A STUDY ON THE INFLUENCE OF PROCESSING PARAMETERS ON DAMPING, ELASTIC AND ULTIMATE PROPERTIES OF COMMINGLED FLAX-POLYPROPYLENE COMPOSITES

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Keywords: biocomposites, flax, compression moulding, vibration, damping, tensile

ABSTRACT

Natural fibres such as flax are a promising replacement for synthetic fibres in polymer composites and achieve comparable specific properties due to their low density. Recent developments in the production of technical flax fabrics allow the use of sustainable natural fibres in the manufacture of structural composite parts. One of the other advantages of natural fibre composites is their high damping properties compared to that of glass and carbon fibre composites. The material properties that can be achieved with plant fibre based composites are highly sensitive to the composite manufacturing process. Although plant fibres have been shown to improve the damping coefficient of composite materials, there are no systematic studies of the effect of the processing conditions on the damping properties in correlation with the stiffness and strength of the composite. In this paper, the damping properties of flax fibre reinforced composites were investigated using the natural frequencies and modal damping obtained from vibration tests. The material selected for the study was flax/polypropylene (PP) fabric supplied by Composites Evolution as Biotex flax/PP. The composite laminates were manufactured by thermocompression using different conditions for temperature, pressure and consolidation time. The damping measurements were achieved on cantilever beams of the flax/PP composite that were clamped in a bench at one end and excited by a short impulse. A laser vibrometer sensor was used to measure the impulse response of the beam and the time signal was converted to frequency response by FFT. It was found that the damping factor is sensitive to the processing conditions and that factors such as porosities, fibre-matrix interface will affect the vibration response of the composite.

1. INTRODUCTION

Natural fibres have been studied as potential replacement to traditional composite reinforcements due to their low density, high specific properties, relative abundance, low cost of raw material, and positive environmental profile [1]. Bast fibres such as flax, hemp, kenaf, jute and ramie exhibit superior flexural strength and elastic modulus and are used in applications with structural requirements as they have high cellulose content and low microfibrillar angle [2]. The specific longitudinal stiffness of flax in tension is higher than that of glass fibres due to the lower density of flax [3] and in the case of plate bending, the specific flexural stiffness of flax composites outperforms steel and aluminium. However, the material properties that can be achieved with the plant fibre composites depends on several factors including the type of the plant fibre, the length of fibres, the resin system used, the composite manufacturing process, and the fabric architecture. Natural fibre thermoplastic composites with short flax fibres and polypropylene manufactured using extrusion had an elastic modulus of 1.5-3.8 GPa and maximum strength of 20-33 MPa [4]. However, it is evident that the structural potential of plant fibres as reinforcing agents can only be realized when the highest reinforcement efficiency is employed and this is typically achieved with twistless long continuous fibres. There is limited literature on the mechanical characterisation of plant fibre based thermoplastic composites reinforced with woven long continuous fibres are used.

Natural fibres used as reinforcement in composites are exposed to high temperatures during processing. Liang et al. [5] noted that the manufacturing conditions of plant fibre-based composites are limited by the thermal degradation of fibres at high temperatures. This is due to the presence of thermally sensitive constituents such as cellulose, hemicelluloses and lignin and the high melting point of thermoplastics. The thermal degradation of flax fibres is dependent on both the temperature of exposure and the duration. Gourier et al. [6] studied the effect of thermal cycles on the mechanical behaviour of elementary fibres of flax and found that modification of the fibre structure and of the interactions between components occurred at high temperatures such as 250° C. In order to minimise the damage of the fibre, Van de Velde and Baetans [7] recommended that composite production temperatures higher than 180° C have to be avoided, unless the duration is short. It was reported that 15 minutes of exposure of flax to 180° C results in lower residual tensile stress and strain than two hours exposure to 120° C. Nassiopoulos and Njuguna [8] revealed that deformational behaviour of flax-PLA composite changes from brittle to more ductile-like characteristics with increasing temperature and concluded that the material properties exhibited a strong dependence on temperature. Bourmaud et al. [9] studied the effect of the processing temperature on mechanical performance of unidirectional flax fibre composites in polyamide 11 matrix and found that a thermal cycle of 8 minutes at 210° C reduced both the stiffness and strength of the composite. John and Anandjiwala [10] also noted the low thermal stability of natural fibres and recommended the use of polypropylene as matrix as it has a relatively low processing temperature. Van de Velde and Kiekens [11] studied the effect of processing parameters, temperature and time, on the mechanical properties of non-woven flax/PP composites and recommended that temperature of 200° C was sufficient to obtain good mechanical properties. In this paper, the effect of processing conditions on the mechanical properties of woven commingled fabric of flax fibre and polypropylene is studied to improve the understanding of the process-structure-property relationship of these composites.

