netherlands) in secondary electron mode at an accelerating voltage of 3 kV.
Three point bending tests were performed on sandwich beam samples with a Minimat microtensile testing machine with 200 N load cell at a deformation rate of 0.5 mm/min. The bending stiffness was, for the sake of comparison, evaluated according to standard ISO/DIS 5628 [5] for paper and board as

$$S_b=(k_b l^3)/(3w) \qquad (1)$$

where k_b is the slope in the linear region of the load-deformation curve, l the span length and w the width. The stress-strain behaviour, σ-ε, was obtained from the Euler-Bernoulli beam theory with the following equations

$$\sigma=(3Pl)/(3wh^2) \qquad (2)$$

where P is the recorded force and h is the thickness of the beam, and

$$\epsilon=(6\delta h)/(l^2) \qquad (3)$$

where δ is the midpoint deflection of the beam. The apparent density was determined by weighing and measuring the volume of each sample.

3 Results and Discussion

The stratified wood fibre/PLA compounds (Fig. 2a) were successfully foamed with supercritical CO_2. Fig. 1a-b show the structure of two different beam samples at the macroscopic level.

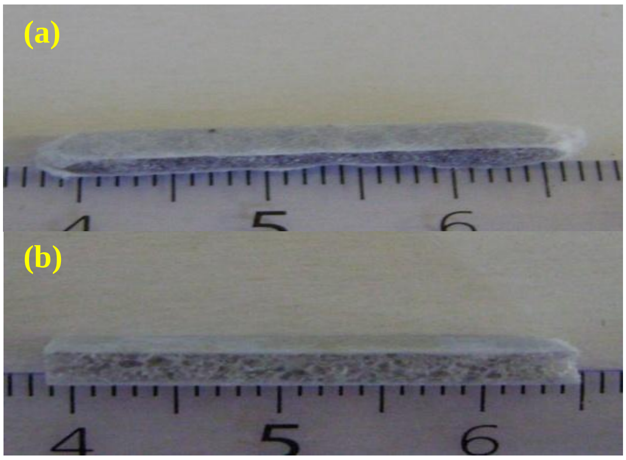

Fig.1. Sandwich beam with wood fibre faces and a PLA/1 wt% wood fibres core formed under (a) free and (b) confined expansion. Ruler in cm scale.

In case the stratified preforms were allowed to expand freely, the obtained sandwich materials (Fig. 1a) had an inhomogeneous expansion across the length leading to wrinkling of the skins. Due to PLA shrinkage, separations of the skins and core are clearly seen especially at the boundaries of the specimens. Fig. 1b shows that confined foaming resulted in sandwich materials with flat skins which were well impregnated by the PLA (as confirmed also by the SEM micrographs in Fig. 2) as opposed to materials formed under free expansion.

Confinement of the expansion resulted in an increased pressure applied by the growth of the foam core against skins. No lateral polymer shrinkage was observed and skin surfaces appeared well wetted by the PLA. The beam longitudinal axis (*x*-axis) remained straight and the cross section could be considered rectangular with constant dimensions along the *x*-axis (i.e. thickness of 3 mm).

3.1 Microstructure

Fig. 2a-d show a global view in the *x-z* plane of the preform before foaming (Fig. 2a) and the obtained sandwich materials with different wood fibre content in the core (Fig. 2b-d). In the stratified preform the fluffy wood fibre layers can be clearly distinguished from the compacted PLA fibres in the core. Addition of wood fibres in the core changed the foaming behaviour and thus final microstructure. Foam expansion decreased with increasing wood fibre content, while the number of cells and homogeneity increased. This translated into a density increase of the core of the sandwich material as seen in previous work [1,4]. It is also known that wood fibre surfaces act as nucleating agent [6].

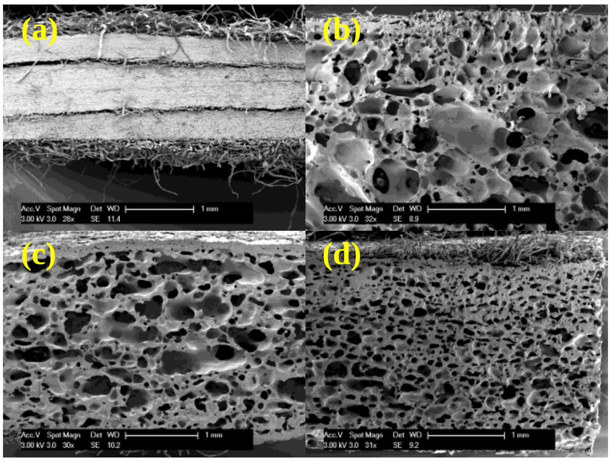

Fig.2. Micrographs of (a) preform with PLA fibre core, and sandwich materials with PLA foam core with (b) 1 wt%, (c) 5 wt% and (d) 10 wt% wood fibres. Scale bars 1 mm.

