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The mechanical properties of woven tape all-polypropylene composites

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Abstract

The creation of highly oriented, co-extruded polypropylene (PP) tapes allows the production of recyclable "all-polypropylene" (all-PP) composites, with a large temperature processing window (>30 °C) and a high volume fraction of highly oriented PP molecules (>90%). This paper describes all-PP composites made from woven tape fabrics and reports the tensile and compressive properties of these, with reference to composite processing conditions and compares these mechanical properties to those of commercial alternatives.

Keywords: A. Recycling; A. Tape; D. Acoustic emission; A. Fabrics/textiles

1. Introduction

In a series of academic theses [1–3], composite materials in which both the fibre and the matrix are based on polypropylene (PP) have been described. The behaviour of these composites will be presented in a range of publications. These so-called "all-PP" composites are designed to compete with traditional thermoplastic composites such as glass fibre reinforced PP, so must possess comparable or superior mechanical properties. One of the main advantages of all-PP composites is the enhanced recyclability which is achieved by using the same polymer for both fibre and matrix phase of the composite. Unlike glass fibre reinforced PP (GFRPP) composites, all-PP composites can be entirely melted down at the end of the product life for recycling into PP feedstock. This enhanced recyclability is desirable to satisfy new environmental legislation which is currently targeting high volume industries particularly the automotive industry [4].

The mechanical properties of PP can be improved in a given direction by orienting the polymer molecules in the direction of loading. Numerous routes have been described to achieve this molecular orientation such as melt spinning, gel (solution) spinning, and solid state deformation [5]. However, in these cases, orientation is achieved in only one direction and while this "self-reinforcement" is suitable for fibre applications, it has limited potential to other applications that require load bearing in more than one dimension. Thus, to apply the improved mechanical properties achieved through molecular orientation, alternative/additional routes must be devised to produce PP which possesses molecular orientation in two- or three-dimensional geometries. Reported routes include the roll drawing of PP sheets [6–8], flow-induced crystallisation during sheet extrusion [9,10], uniaxially oriented PP films welded together in multidirectional plies [11], stacking of oriented PP fibres together with isotropic propylene–ethylene copolymer films [12], and stacking biaxially oriented PP films, which have been co-extruded with a propylene–ethylene copolymer film to facilitate bonding at temperatures below the melting temperature of the films [13]. However, while roll drawing and flow induced crystallisation produce molecular orientation in two-dimensional geometries, molecular orientation is still limited to one direction: the rolling, or extrusion direction. Bidirectional properties

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can be achieved by welding alternatively rotated stacks of uniaxially oriented sheets together, and tensile moduli of 5 GPa and tensile strengths of 150 MPa have been reported using this method [11].

However, an alternative route to create reinforced polymer laminates by welding highly oriented PP fibre bundles or PP tapes by careful heating has been devised at University of Leeds, UK [14–19]. This so-called "hot compaction" route provides an interesting alternative to achieve entirely PP sheet structures with high volume fractions of reinforcement and hence high mechanical properties (a tensile modulus of 5.1 GPa and a tensile strength of 182 MPa have been reported [20]). The main complication with this process route is that this process is limited by a small temperature processing window. Excessive temperature results in relaxation and hence a loss of molecular orientation, while insufficient temperature leads to a poor interfacial bonding between fibres/tapes. This small temperature processing window reduces the versatility of this processing route. However, the temperature processing window is adequate for carefully controlled consolidation of biaxially oriented laminates (for example in a hot press or double belt press [21]), which can subsequently be thermoformed.

However, the drapability of unconsolidated fabrics makes a fabric structure more desirable than consolidated sheet precursors for forming processes. The focus of the research reported in this paper is to create all-PP laminates with excellent bidirectional mechanical properties, a high volume fraction of reinforcement and a large temperature processing window which allows direct forming of three-dimensional geometries for unconsolidated fabric precursors. This can be achieved by consolidating fabrics woven from a highly oriented, co-extruded PP tape. The tape is composed of three layers: a central highly oriented PP homopolymer core, with a thin skin layer (5.5% tape thickness) of a propylene–ethylene copolymer on either side. After co-extrusion, the tape is highly drawn in a two stage, solid state drawing process to achieve high modulus and strength by molecular orientation. The copolymer skin layer possesses a melting temperature below that of the homopolymer core, thus facilitating bonding of the tapes at temperatures below the melting temperature of the core. Thus, the fabric can be consolidated into a laminate without significant thermal relaxation of the highly oriented, high modulus homopolymer core [22].

The mechanical properties of any highly oriented polymer structure is dependant on microstructure and so thermomechanical history [23,24]. The consolidation of these co-extruded PP tapes into a coherent composite material requires heating, but highly oriented polymers experience shrinkage forces in the drawn direction upon heating due to thermal energy allowing relaxation to a higher entropy [25–27]. Thus, there is always a risk that shrinkage and relaxation may occur during composite consolidation. If complete relaxation is allowed to occur, then the high tensile properties achieved during the tape drawing process will be reduced and so not be transferred to the resulting composite. In order to limit or prevent this relaxation, it is important to carefully define the parameters used to consolidate a collection of tapes into a load bearing structure. The composite production parameters considered here are temperature and pressure applied during consolidation, although the effect of temperature on the interfacial properties of all-PP composites have been assessed [2] and will be the subject of a subsequent paper. In order to make a coherent all-PP composite that possesses a strong interfacial strength and retains the high mechanical properties of the constituent tapes, the following processing conditions must be applied:

(i) Sufficient pressure to prevent shrinkage by lateral constraining and encourage good interfacial contact for bonding, but not enough to encourage flow of homopolymer core, which would result in a loss of properties.
(ii) Sufficient temperature to melt the copolymer layer to enable fibre bonding, but not so much as to encourage shrinkage, relaxation or melting of the oriented homopolymer phase.