One advantage of natural fibre composites compared to synthetic fibre composites is their damping properties. Duc et al. [12] reported that the damping of flax fibre reinforced composites was higher than that of carbon and glass fibre reinforced composites, with unidirectional flax fibre epoxy composite exhibiting a 100% increase in loss factor compared to glass fibre reinforced epoxy. Typically, damping and stiffness are inversely related with low damping value indicating high elasticity and high damping suggesting a material with high, non-elastic strain component. Similarly, an improvement in the fibre/matrix interface results in the reduction of damping factor, since mobility of the molecular chains at the fibre/matrix interface decreases. El Hafidi et al. [13] noted that damping is induced by several microscopic mechanisms, such as viscoelastic elongation of the matrix and/or fibres, and local friction at the interface between both components. For the natural fibre composites such as flax fibre reinforced polymer composites, the friction between elementary fibres inside bundles represents an additional mechanism of energy dissipation, which also contributes to the overall damping. The entanglement of the fibres, void in the lumen, heterogeneity of the cell walls and reversible hydrogen bonding between the different components of the cell walls are also considered important contribution to the intrinsically good damping properties observed in plant fibres [14].

There are many different definitions and ways of measuring damping such as the loss factor, the quality factor, the specific damping capacity, the logarithmic decrement or the damping ratio. There is limited literature on the damping properties of flax-PP composites, especially on the effect of processing conditions on the vibration response of composites. Duc et al. [12] used DMA device in the single cantilever mode to characterise the damping behaviour of the composites under flexural loading. Le Guen et al. [14] quantified the damping coefficient of flax fibre-reinforced composites and flax-carbon hybrid composites and used rule of hybrid mixtures to study the impact of reinforcement hybridisation on the damping coefficient, and tensile and flexural elastic modulus and strength of hybrid composite. Le Guen et al. [15] also used vibration in longitudinal and flexural modes and found that improved damping performance due to polyglycerol additives. This was explained by the formation with polyglycerol of hydrogen bonded bridges between concentric layers of cell walls, causing friction between the two layers through stick–slip molecular motion. Monti et al. [16] studied the flexural vibration response of bio-based sandwich and laminates beams using free vibration tests and analysed the damping property of different configurations of unidirectional and cross-ply composites as well as sandwich structures with different core thicknesses. In this paper, the flax thermoplastic composite is manufactured with different processing parameters of temperature, pressure and duration. The different composites are then tested in uniaxial tension to measure the elastic and ultimate properties. Free vibration tests are conducted on cantilever beams of selected composites and the damping properties are compared statistically to investigate the effect of processing conditions.

2. MATERIALS AND METHODS

2.1 Manufacturing of composite samples

The woven commingled fabrics of Flax/PP were supplied by Composites Evolution as Biotex Flax/PP with surface density of 400g/m^2 and made from twistless natural flax fibre and polypropylene (PP) fibre in a balanced 2x2 Twill architecture. The relatively low melting point of polypropylene makes it a more suitable matrix for flax fibres as thermal characterisation analysis by TGA show that the fibres begin to degrade at temperatures higher than 200 °C. The flax fibre volume fraction of the fabrics was 40%, with an estimated thickness of 0.3-0.35 mm per ply. The composites were fabricated using a compression moulding method in which the fabrics are moulded into rigid fibre-reinforced thermoplastic composite parts by applying heat and pressure to melt the thermoplastic, wet-out the flax and consolidate. Eight plies of the flax/polypropylene commingled fabric were cut to 280 mm x 280 mm and placed between the two rigid steel plates in a hydraulic press. The fabrics were pre-dried before composite production to remove the excess moisture. A schematic diagram showing the upper and lower plate of the mould and the woven fabric and the thermopress system (induction press system EdyCo platform, Mines Albi) used for the fabrication of the plates is shown in Figure 1.

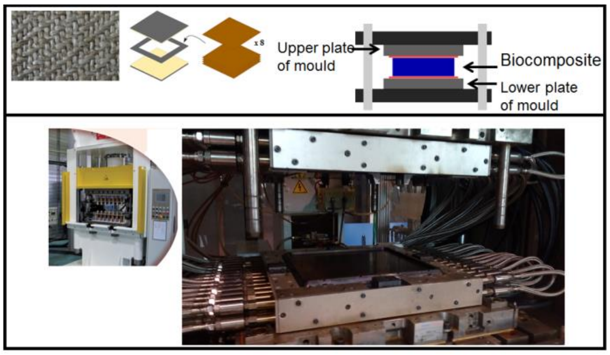


Figure 1. Schematic diagram of thermocompression process and the induction press used for manufacturing flax/PP composites

The mechanical properties of the thermoplastic composite are highly dependent on the processing conditions. Therefore several processing parameters were considered for this study. All samples were prepared in the thermopress following a specific cycle comprised of three phases: an initial heating period, an isotherm (or constant temperature phase) and the final cooling down phase. The composite plates were manufactured using different experimental parameters for temperature, pressure and consolidation time.