Fig. 3a-d highlight the interface between the core and the skins for different samples. Fig. 3a shows a beam sample where the core was allowed to expand freely. For the bottom layer, that was always in contact with the supporting plate, but is shown at the top of Fig. 3a the wood fibres seemed to be quite well embedded into the matrix.

However at the top layer, i.e. the layer that followed the foam core rise, the structure of the skin resembles that of an unprocessed preform (Fig. 3a) with very little or no PLA impregnating the initial fluffy wood fibre layer.

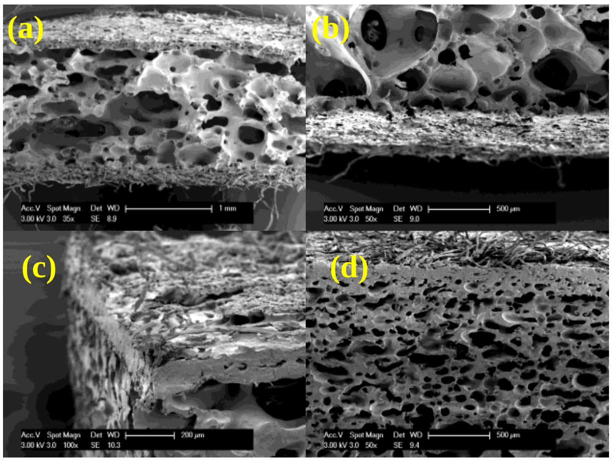

Fig.3. Micrographs of interface between the core and the skins for sandwich materials with (a) 1 wt% wood fibres in the core allowed to expand freely compared with materials foamed under confinement with (b) 1 wt%, (c) 5 wt% and (d) 10 wt% wood fibres in the core. Scale bars: 1 mm in (a), 0.2 mm in (b) and 0.5 mm in (c)-(d).

Sandwich beams for which the core expansion was limited had better overall skin-core adhesion. Fig. 3b shows a bottom skin for a sandwich with 1 wt% wood fibres in the core, while Fig. 3c shows the beam edge and skin-core interface for the top skin of a foam with 5 wt% wood fibres in the core. The top skin for a sandwich beam with 10 wt% wood fibres in the core is shown in Fig. 3d.

It is clear that by limiting the expansion with addition of wood fibres the skin-core adhesion is influenced. Higher wood fibre content in the core leads to less core expansion which reduces the pressure on the perforated confinement plate and less PLA will flow through and impregnate the wood fibre skins in particular at the top skins.

3.2 Mechanical Properties

Eqs. (2)-(3) were used to obtain the stress-strain behaviour from the recorded load and deformation. It should be noted that these curves are obtained from bending tests, and only the initial parts of the stress-strain curves can to some extent reflect the axial tensile or compressive behaviour. Fig. 4 show some typical curves for sandwich samples with different wood fibre content in the core compared with a curve for a neat PL01 beam foamed under same conditions. The sandwich materials are stiffer and stronger than the neat PL01 beams but display a more brittle behaviour.

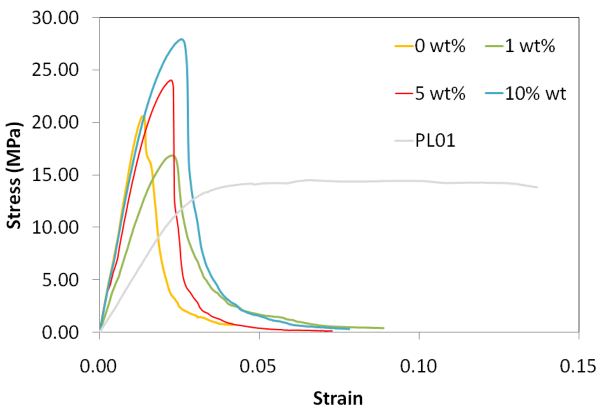

Fig.4. Stress-strain curves for different sandwich materials compared with the stress strain curve for a pure PL01 beam.

A measure of the bending stiffness can be obtained using Eq. (1). Fig. 5 shows the bending stiffness for pure PL01 foam beams as compared with sandwich beams where the PLA core wood fibre content was 0, 1, 5 and 10 wt%.

It is clear that there is an increase in the bending stiffness with use of wood fibre skins (comparing at same density). Addition of wood fibres in the core further increased the bending stiffness (by adding 1 wt%) due to higher stiffness of the core, a combined effect of the density increase (Fig. 2b-d) and wood fibre reinforcement [1,4].