This theoretical processing window is shown schematically in Fig. 1, although conflicting processes complicate the exact geometry of the window. As consolidation temperature is increased, viscosity of the copolymer decreases and so a lower minimum pressure would be predicted for adequate consolidation. However, as temperature is increased, shrinkage forces in the tapes increase [28] and so *greater* pressure is required to prevent tape shrinkage. As pressure is increased, the maximum applicable temperature decreases since at high pressures, flow and hence relaxation, is encouraged, and so a combination of high temperature and pressure aids relaxation by lateral flow of the composite in the mould.

Even though Fig. 1 presents a clearly defined processing window, in reality this is not the case. Within the processing window, mechanical properties are expected to vary as

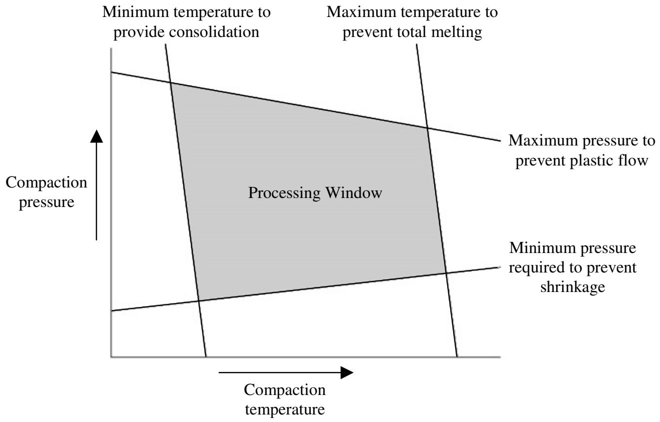


Fig. 1. Temperature–pressure processing window for all-PP composite consolidation.

the microstructural mechanisms of shrinkage and lateral flow compete. By creating a range of composite specimens with woven fibre architectures over a range of processing temperatures and pressures, the effect on mechanical properties can be established. A previous study [2,29] established that the temperature processing window of unidirectional all-PP laminates exceeded 20 °C without a significant effect on tensile modulus or strength of the laminates. The aim of this paper is to characterise bidirectional all-PP composite laminates by investigating the tensile and compressive strength and tensile modulus of all-PP composites made from consolidated woven tape fabrics, as a function of compaction temperature and pressure.

2. Specimen preparation

The experimental tape used throughout this paper is a co-extruded three layer tape, with an A:B:A (copolymer:homopolymer:copolymer) structure, was manufactured at Lankhorst Indutech BV, the Netherlands. The tape properties are summarised in Table 1. This tape was subsequently woven into a balanced, plain weave fabric possessing an areal density of ~100 g m^{-2} by BW Industrial BV, the Netherlands.

Layers of this fabric are cut into square pieces measuring 180 mm × 180 mm, and stacked in a close fitting mould up to a maximum depth of 7 mm. The mould is then subjected to heat and pressure to consolidate the fabric plies into a biaxially reinforced composite laminate. To establish the effect of varying compaction pressure on the mechanical properties of all-PP composites, two different techniques are applied to compact the woven composite laminates. For pressures in the range 1–15 MPa (~10–150 bar), a conventional hot press is used, while for consolidation with a pressure of 0.1 MPa (1 bar), vacuum bagging technology is used. The nomenclature of specimens produced together with processing route and conditions is shown in Table 2.

To consolidate all-PP composites in a hot press, the fabric plies are placed in a mould, which is positioned between the platens of a 500 kN manual hot press equipped with electronically controlled heating platens and water-cooled cooling platens. Due to the inaccuracy of pressure control of the hot press at very low pressures, specimens compacted at 0.1 MPa (1 bar), are consolidated by vacuum bagging, rather than hot pressing. To manufacture vacuum bagged specimens, a mould is sealed in a vacuum bag connected to a vacuum pump that applies a negative pressure inside the vacuum bag, and so atmospheric pressure (~0.1 MPa) is used to compact the plates (see Fig. 2). The entire bag is placed inside a circulated hot air oven and consolidation occurs.

Table 1
Mechanical properties of PP precursor tape for all-PP composites

Width [mm]	2.15
Thickness [μm]	60
Density, ρ [g cm^{-3}]	0.732
Draw ratio, λ	17
Composition [A:B:A]	5.5:89:5.5
Tensile modulus [GPa]	15
Tensile strength [MPa]	450
Strain to failure [%]	7

Table 2
Summary of all-PP laminates created for tensile testing

Specimen	Compaction temperature (°C)	Compaction pressure (MPa)	Production route
W140L	140	0.1 (Low)	Stacked plies, vacuum oven
W140M	140	1.2 (Med.)	Stacked plies, hot press
W140H	140	12.4 (High)	Stacked plies, hot press
W160L	160	0.1 (Low)	Stacked plies, vacuum oven
W160M	160	1.2 (Med.)	Stacked plies, hot press
W160H	160	12.4 (High)	Stacked plies, hot press

The time–temperature and time–pressure profiles for both methods are identical. It is important to note that pressure is applied before heating starts and then remains constant during consolidation until the specimen has been cooled to a release temperature (approximately 40 °C). The time–temperature and time–pressure profiles for both methods are shown in Fig. 3. The specimen is placed in

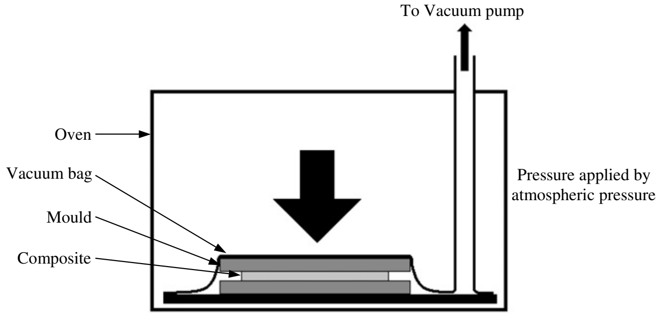


Fig. 2. Schematic set-up for consolidation of all-PP composites in a vacuum oven for consolidation at low pressure (vacuum bagging).