The conditions were chosen based on thermal analysis of the flax-PP fibres. The peak melting temperature of the polypropylene was found to be approximately 168 °C. The chosen temperature should be sufficiently high as to completely melt the PP matrix. At higher temperatures, the viscosity of the resin is low allowing the flow of the matrix and perhaps better diffusion. However, at high temperatures, there is degradation of the flax fibre. 18 composite plates were manufactured and the parameters used are summarised below in Fig 2. The nomenclature for the samples follows the rule **xC_ym_zb**, which corresponds to the temperature in °C degrees, consolidation time in minutes and pressure in bars, respectively.

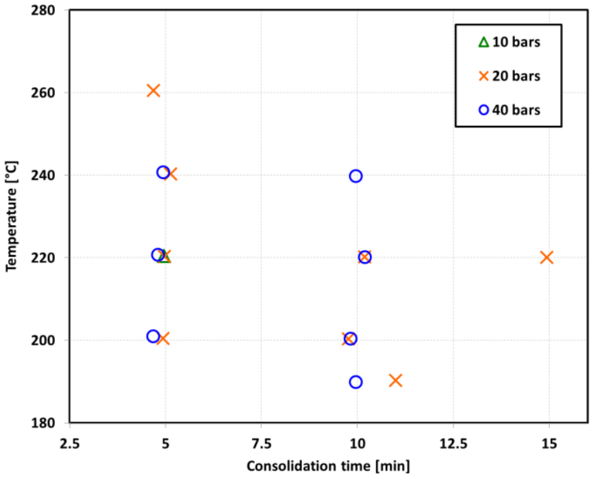


Figure 2. Summary of experimental conditions used in press for fabrication of flax/PP composite

The temperature of the press, the pressure applied, generator power applied for the heating and the platen position were recorded for each condition. A typical processing cycle for the manufacturing of flax/PP composite is shown in Figure 3. It can be seen that the isotherm corresponding to this cycle is at 200 °C. Maximum power is applied to the press for the heating cycle and power is shut off before the press reaches the assigned temperature as there is a delay. The temperature is allowed to stabilise for 2 minutes before the pressure (20 bars in this case) is applied and the clock is started to measure the duration of the consolidation phase. At the end of the consolidation phase (10 minutes in this case), the cooling system is turned on and the temperature is allowed to decrease rapidly until 140 °C. The cooling rate is reduced at this temperature to aid the crystallisation of the polymer. The pressure is maintained during the entire cooling cycle and it can be seen from the platen position that there is thickness reduction during the consolidation.

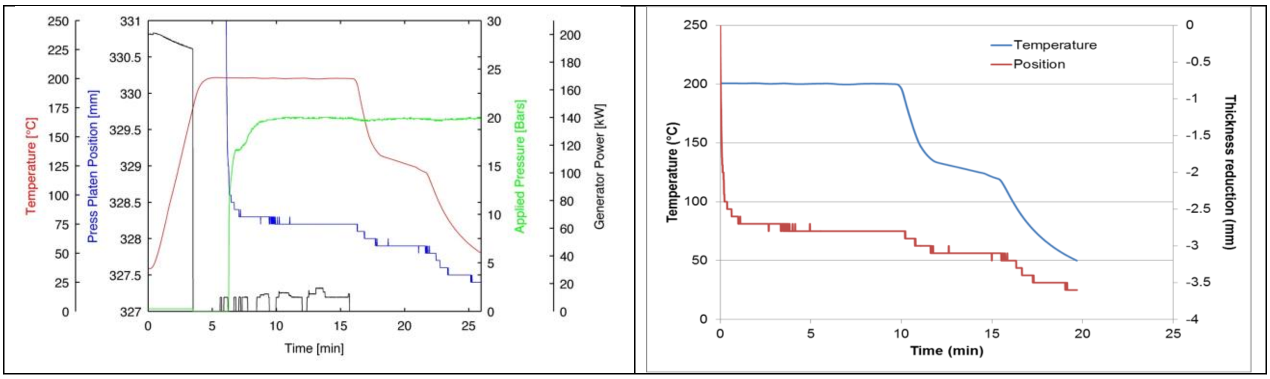


Figure 3. Typical processing cycle data from induction press

A typical SEM micrograph of the flax fibre reinforced PP matrix composite manufactured using the induction press is shown in Figure 4. The composite specimen in the figure corresponds to plate manufactured at 190°C and consolidation time of 10 minutes. During the compression moulding, the PP fibres in the commingled yarn melts and diffuses within the flax yarns and bundles before the freezing of the matrix during the cooling phase. It can be seen in the left that the composite consists of both longitudinal and transversal fibres (warp and weft). Flax fibres are typically in the form of fibre bundles with individual elementary fibres visible in the Figure 4(b). We can also note the microstructure of the fibre with lumen and cell walls as well as some macro porosities in the composite.

