EXPERIMENTAL INVESTIGATION INTO VOID CONTENT AND MECHANICAL PROPERTIES OF CURED FLAX/EPOXY PREPREGS

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Keywords: flax, mechanical properties, voids, prepreg

Abstract

Flax fibres have shown good potential in reinforcing composite materials but have posed several processing challenges which lead to a higher likelihood of process-induced voids. This study investigated the influence of voids on static tensile, interlaminar shear and moisture absorption properties of woven flax/epoxy composites. Samples were manufactured by conventional vacuum bagging techniques in an autoclave from two commercially available prepregs at pressures of 1, 3 and 5 bars. This led to void contents ranging from 0 to 25% as determined by optical microscopy and image analysis. The results show that among the studied properties, interlaminar shear and moisture absorption were most affected by the presence of voids. The latter was most critical and showed a 33% increase in mass gain after 24 hours in a saturated moisture absorption test with only a 3.5% increase in void content.

1. Introduction

Bast fibres such as flax are gaining significant attention from the composite materials community due in part to their low density, good mechanical properties and several environmental benefits [1]. However, these benefits are accompanied by some challenges in processing such as lower permeability for a given fibre volume fraction and fabric architecture [2] when compared with conventional reinforcements and potentially high moisture content in the fibres due to their hydrophilic nature. These challenges inevitably lead to a higher likelihood of process-induced voids which are known to be one of the most serious defects in composite materials [3]. The aim of the current study is to investigate the effect of voids on static tensile, interlaminar shear and moisture absorption properties in woven flax/epoxy composites.

Several studies have investigated the effect voids on mechanical properties in conventional composite materials. It is widely agreed upon that voids most significantly degrade the strength of composite materials [3]. The properties that are mostly affected are usually matrix-dominated such as compression [4], flexural [5, 6] and interlaminar shear [5, 7]. The tensile properties in unidirectional composites are generally observed to be void-insensitive [8]. On top of a reduction in static properties, fatigue life [9] and moisture absorption [10] are also known to be compromised by voids. Although there has been much work done in this area for conventional composites, little has been done in the case of bio-based composites.

Therefore the influence of voids on certain mechanical properties was selected as the focus of this study.

2. Experimental

2.1. Materials

Two different hot-melt flax prepregs (BL200 and BL550) based on 2x2 twill weaves were kindly supplied by Lineo NV. The prepregs varied in yarn architecture and spacing which resulted in different areal weight and crimp values. Both systems employed the same epoxy resin system (Huntsman LY5150) and the fabrics were treated by a patented sizing (Patent No. US8080288). Selected properties of these material systems relevant to the experimental results are provided in Table 1 with scans of the fabrics in Figure 1.

	Areal weight (g/m^2)	Fibres in Warp Direction (%)	Warp Crimp[a] (%)	Weft Crimp[a] (%)	Resin Mass Content (%)
BL200	196±2	51.2	1.2±0.1	1.6±0.1	52.1±1.8
BL550	543±2	51.3	2.1±0.5	7.9±0.7	46.8±2.0

[a] Crimp measured by image analysis on samples processed at 3 bars

Table 1. Selected properties of Lineo flax/epoxy prepregs

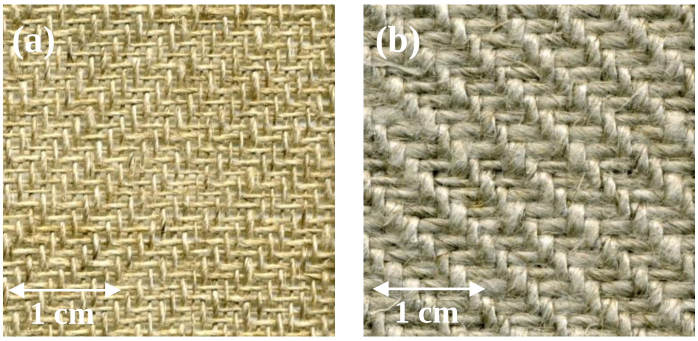


Figure 1. Dry 2x2 twill flax weaves used in the studied prepregs a) BL200 b) BL550

2.2. Sample preparation

Laminates were manufactured in an autoclave at pressures of 1, 3 and 5 bars in order to produce composites of varying void content. The cure cycle was selected based on the resin specifications (Figure 4). A 60 minute debulk at 50 °C and a pressure of -0 .7 bars was added at the beginning of the cure cycle to encourage air and moisture evacuation. This same level of vacuum was applied throughout the entire process. A conventional vacuum bagging arrangement incorporating "edge breather" [11] was employed (Figure 2(a)). Plates were manufactured with dimensions of 30 x 30 cm^2. The number of layers was selected to comply with thickness requirements stated by ASTM D3039 and ASTM2344 (3 and 6 for the BL200 and BL550 respectively). In the case of the BL550, two layers were placed in the warp direction and one in the weft direction. For the BL200, the difference in warp and weft properties was neglected since the difference in crimp and fibre content was slight (see Table 1).