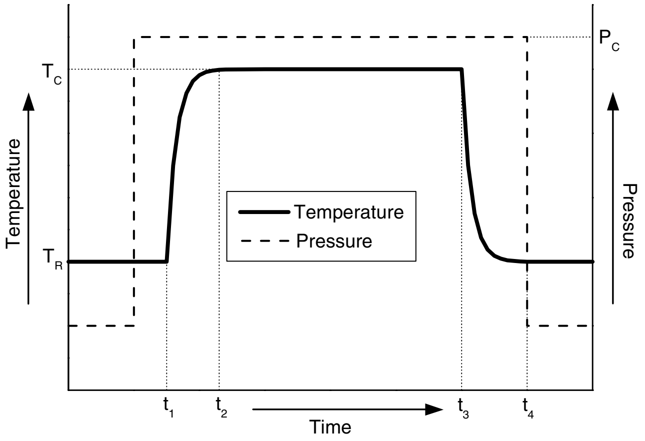


Fig. 3. Time–temperature and time–pressure profiles during consolidation of all-PP composites.

the hot press or vacuum oven at the release temperature (T_R) of 40 °C. At this temperature, there is no loss of orientation in the absence of applied pressure. It is important to note that the compaction pressure (P_C) is applied before heating starts and then remains constant during consolidation at the compaction temperature (T_C) until the specimen has been cooled to T_R. Heating a specimen from T_R to T_C ($t_2 - t_1$) takes approximately 10 min while cooling from T_C to T_R ($t_4 - t_3$) is possible in approximately 5 min, although these durations are specific to the equipment used. The consolidation time ($t_3 - t_2$) is kept constant at 10 min, while the entire cycle from insertion of the mould into the hot press or vacuum oven, until removal of the consolidated plate ($t_4 - t_1$) has a duration of approximately 25 min. Table 3 summarises the parameters of time, temperature and pressure applied in the creation of the all-PP composites described in this paper. The application of pressure acts to consolidate tapes, encourage copolymer flow and prevent shrinkage by lateral constraining.

All specimens for mechanical testing were cut from laminates consolidated as described above. Test specimens were cut by using a razor blade, to the dimensions listed in Table 4. The dimension of the specimens was limited in some cases by the dimensions of the mould used to manufacture the specimens. For the microscopy images presented in Fig. 13, the all-PP composites are cut with a glass knife, after cooling by liquid nitrogen, using a Leica RM2165 microtome. The images are captured using a Zeiss Stemi SV11 stereomicroscope equipped with a high resolution, digital camera.

Table 3
Consolidation parameters for the production of woven tape all-PP composites

Consolidation method	Vacuum oven	Hot press
Release temperature, T_R [°C]	40	40
Compaction pressure, P_C [MPa]	1–15	0.1
Heating time, $t_2 - t_1$ [min]	10	10
Compaction time, $t_3 - t_2$ [min]	10	10
Cooling time, $t_4 - t_3$ [min]	5	5

Table 4
Test specimen dimensions for tensile and compressive testing of consolidated all-PP laminates

Test	Specimens	Tape orientation	Dimensions[a]
Tension	W140/160/L/M/H	[0°/90°]	150 mm × 12 mm × 1 mm
Tension	W140/160/L/M/H	[±45°]	150 mm × 25 mm × 1 mm
Compression	W140C/W160C	[0°/90°]	15 mm × 5 mm × 5 mm

[a] Dimensions given as length × width × thickness. Exact specimen thickness depends on processing parameters.

3. Mechanical testing of woven tape all-PP composites

Tensile strength and modulus were measured using an Instron 5584 tensile testing machine, equipped with a 5 kN load cell and Merlin data acquisition software. Specimens were placed in specially designed composite grips which allow 10° free rotation to reduce the effect of moments in off-axis specimens. The gauge length of tensile test samples was 120 mm and tests were performed at 2 mm min^{-1} with a small preload (~1 N). To fully characterise the samples, two types of tensile tests were performed: tensile deformation at low strain to determine moduli and Poisson's ratio, and deformation to failure to determine strength and strain to failure. To determine the tensile modulus for each specimen, strain was measured by strain gauges placed in the direction of tensile loading at low strains (<2.5%). The Young's modulus was calculated in all cases using a range of 0.05–0.2% strain; in all cases this proved to be a linear and reproducible region with very little deviation. To determine Poisson's ratio, an additional strain gauge was placed at 90° to the direction of loading. To determine the tensile strength and strain to failure of the specimens, high strain (>2.5%) data from the cross-head displacement was used to measure extension. Five specimens were tested for each case to ensure reproducibility. Specimens which failed within, or very close to, the gripped region of the specimen were discarded.

Compressive tests, denoted by a "C" suffix in Table 4, were performed on end-loaded specimens manufactured in the same way as tensile specimens, but cut from thicker (5 mm) plates. Compressive tests were performed using a Hounsfield H25KS tensile testing machine configured for compressive loading and equipped with a 2.5 kN compression load cell and compression plates, in accordance with ASTM D695-96 and as described elsewhere [2]. The specimen geometry is shown in Table 4. Tests were performed by applying compressive loading at a rate of 1.3 mm min^{-1}. The recorded load increases with compression until ultimately tends reaches a uniform stress. This plateau of constant stress is recorded as the compressive strength. As before, five specimens were tested for each case to provide an accurate measure of the compressive strength. Compressive strength tests were performed along one tape direction [0°/90°] of woven tape specimens. All tests reported in this paper were performed at room temperature, but the tensile and flexural properties of all-PP composites at elevated temperatures have also been investigated [2] and will be presented in a subsequent publication.

During tensile testing, acoustic emissions of the specimens during failure was also monitored using a 15 mm differential sensor, connected to a 40 dB 1220A preamp and AEWin data acquisition software manufactured by Physical Acoustics Ltd, UK. An analogue filter was applied with upper and lower thresholds of 2 MHz and 100 kHz, respectively, and a gain of 60 dB. A high viscosity grease was used to couple the acoustic emission sensor to the specimen.