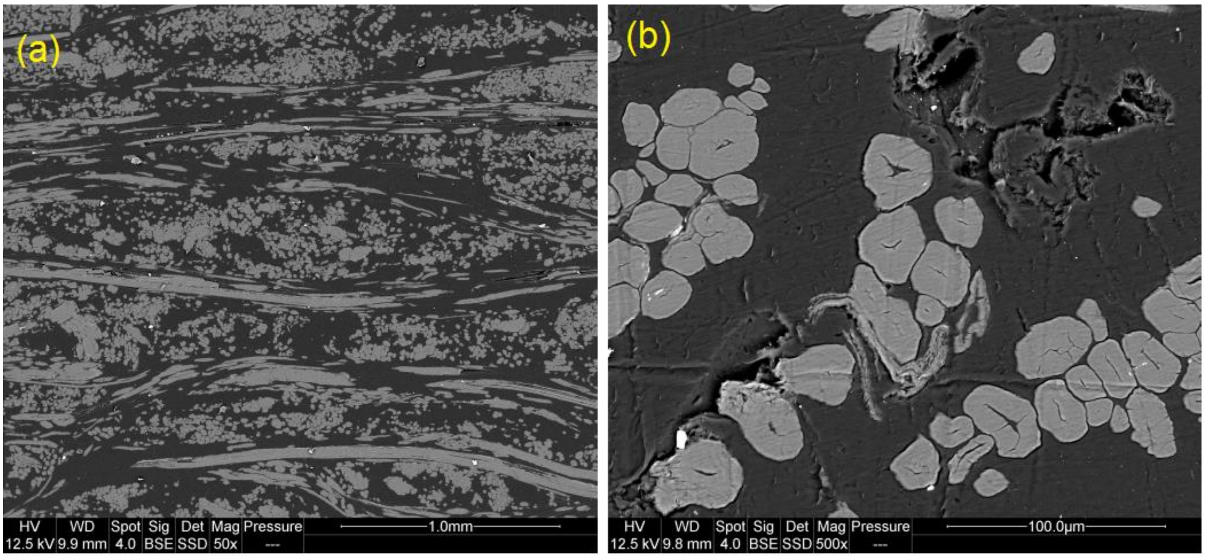


Figure 4. SEM image of fracture surface for composite manufactured at 240°C

2.2 Tensile testing of composite samples

A Zwick Z010 testing machine, shown in the Figure 5 was used to perform tensile tests based on the ISO 527-4 standard. These tests were all performed in room temperature in a controlled environment (ca. temperature of 25°C and 50% humidity) after having been conditioned for at least three hours in a humidity controlled oven (temperature of 30 C and 3.5% humidity). The width and thickness of each sample was measured before every test (ca. 18 mm-21 mm and 2.4 mm-2.8 mm, respectively) and the length between the fixing jaws was 110 mm (length of actual samples: 165 mm). Five samples were tested per condition and each test was conducted in two stages; a first stage at a crosshead speed of 1 mm/min using an extensometer to determine the tensile modulus (calculated by taking the slope of a linear trendline of the stress-strain curve between a displacement of 0.05% and 0.3%) and a second stage at a cross-head speed of 5 mm/min until rupture to determine the tensile strength. A typical stress strain curve showing the two loading phases is shown in Figure 5.

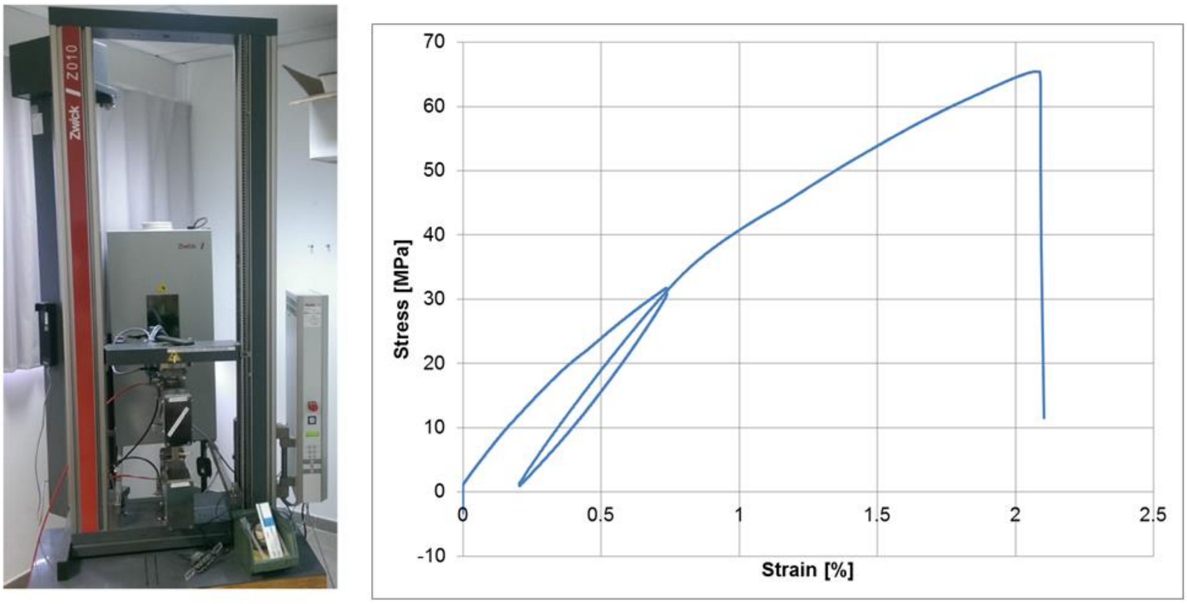
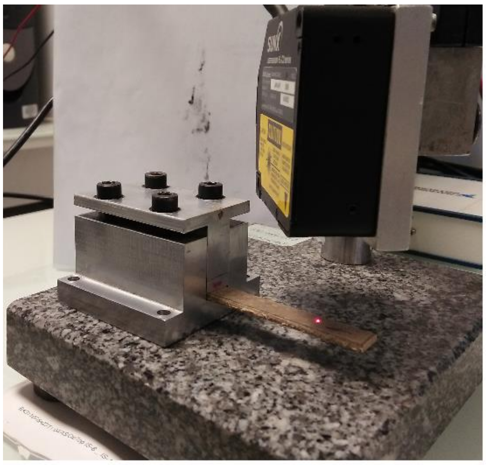