Sample data obtained from the thermocouples is provided in Figure 2(b) along with the evolution of the resin storage and loss moduli as determined by parallel plate rheometry on the neat resin (Araldite LY5150). The gel time was taken to be the point which G' and G'' cross in a 25 mm parallel plate oscillation experiment. These experiments were carried out at a frequency of 1 HZ and strain of 0.1% on a TA AR2000 rheometer.

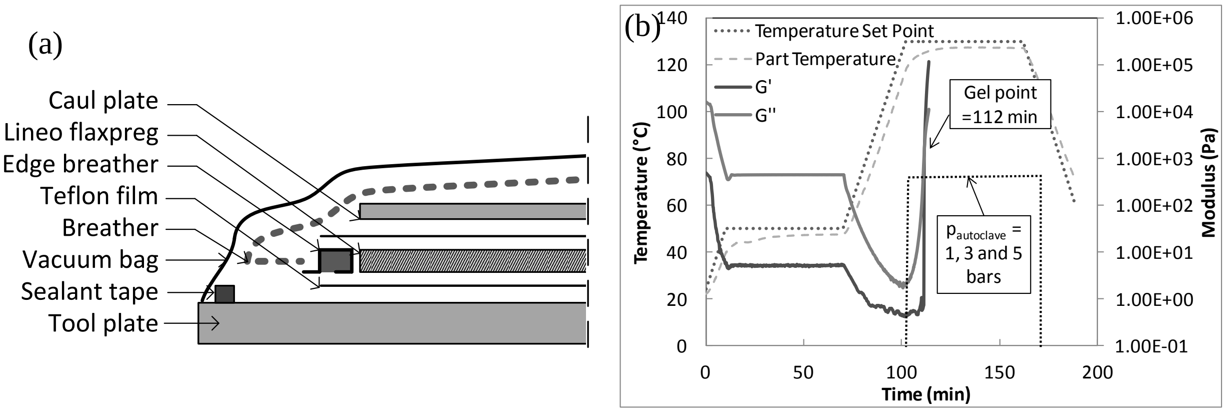


Figure 2. Process parameters (a) temperature, storage and loss modulus evolution and (b) vacuum bag arrangement

3. Mechanical characterization

3.1. Void analysis

Optical microscopy was selected to measure the void content due to the inappropriateness of the commonly used resin-burn off technique (ASTM D2734) for composites reinforced with cellulose-based fibres. Samples of 45 mm in length were cut by water jet from the center of the laminates and the cross-sections were polished. Images of the cross sections at a magnification of 50x were then captured. Upon examination of the images, it was noted that the contrast of the voids with the rest of the composite was very low. To better distinguish them, they were filled in manually with black pixels. The images were then converted to 8-bit greyscale and a threshold function was applied to isolate the filled in regions whose area was finally measured by the software, ImageJ. A summary of the results along with the sample cross sections is shown in Figure 3.

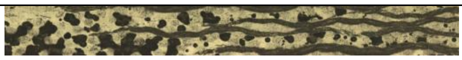
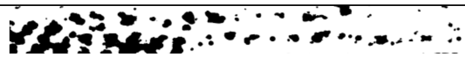
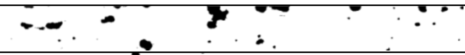
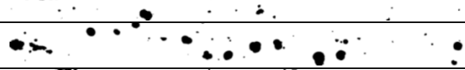
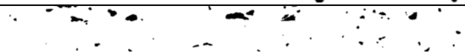
	P (bar)	Polished cross section	Image analysis	Void content (%)
BL200	1			21.4±4.4
	3			3.7±1.1
	5			4.5±0.9
BL550	1			3.5±2.1
	3			0.2±0.1
	5			0.03±0.02

Figure 3. Representative cross-sections of flax/epoxy prepregs processed at 1, 3 and 5 bars.

The image analysis results revealed that, due to the difference in applied pressure, composites of void content ranging from below 1% to over 20% were produced. The relationship between void content and autoclave pressure was non-linear (Figure 4) likely due to the non-linear stress-strain behaviour of the fibre bed.