Table 5
Compressive strength of unidirectional tape all-PP composites compared to woven tape all-PP composites

Compaction temperature	UD σ_{11C} [MPa] [29]	UD σ_{22C} [MPa] [29]	Woven σ_{11C} [MPa]
140 °C	39.2	10.8	34.4
160 °C	55.8	12.0	43.2
Percentage difference	42%	12%	26%

4. Results and discussion

4.1. Compressive properties of woven tape all-PP composites

Table 5 shows the compressive strength of a woven tape composite, $W\sigma_{11C}$, compacted at 140 and 160 °C. Previous research [2] revealed that there is no increase in the transverse compressive strength of the unidirectional composite with increasing compaction temperature but there is an increase in compressive strength in the longitudinal direction. An increase can also be seen in the compressive strength of the woven composite with increasing compaction temperature.

A photograph of a typical failed woven composite compression specimen is shown in Fig. 4. In these photos, the dark regions are tapes travelling out of the plane of the paper while the light regions are tapes travelling left to right in plane of the paper. The tape structure is clearly buckled, with some interfacial failure in the buckled region. The increase in compressive strength of the woven tape composite with increasing compaction temperature can be attributed to the increase in buckling stability of the tapes due to and increase in interfacial adhesion with increasing compaction temperature [2]. The interfacial properties of all-PP composites will be presented in more detail in a subsequent paper. However, since failure is by buckling, the bending stiffness must also contribute to the compressive strength.

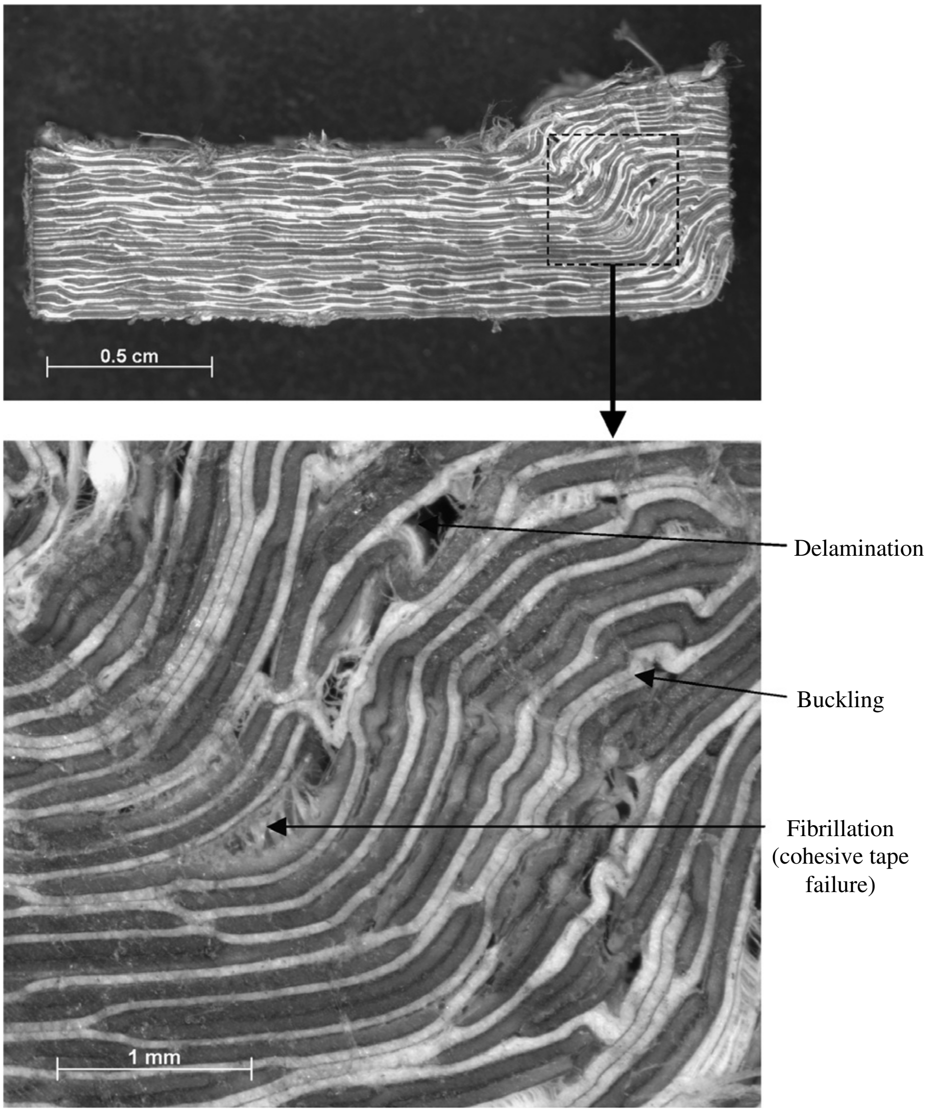


Fig. 4. (Top) Optical micrograph of a cross-section of an end-loaded compression test specimen (W160C) which has failed by buckling, and (bottom) increased magnification of buckled region showing tape buckling and delamination. (Compression loading direction is horizontal.)

The bending stiffness of an all-PP woven fabric composite is mainly governed by the stiffness of the tapes in the longitudinal direction, and so the transverse tapes do not contribute greatly to the bending stiffness during buckling. This is supported by considering that the increase of the compressive strength of the woven composite specimen with increasing compaction temperature is approximately half that of the increase of the unidirectional specimen with increasing compaction temperature. This seems to be due to the 50% (see Table 5) volume fraction of tapes in the longitudinal direction providing bending stiffness in the woven tape composite. A higher magnification of Fig. 4 shows that in the buckled region of the failed specimen, some delamination is seen with some fibrillation traversing the delaminations. It is likely then that the increase in compressive strength with increasing compaction temperature is due to the increase in interfacial strength resulting from higher compaction temperatures.

The drawing mechanism increases tensile properties of polymer structures by aligning the microstructure of the material in the drawing direction [30–32], however it is unlikely that any increase would be seen in compressive properties of tapes. The compressive strength seen here is very close that of bulk PP (~50 MPa), and since tensile tests have shown that there is very little affect of processing temperature in this region on the microstructure of the PP tapes, it is clear that no real benefit is gained by using these highly oriented PP's rather than bulk PP for purely compressive applications. Even highly oriented polyethylene fibres which show a much higher tensile strength (3 GPa) exhibit a very low compressive strength (70 MPa) [33]. This poor compressive strength must be taken into account when designing all-PP products which will experience compressive or flexural loads.