Figure 5. Tensile testing setup and typical stress - strain curve of flax/PP composite

2.3 Vibration testing of composite samples

The damping measurements were achieved on cantilever beams of the flax/PP composite that were clamped in a bench at one end and excited by a short impulse. Initially, three processing conditions corresponding to the extreme cases were chosen for the vibration tests. The setup used for the vibration test and typical timeform signal is shown in Figure 6. The dimensions of the beam were 80 mm in length and 10 mm width. However, some part of the beam was clamped in the fixture so that 60 mm was the free length of the beam. The clamping torque of 2 Nm was kept constant by using a torque wrench. The vibration tests were performed at room temperature and humidity. A short impulse was applied at the free end of the beam, similar to the method used by Regazzi et al. [17]. For each beam, the tests were repeated at least five times and the results were averaged. A SunX laser vibrometer sensor was used to measure the transient response of the beam and the time signal was converted to frequency response by FFT. The advantage of the laser sensor was that it is a non-contact measurement and there is no added mass on the composite beam (contrary to the use of an accelerometer). The eigen-frequency and the damping factor of the fundamental flexural mode of vibration were determined using a rational polynomial fit of the frequency response function in Modalview software.



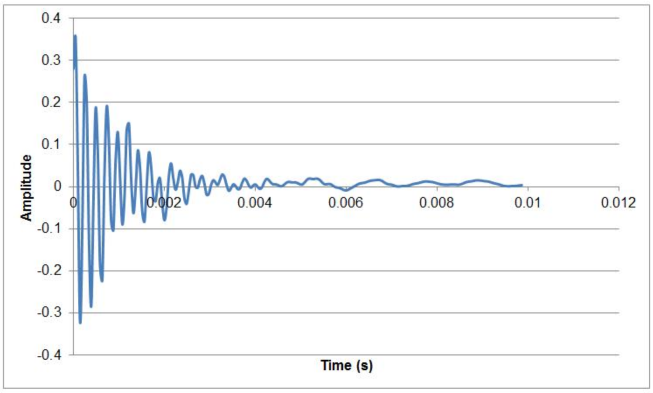
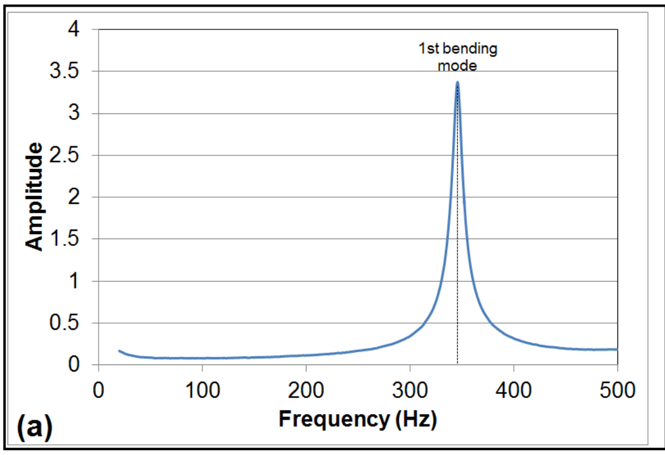


Figure 6. Setup for vibration test and typical time signal measured by laser vibrometer

The time signal is converted to the frequency domain. A typical frequency response with the first flexural mode identified for a flax-PP beam is shown in Figure 7. It can be seen that the corresponding resonant frequency is approximately 350 Hz. The real part and the imaginary part of the complex frequency response function were plotted against each other to provide a Nyquist plot, which is nearly a complete circle as expected in the vicinity of a resonance peak.