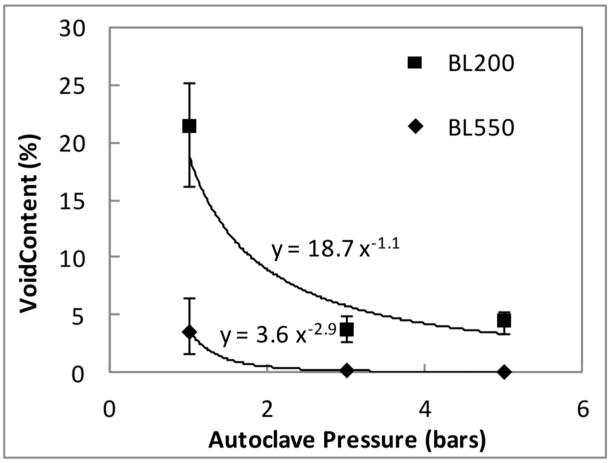


Figure 4. Void content versus applied autoclave pressure

It should be noted, that the analysis of voids was limited to inter-yarn voids. In the case of bast fibre reinforced composites, voids can not only occur in the resin but also within yarns and even the fibres themselves [12]. Therefore, the actual void content is likely higher. The influence of these intra-yarn and intra-fibre voids was beyond the scope of the current study.

3.2. Static tension

Tension tests were carried out in accordance with ASTM D3039 on an Instron servo-hydraulic testing machine with a 100kN load cell and 50 mm extensometer. A specimen area of 25 x 250 mm^2 was selected. Samples were cut by a water jet cutter and E-glass epoxy tabs were bonded to them. A loading rate of 2 mm/min was applied during the tests. The portion of the stress/strain curve up to 0.2% strain was used to calculate the Young's modulus. The results are summarized in Table 2 with representative stress/strain curves in Figure 5.

Prepreg	Autoclave Pressure (bars)	Fibre Volume Fraction[a] (%)	Young's Modulus (GPa)	Strength (MPa)	Elongation (%)
BL200	1	31.4±0.2	8.8±0.4	95±7	1.77±0.15
BL200	3	34.3±0.4	9.7±0.4	102±4	1.74±0.05
BL200	5	35.8±0.4	10.0±0.2	104±3	1.68±0.06
BL550	1	40.6±0.7	9.5±0.2	88±3	1.79±0.05
BL550	3	42.3±0.3	11.1±0.2	101±2	1.68±0.08
BL550	5	44.7±0.6	11.2±0.6	94±2	1.39±0.06

[a] as computed per ASTM 3171 assuming $\rho_{fibre} = 1.53$ g/m^3

Table 2. Summary of tensile testing results

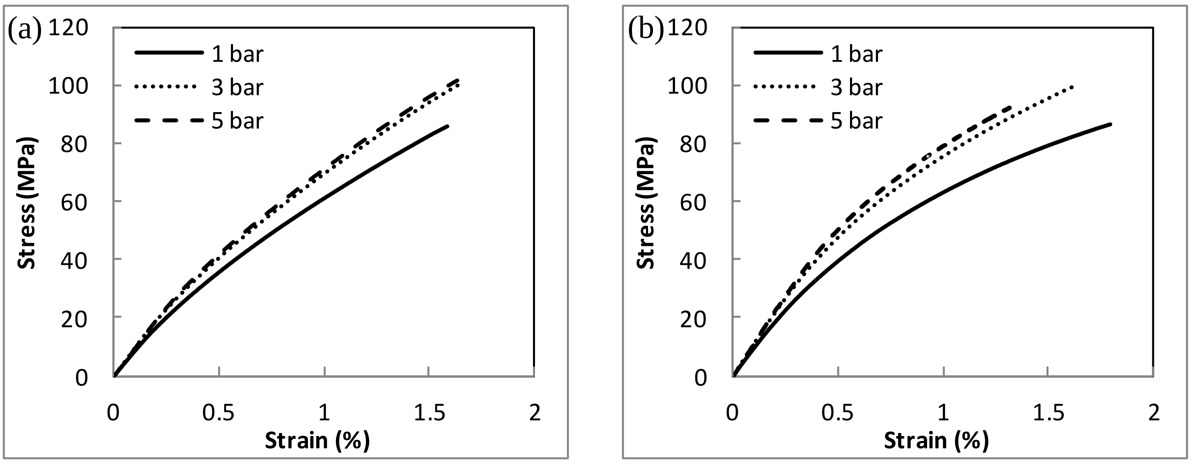


Figure 5. Representative stress/strain curves for (a) BL200 and (b) BL550

From the above results, it first appears that the voids have a relatively large influence on the tensile properties. However, taking into account the increase in fibre volume fraction due to increased pressure, the effect of the voids on tensile properties can be better understood. A normalized modulus and strength were determined by dividing the properties in Table 2 by the fibre volume fractions. The results are plotted in Figure 6.