4.2. Tensile properties of woven tape all-PP composites

The tensile properties of woven tape composites are presented in Figs. 5 and 6, which show tensile strength and modulus of woven tape composites loaded at $\theta = 0°/90°$ and $\theta = \pm45°$, where θ is defined as the angle between the tape direction and the loading direction. From Fig. 5, the values obtained for processing temperatures of 140 and 160 °C both show very similar strengths but as $\theta = \pm45°$, a difference in strength between specimens with these compaction temperatures is observed. As the processing temperature increases, the off-axis strength of these composites increases. This can be expected because the strength of off-axis composites is due to interfacial bond strength between tapes resisting delamination and so tape pull-out; at $\theta = 45°$, failure occurs by intraply shearing and tape pull-out. As compaction temperature increases, the interfacial strength increases, and so superior load transfer increases the total strength of the composite structure.

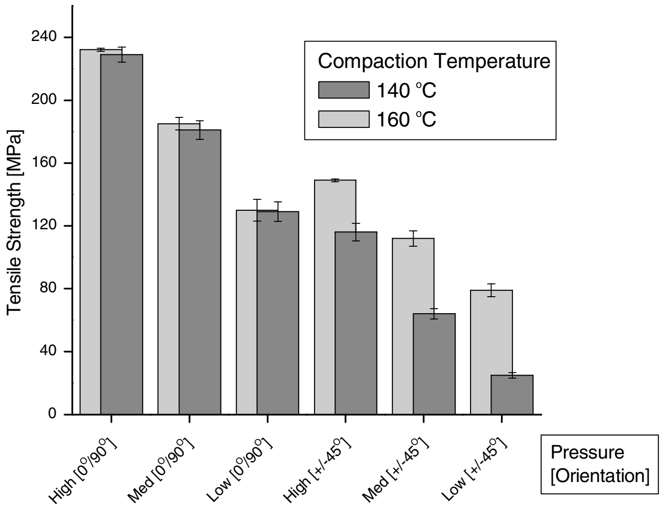


Fig. 5. Tensile strength of woven tape all-PP composite laminates consolidated at two different temperatures (140 and 160 °C) and three different compaction pressures (0.1 MPa (Low), 1.2 MPa (Med.) and 12.4 MPa (High)), showing a large effect of consolidation pressure for both [0°/90°] and [±45°] loading directions, but a negligible effect of consolidation temperature in specimens loaded in the [0°/90°] direction.

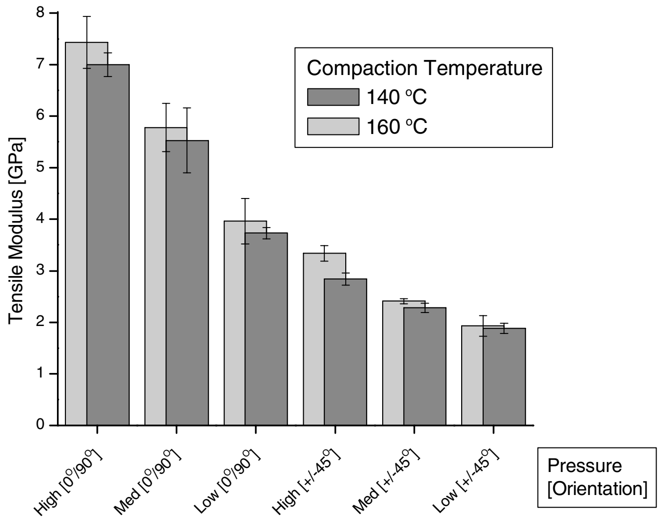
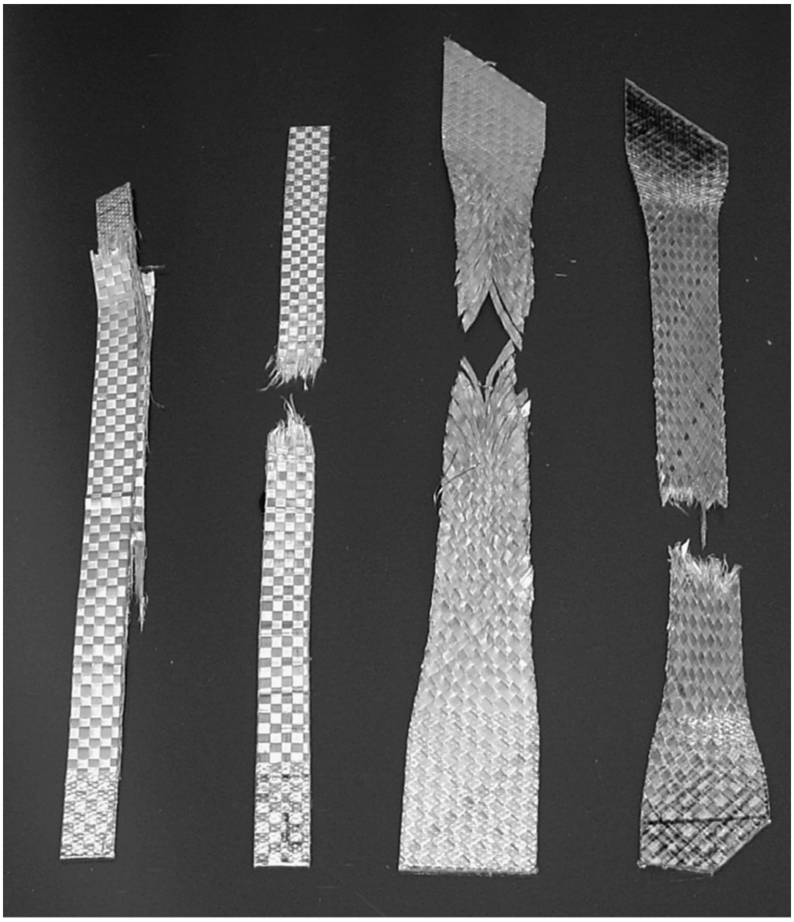


Fig. 6. Tensile modulus of woven tape all-PP composite laminates consolidated at two different temperatures (140 and 160 °C) and three different compaction pressures (0.1 MPa (Low), 1.2 MPa (Med.) and 12.4 MPa (High)), showing a large effect of consolidation pressure for both [0°/90°] and [±45°] loading directions, and only a small effect of consolidation temperature.