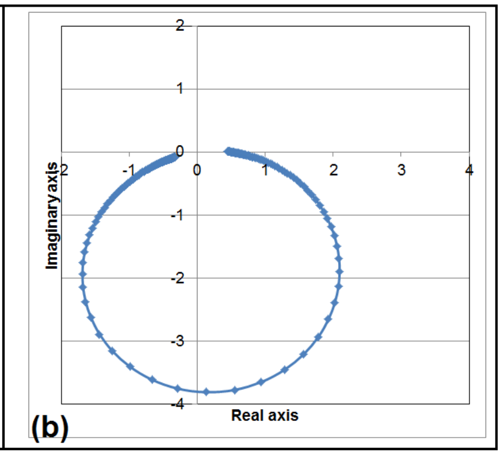


Figure 7. Typical frequency domain signal with first bending mode and Nyquist plot

3. RESULTS AND DISCUSSION

A box plot showing the modulus and strength for selected subset of processing cycles is shown in Figure 8. The fibre volume fraction of the composites varied from 39% to 41.1%. It can be seen from the variation of the modulus and strength that there is a positive effect of the fibre content on the mechanical properties. The increase in the modulus of the composite with higher fibre weight fraction is an indication of stiffening due to the fibres, while the increase in the strength of the composite is explained by the fact that the addition of flax fibres leads to uniform and effective stress transfer within the composite.

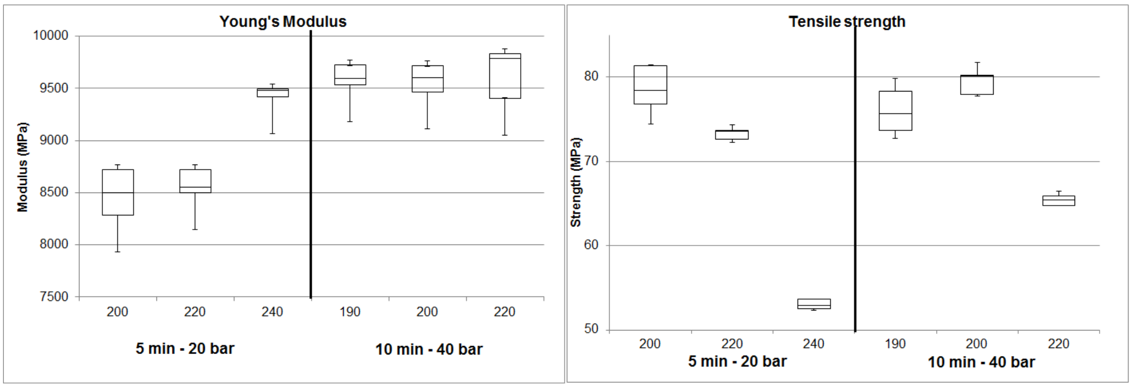


Figure 8. Boxplot of tensile modulus and strength for different processing cycles

Figure 9 shows the variation of both the tensile strength and modulus in Ashby style chart to study the effect of the processing conditions. It can be seen that the effect of the temperature on the initial modulus is negligible but there is significant effect on the tensile strength of the composite. The composites manufactured at 240°C have the lowest strength with almost 35% reduction in strength compared to composites manufactured at 200°C. According to Bourmaud et al. [8], at higher temperatures there is a significant modification of the flax fibres such as its biochemical composition and macromolecular arrangement. These modifications affect the cellulose micro fibrils, and therefore lead to a decrease of Young's modulus and strength.