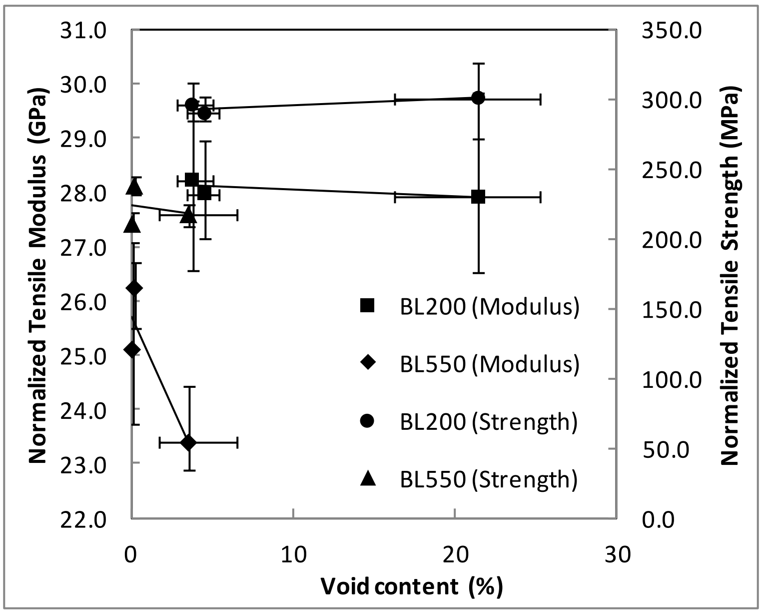


Figure 6. Normalized tensile modulus and strength versus void content of BL200 and BL550 flax/epoxy prepregs processed at 1, 3 and 5 bars

Figure 6 reveals that, in general, the void contents do not have a significant effect on the tensile modulus or strength for the materials systems studied when the fibre volume fraction is taking into account. An exception was the BL550 system which showed a 10% decrease in normalized modulus with increasing void content. The reasons for this result are not yet fully understood so it was deemed inconclusive at the time of this writing and is a topic for future investigation.

It is interesting to note that the BL200 system had a higher normalized modulus. This is likely due to much lower crimp values as given in Table 1 and differences in yarn architecture.

3.3. Interlaminar Shear

Interlaminar shear strength (ILSS) was obtained by the short beam test carried out in accordance with ASTM D2344 on a MTS load testing machine with a 5kN load cell. A sample area of 50 x 150 mm^3 was selected and a load rate of 0.5 mm/min was applied. The span to depth ratio was set to 4. This configuration resulted in a mode of failure of interlaminar shear for both systems as observed under a microscope. Unpolished cross-sections of selected fracture surfaces are shown in Figure 7 along with a plot of normalized interlaminar shear strength versus void content.

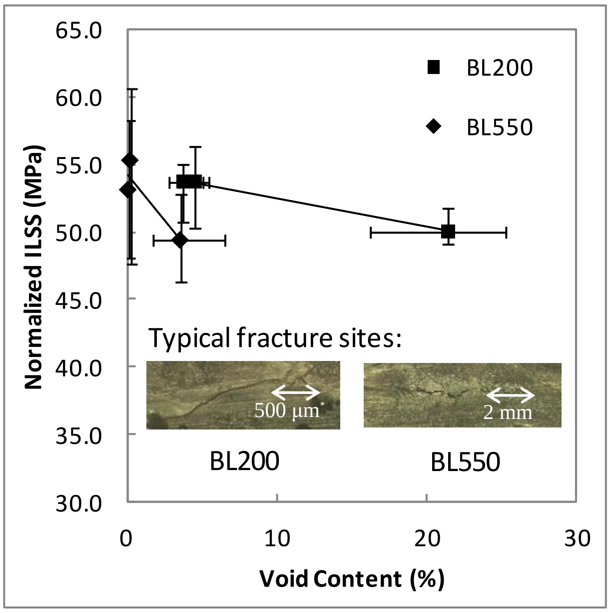


Figure 7. Normalized interlaminar shear strength versus void content for BL200 and BL550 flax/epoxy prepregs processed a 1, 3 and 5 bars

Contrary to the tensile properties, the ILSS exhibited a consistent decrease in properties with increasing void content. For the BL550 system, the normalized strength decreased 11% with a 3.5% increase in void content. The rate of decrease was less pronounced in higher ranges of void content which is typical of observations made by other authors for unidirectional carbon fibre/epoxy composites [6].