Fig. 7 shows photographs of tensile testing specimens which have failed by a variety of mechanisms. The two specimens on the left of the photograph are [0°/90°] specimens compacted at 140 and 160 °C, and 0.1 and 1.2 MPa, respectively. The main difference between these two specimens is the improved interface as established earlier and this clearly affects the failure mode of these two specimens. Specimen W140L shows large amounts of delamination with entire plies of fabric debonding from the composite together with only small fabric breakage along the length of the composite.



[0°/90°]W140L *[0°/90°]W160M* *[±45°] W140L* *[±45°]W160H*

Fig. 7. The range of failure modes of woven tensile specimens. Specimens consolidated at 140 °C/0.1 MPa and loaded at [0°/90°] show widespread delamination, whereas those processed at 160 °C/1.2 MPa and loaded at [0°/90°] show local fibrillation. Specimens consolidated at 140 °C/0.1 MPa and loaded at [±45°] show debonding and interlaminar shearing over the entire specimen, whereas those processed at 160 °C/12.4 MPa and loaded at [±45°] show much more localised damage.

Specimen W160M shows a completely different failure mode; a single failure is seen in the centre of the specimen length with some very localised micro-fibrillation, but no complete tape pull-out. Since tests on unidirectional specimens showed a negligible decrease in tensile strength of the longitudinal tapes with increasing compaction temperature [2,29], this change in failure mode must be due to increased interlaminar strength allowing local stresses to build up to reach the longitudinal tensile strength of the tapes.

A similar effect is seen in the case of woven fabric composites tested in the [±45°] direction. Unlike in the case of loading W140M in the [0°/90°] direction, shear deformation over the length of the specimen is clearly visible by a reduction in specimen width following loading in the [±45°] direction. This reduction in width is accompanied by an increase in specimen thickness caused by intraply shearing (the so-called trellis effect [34]). Fig. 8 shows composite optical micrographs of the failure surfaces of (a) W140L and (b) W160H. Ultimately, as in the case of [0°/90°] loading, failure of W140L is by a tape pull-out mechanism following rotation of the tapes to align nearer to the loading direction, as can be seen at the failure location. Specimen W160H also shows a high degree of deformation over the whole specimen length due to tape shearing, but in this case, tape pull-out is absent at the failure location. Instead, tape breakage occurs and the failure site is similar in appearance to the failure site of [0°/90°] specimen, with a ''brush-like'' fracture surface showing extensive micro-fibrillation. In either case, tapes have rotated from 45° to 27° to the loading direction.

The concept of intraply shearing and fibre pull-out is reinforced by Fig. 9, which shows the strain to failure of woven tape composites. At $\theta = 0°/90°$, the strain to failure is very low, approximating the strain to failure of a single tape, since this failure mode is dominated by tapes loaded in tension. As θ increases, increasing amounts of shearing and tape debonding lead to a much greater apparent strain to failure of the composite system of $\sim$40%. The effect of processing parameters is less clear, but specimens compacted at lower temperatures show a greater strain to failure due to the lower interfacial strengths allowing greater tape pull-out and debonding.

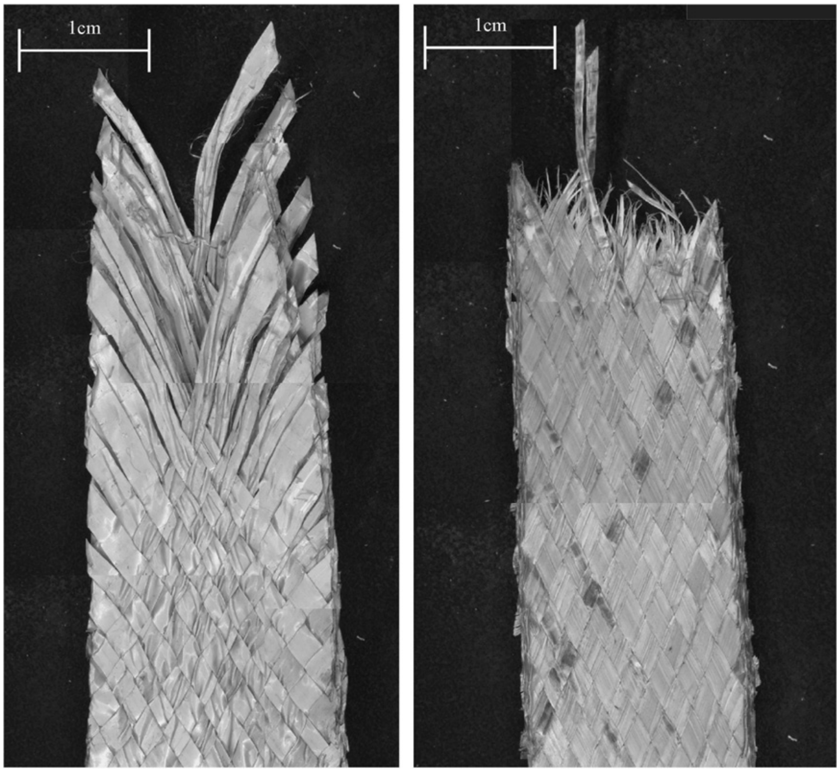


Fig. 8. Optical micrographs showing tape debonding and interlaminar shear failure modes of woven all-PP composites compacted at 140 °C/0.1 MPa, compared to the localised failure of specimen consolidated at 160 °C/12.4 MPa and loaded at [±45°].