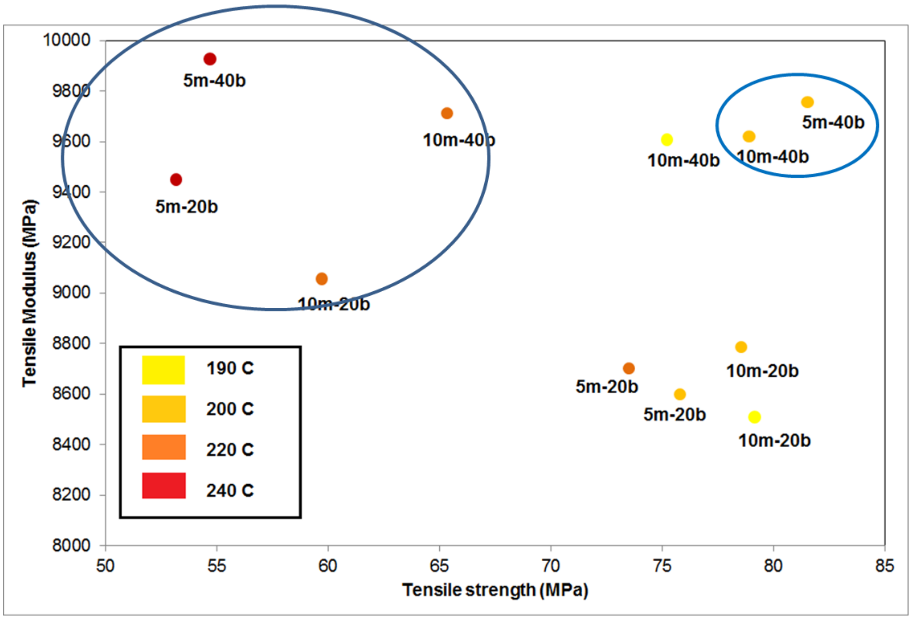


Figure 9. Ashby chart of modulus and strength for flax/PP composite

Figure 10 compares the stress-strain curve corresponding to the two extreme cases of 190 C and 240 C. It can be seen that the initial modulus is almost identical for the two composites. However, due to thermal degradation, the flax fibre is more brittle and this results in the composite failure. It can be seen that the strain properties are more influenced by thermal treatment and the failure strain for the composite manufactured at 240 C is 1.3% compared to the 2.4% for 190 C composite.

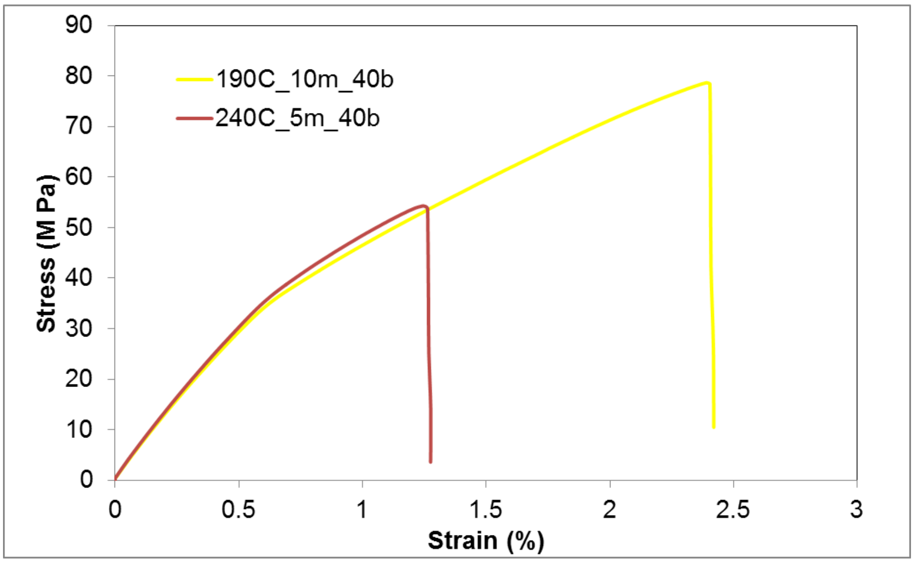


Figure 10. Comparison of extreme cases (190 C and 240 C composite)

Initially, two composite samples were selected for the vibration tests corresponding to the two extreme cases for the tensile tests. However, it can be seen that the initial modulus is almost identical for the two extreme cases. It was hypothesised that since the vibration response is measured at low strains, there may not be a strong effect of processing conditions on damping. Therefore, a third sample corresponding to the lowest modulus in the Ashby chart (200C_5m_20b) was also chosen.

The frequency-amplitude graph for each sample was analysed using Modalview software. The frequency range around the resonance peak was chosen according to the classical 3db method and an automatic polynomial fit was applied to extract the eigen frequency and damping factor. Table 1 shows the results of the damping factor (in %) measured for composites manufactured at 190C_10m_40b. Five samples were tested and multiple trials were conducted for each sample. It can be seen that the variation of the damping factor for the multiple trials on same sample is very low (standard deviation of 0.01 to 0.02). This confirms that the vibration measurement system that is proposed is highly repeatable. However, it can be seen that the variation between the samples of supposedly the same material is rather high, varying from 1.43 to 1.54 (standard deviation of 0.04). This can be explained by the fact that the damping factor is highly sensitive to local microstructure and presence of voids, any variation in the fibre volume fraction or in the intrinsic properties of the components will have a strong influence on the measured damping. It was also found that while the eigen frequency was very stable for the measurement, the damping factor was influenced by the impulse magnitude and by the selection of the frequency range for the calculation. It is also interesting to note that the dynamic modulus assessed from the eigen frequency by using the classical Euler-Bernoulli formula are well correlated to the modulus measured by the tensile tests. For example, the tensile modulus of 240C_5m_40b is 9.8 GPa and the dynamic modulus for the same sample is 9.6 GPa.