3.4. Water absorption

The final property which was investigated was the saturated water absorption. This test was carried out in accordance with ASTM D570 and a sample area of 25x45 mm^2 was selected. To minimize in-plane absorption, the sides of the samples were sealed with a silicone sealer. The weight of the silicone was determined based on the difference in dry sample weight before and after the application of the silicone. The drying process consisted of heating in a convection oven for 24 hours at 50 °C followed by cooling in a desiccant chamber.

The specimens were weighed after submersion in distilled water at 2 and 24 hours. Prior to weighing, they were dabbed with a lint free cloth in order to remove excess liquid. The amount of water absorbed under these conditions and a plot of void content versus effective water equilibrium after 24 hours are given in Table 3 and Figure 8 respectively.

	Processing Pressure (bars)	*Mass gain after 2 hours (%)*	*Mass gain after 24 hours (%)*
BL200	1	5.1±0.6	12.5±1.5
	3	4.7±0.4	9.4±1.3
	5	3.5±0.2	6.4±0.4
BL550	1	2.9±0.3	6.5±0.5
	3	2.3±0.2	4.9±0.4
	5	2.7±0.3	5.3±0.4

Table 3. Mass gain after 2 and 24 hours submersion of cured flax/epoxy prepreg samples processed at varying pressure in water

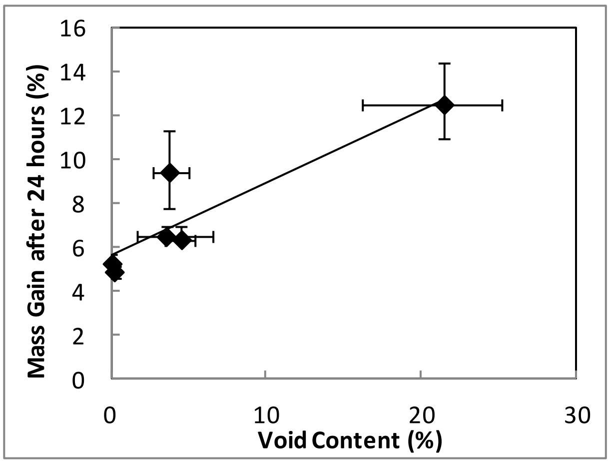


Figure 8. Void content versus mass gain after 24 hours in water absorption test (ASTM D570)

The results indicate a large dependence of water absorption rate on the level of voids. The effective moisture equilibrium after 24 hours increased 33% with only a 3.5% increase in void content. Furthermore, the amount of scatter was generally higher for higher void contents. From Figure 8, it can also be seen that the relationship between the effective moisture equilibrium after 24 hours and void content was in general linear. The outlying point indicates that the level of voids was not well distributed throughout the manufactured plates. It suggests that the void content in the moisture absorption samples was higher than in those used for image analysis for the BL200 system processed at 3 bars.

In summary, the results of the water absorption tests are deemed to be most critical of all mechanical properties investigated in this paper. This is due to the fact that water absorption has been previously shown to in turn lead to a reduction in mechanical properties in some natural fibre-reinforced plastics [13]. Therefore, the achievement of void-free bio-based composites should be a top priority in order for these materials to achieve their full potential.

4. Conclusions

This study demonstrated the negative effect of voids on the mechanical properties of woven flax/epoxy composites. Samples of varying void content were manufactured from two different flax/epoxy prepreg systems. By means of compaction experiments, the voids were shown to originate primarily from resin starvation. The void content of the cured prepregs (as determined by image analysis) were shown to be highly dependent on the processing pressure. The laminates processed at varying pressure were subjected to a mechanical characterization which indicated a reduction in interlaminar shear strength as well as an increase in moisture absorption rate with increasing void content. Tensile properties were shown to be affected to a much lesser extent. For the normalized interlaminar shear strength, a decrease in 11% was noted for a 3.5% increase in voids. At higher void contents this effect was less pronounced. This study emphasizes the importance of developing process technologies for bio-based materials that minimize void content. Furthermore, it provides valuable data on the expected reduction in selected mechanical properties due to the presence of voids in woven flax/epoxy composites.

Acknowledgements

The financial support of the Network for Innovative Plastic Materials and Manufacturing Processes (NIPMMP) and Ministry of Economic Innovation and Exportation Development (MDEIE) is acknowledged. The authors would also like to thank Lineo NV for providing materials and the technical staff of the Catholic University of Leuven for their assistance.

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