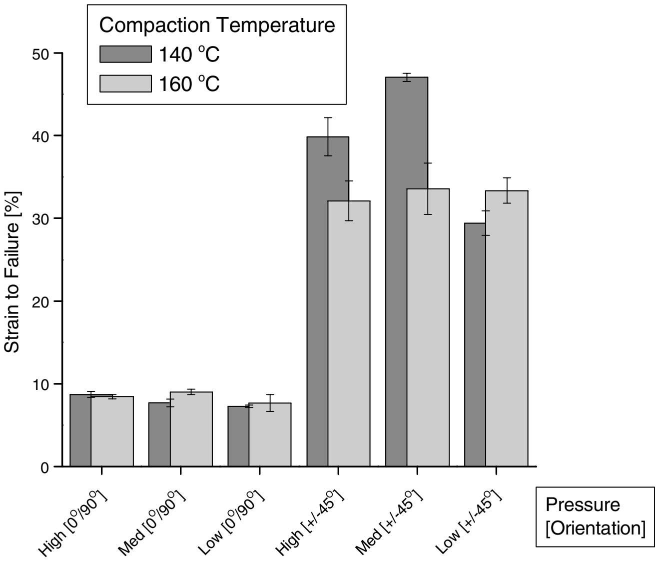


Fig. 9. Strain to failure of woven tape all-PP composite laminates consolidated at two different temperatures (140 and 160 °C) and three different compaction pressures (0.1 MPa (Low), 1.2 MPa (Med.) and 12.4 MPa (High)), showing a large increase in strain to failure in specimens loaded at [±45°] due to tape debonding and interlaminar shearing compared to those loaded at [0°/90°].

The effect of processing conditions on the tensile modulus of woven tape all-PP composites is much smaller. Since the modulus depends on the microstructure of the tapes, whereas the tensile strength depends on a combination of tape microstructure and composite macrostructure, it has already been shown for a unidirectional specimen that the modulus is largely unaffected by processing temperature within this range [29]. However, increasing the applied pressure during consolidation seems to increase the modulus of a composite loaded at $\theta = 0°/90°$ and $\theta = \pm 45°$. The greatest modulus seen is for the specimen with the greatest applied pressure and temperature, is 53% of E_{11} of the tape, and since the volume fraction of longitudinally oriented tapes in the woven fabric is 50% and the transverse modulus, E_{22}, of tapes is relatively small ($E_{22} = 1.5$ GPa) [2,29], this agrees with the rule of mixtures.

If the woven composite is considered as two stacked unidirectional plies with 90° rotation between plies, the modulus of the composite (E_{calc}) can be predicted by using laminate theories [35] and compared to the experimentally measured values presented earlier. If the composite is considered to be loaded in the [0°/90°] direction, then the modulus can be calculated as follows. The modulus, E_{c11}, of the ply containing tapes oriented in the 0° direction (parallel to the direction of loading), is given by the rule of mixtures:

$$E_{c11} = E_{f11} V_f + E_m (1 - V_f) \qquad (1)$$

In which E_{f11} is the tensile modulus of tapes in the tape direction ($E_{f11} = 15$ GPa), V_f is the volume fraction of tapes ($V_f = 0.89$) and E_m is the tensile modulus of the matrix copolymer ($E_m \approx 1$ GPa). This predicts a value for the tensile modulus of this ply, $E_{c11} = 13.46$ GPa. The tensile modulus, E_{c22}, of the ply containing tapes oriented in the

90° direction (normal to the direction of loading) is predicted by the Halpin–Tsai equation [35]:

$$E_{c22} = E_m \left(\frac{1 + \xi \eta V_f}{1 - \eta V_f} \right) \tag{2}$$

$$\eta = \frac{(E_{f22}/E_m) - 1}{(E_{f22}/E_m) + \xi} \tag{3}$$

$$\xi = 2\frac{a}{b} \tag{4}$$

where E_{f22} is the transverse modulus of the tape ($E_{f22} = 1.5$ GPa) and a and b are the width ($a = 2.15$ mm) and thickness ($b = 65$ μm) of the tape, respectively. It is important to note that since the tape is highly oriented in one direction, the tape itself is highly anisotropic and so $E_{f22} \neq E_{f11}$. The high volume fraction of fibre, together with the proximity of E_{f22} and E_m result in E_{c22} being approximately equal to E_{f22}, $E_{c22} = 1.45$ GPa. Combining E_{c11} and E_{c22} predicts the tensile modulus of the combined laminates, $E_{c(Calc)} = 7.45$ GPa. However, due to the compaction process, varying compaction conditions alter the density of the composites by causing void closure and so affect apparent measurements. For this reason, the specific tensile properties must be compared since this takes into account changes in density due to interlaminar void closure. The calculated specific modulus of the composite, $E^s_{c(Calc)}$, i.e., normalised for composite density is given below:

$$E^s_{c(Calc)} = \frac{E_{c(Calc)}}{\rho} \tag{5}$$

where ρ is composite density. The density of the composite processed at 140 °C and 1.2 MPa is 0.700 g cm^{-3} while the density of the composite processed at 160 °C and 1.2 MPa is 0.780 g cm^{-3} (see Table 7). The calculated specific moduli of composites processed at 140 °C, $E^s_{c(Calc)140}$, and 160 °C, $E^s_{c(Calc)160}$, are 10.65 GPa g cm^3 and 9.55 GPa g cm^3.

$$E^s_{c(Exp)} = k_1 k_2 E^s_{c(Calc)} \tag{6}$$

where k_1 and k_2 are efficiency factors which describe the major discrepancies between the model and the experimentally measured values. k_1 describes the loss of tensile modulus caused by molecular relaxation which occurs in the highly oriented homopolymer core during heating the stacked plies laminates in order to achieve bonding. k_2 describes the effect off-axis loading of tapes in the woven plies due to tape crimping in the woven structure [36,37]. The use of a thin tape geometry rather than a circular cross-section fibre geometry which is normally seen in conventional tow or roving based composites, should reduce the degree of crimping to maximise k_2.