Table 1. Damping factor in (%) for 190C_10m_40b composite

	Sample 1	Sample 2	Sample 3	Sample 4	Sample 5
Trial 1	1.57	1.44	1.43	1.45	1.47
Trial 2	1.52	1.45	1.42	1.46	1.43
Trial 3	1.53	1.49	1.43	1.48	1.43
Trial 4	1.53	1.47	1.44	1.49	1.45
Trial 5	1.53	1.47	1.42	1.47	1.46
Mean	**1.54**	**1.46**	**1.43**	**1.47**	**1.45**
SD	**0.02**	**0.02**	**0.01**	**0.01**	**0.02**

The mean damping factor, which is taken as the mean of the means of the different composite beams, was found to be 1.47, 1.50 and 1.39% for the 190C, 200C and 240C samples respectively. Figure 11 plots the damping factor measured from the vibration tests and the Young's modulus measured from the tensile tests for the three samples. It can be seen from the error bars that there is a large scatter in the properties of the composite. A statistical comparison of the composites is necessary to identify the degree of correlation

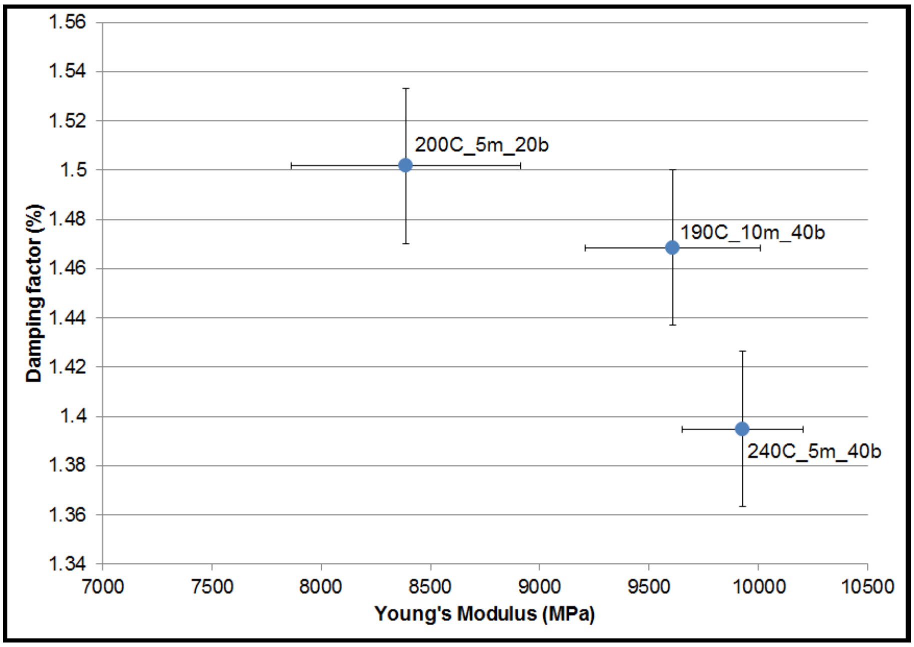


Figure 11. Damping factor vs. Modulus for the three composite samples (190C, 200C and 240C)

A Wilcoxon signed-rank test was performed for the comparison of the three composite samples. The Wilcoxon method is a nonparametric test that can be used to determine whether two dependent samples were selected from populations having the same distribution. This test is recommended as an alternative to the paired Student's t-test for small sample sizes and when the population cannot be assumed to be normally distributed. A p-value of less than 0.01 is considered highly statistically significant while a p-value higher than 0.05 is considered not statistically significant, meaning that there is no effect of the processing conditions.

For the comparison of the 190C and 240°C samples, it was found that the p-value was 2.85E-05 which clearly shows that there is an effect of the processing temperature. However, the comparison of the 190C and 200C samples did not have the same result, with a p-value of 0.0734. Contrary to the initial hypothesis that composites with similar modulus will have similar damping factors, these results show that there was an effect of the higher processing temperatures on the damping factor. We can see that the damping factor is reduced for the samples manufactured at 240 ºC. Therefore the thermal degradation of the fibre and the change in the microstructure such as fibre-matrix interface and the fibre volume fraction will impact the vibration response of the composite. In the future study, the damping parameters of the composites manufactured at all the different conditions are correlated to the elastic properties.

5. CONCLUSIONS

In this study, thermoplastic composites from flax/PP commingled fabrics were manufactured using thermocompression. The influence of the processing conditions on the elastic and ultimate properties from uniaxial tensile test was studied. Additionally, the vibration damping properties were measured using free vibration analysis of cantilever beams with laser vibrometer. It was found that the temperature, pressure and duration of the compression moulding cycle has a strong effect on the modulus and strength of the composite. The strength of the composite was particularly affected by high temperatures which resulted in fibre degradation. The damping properties of the composite from the vibration test also show that it is sensitive to the microstructural parameters such as porosities and fibre volume fraction. In the future study, the damping parameters of the composites manufactured at all the different conditions are correlated to the elastic properties.

ACKNOWLEDGMENTS

This project was funded by the European Union's Horizon 2020 Programme (HERMES project, grant agreement n° 636520).

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