It has been previously proved that wood fibres show reinforcing potential in foams, since they are organized in the foam structure to provide support in the cell walls oriented along the fibres [1,4]. Fig. 6 shows a fracture surface of a sandwich core confirming that wood fibres were contained within the foam cell walls.

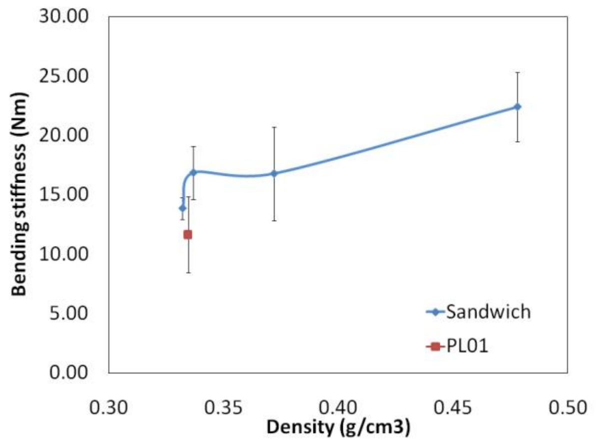

Fig.5. Bending stiffness vs. density.

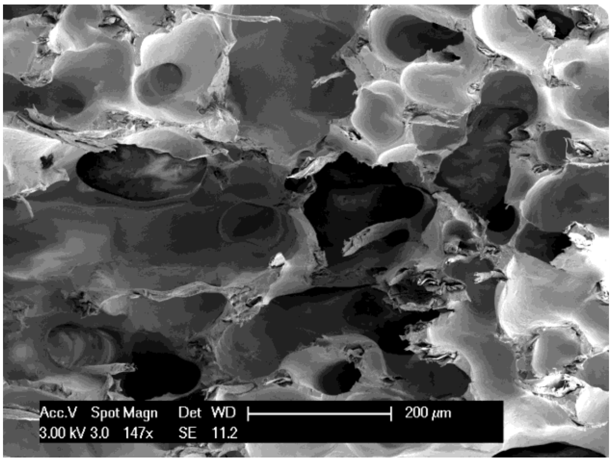

Fig.6. SEM micrograph showing a fracture plane of a sandwich core with 10 wt% wood fibres.

It is noteworthy that the stiffness of the sandwich materials with 5 wt% wood fibre in the core does not increase despite the large increase in density (Fig. 5). This could be an effect of inferior skin-core adhesion due to lower core expansion which could be observed in Fig. 3b-c.

Fig. 7 shows the maximum stress which can be seen as a measure of the flexural strength of the material as function of the density. The strength increased with increased material density. As for the stiffness, there is an increase due to the sandwich effect.

Addition of 1 wt% wood fibres in the sandwich core does not result in significant increase in flexural strength. Therefore the bending stiffness (Fig. 5) can be considered to increase to a greater extent than the strength.

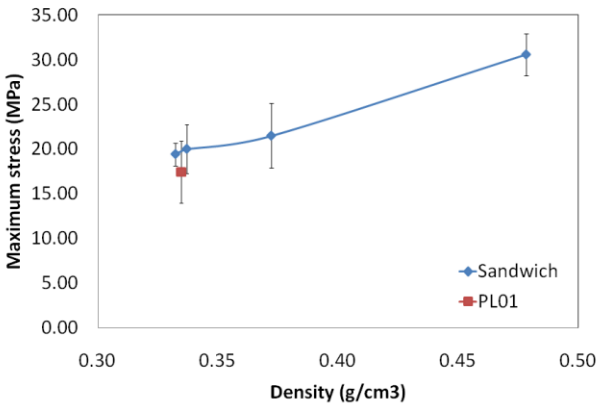

Fig.7. Maximum stress vs. density.

3.3 Conclusions

Stratified wood fibre/PLA compounds were successfully foamed with supercritical CO_2 under confined conditions. Addition of wood fibres in the core influenced the foaming behaviour and final microstructure of the foam cores. Foam expansion decreased, i.e. density increased, with increasing wood fibre content, but the number of cells and homogeneity increased. The foam expansion had a direct effect on the adhesion between the core and the skins. Core materials with higher expansion seemed to have better skin-core adhesion.

The bending stiffness of the resulting sandwich materials was considerably higher compared to neat foams at same density. There was a slight increase in flexural strength but the bending stiffness can be considered to increase to a greater degree than the strength.

Mechanical properties can be tailored depending on the application by changing the properties of the skins and/or core, e.g. by reinforcing with wood fibres, and/or changing the foaming conditions to obtain the confinement which will give the optimal skin-core adhesion.

Finally, one of the advantages of the material developed in this work over similar traditional foam core sandwich materials is that there is no need for an adhesive between the face sheets and the core.

Acknowledgements

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