Both of these factors will reduce the tensile moduli seen experimentally in composite laminates. Since the melting temperature of the copolymer skin layer is significantly lower than that of the homopolymer core, it would be expected that k_1 approaches 1, i.e., the loss of modulus due to molecular relaxation is minimised. However, studies relating the tensile modulus of unidirectional all-PP composites with the tape precursor revealed a 18% loss in specific modulus due to molecular relaxation and tape density increases occurring during the consolidation process at both 140 and 160 °C [2,29], therefore it is assumed that $k_1 = 0.82$. This value can be substituted in Eq. (6) to give $k_2 = 0.91$ and 0.95 for compaction temperatures of 140 and 160 °C, respectively (see Table 6). This effect is quite low compared to that seen in conventional composites. A 20% decrease in modulus due to fibre crimping has been reported for carbon fibre reinforced epoxy composites [38]. The efficiency factor k_2 increases with consolidation temperature, suggesting a reduced crimping of tape properties in the final composite consolidated at 160 °C.

Table 6
Efficiency factors, k_1 and k_2, comparing predicted and experimentally measured specific tensile moduli for all-PP composites consolidated at 140 and 160 °C

Specimen	$E^s_{c(Calc)}$ [GPa g cm^3]	$E^s_{c(Exp)}$[GPa g cm^3]	Efficiency factor	
			k_1	k_2
W140M	10.65	7.90	0.82	0.91
W160M	9.55	7.41	0.82	0.95

In order to further evaluate the effect of the fabric crimp on the mechanical properties of all-PP composites, the theoretical stiffness of the all-PP fabric was assessed by using a two-dimensional finite element model using finite element analysis (FEA) software Abaqus v6.3. The model represents the cross-section of a laminate based on multiple layers of an all-PP plain weave fabric and consists of two unit cells of the fabric placed in parallel (see Fig. 10). In the fabric, the tape orientation is either parallel to the X direction (grey) or to the Z direction (black). The grey and the black layers represent only the homopolymer core of the coextruded tape. The thickness of the copolymer skin was set to 5 μm on both sides of the core. The model has a length $X = 2.14$ mm and a thickness $Y = 0.260$ mm. The length equals half the plain weave unit cell length (i.e., the width of the tape) while the thickness equals four times the thickness of the tape. The following orthotropic properties were considered for the core material: $E_{11} = 15$ GPa, $E_{22} = 1.5$ GPa, $G_{12} = 0.8$ GPa and $\nu_{12} = 0.36$ while the copolymer skin was assumed isotropic ($E = 1$ GPa and $\nu = 0.3$). The longitudinal modulus of the tape (65 μm thick, $E_{11} = 13$ GPa) is then equivalent to the value reported previously and is discussed in a separate publication [2,29]. The region where the grey tape has a 25° angle with the loading direction (X) is due to the crimp of the fabric. This angle is realistic and correlates to experimental observation. In the crimped region of one fabric layer, no tape is parallel to the loading direction so that locally a low stiffness is expected based on the off-axis properties of the tape [2,29]. One fabric layer can then be assumed as a cross-ply element placed in series with a short unidirectional off-axis element. If the fabric layers were orderly stacked, the crimp regions would be aligned and only one fabric layer would be necessary to model the laminate. Here, two non-aligned layers of fabrics were

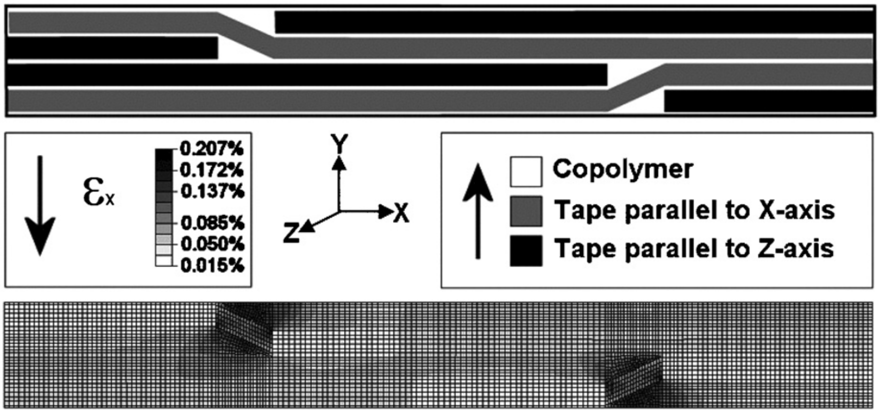


Fig. 10. Finite element model of a woven all-PP laminate. (Top) Geometry and material definition. (Bottom) Mesh and strain distribution, ε_x, when a total strain of 0.1% is applied along the X direction.

considered as a more realistic representation of a woven laminate. The distance between the crimp regions is maximised and equals half the width of the tape. A total strain of 0.1% was applied to the model leading to a calculated Young's modulus of 6.1 GPa. As this value compares well with the measured Young's modulus (5.8 GPa), it can be concluded that the properties of the oriented tape are retained. As expected the modulus of a woven fabric based laminate is lower than that of a non-crimp cross-ply laminate due to the low stiffness of the crimp regions. The distribution of the strain, ε_x, reveals a locally higher deformation in the crimp region (dark shades in Fig. 10).

Figs. 5 and 6 showed that the pressure applied during composite consolidation greatly affects the tensile mechanical properties of the composites in the [0°/90°] direction. This observation will now be explained. It was observed during specimen production that processing temperature and pressure alter the density of the resulting composites. Density tests were performed on samples taken from the same composite plates used for tensile testing, using the Archimedes principle. The relationship between the density of the plates and the processing parameters is presented in Fig. 11. It can be immediately seen that with increasing processing pressure, plate density increases. The effect of increasing compaction temperature is an increase in density. The tape density, ρ_{tape}, is also shown in Fig. 11, and is less than the density of isotropic polypropylene due to the presence of micro-voids [2]. The woven tape specimens, W140 and W160, compacted at 0.1 MPa have a density less than the original tape. This can only be caused by the presence of intertape voids, caused by the surface irregularity of adjacent woven plies, due to tape crimping. In these specimens, consolidation is incomplete and poor interfacial properties would be expected. With increasing compaction pressure, specimens compacted at both temperatures show an increase density, reaching and exceeding the original tape density, ρ_{tape}. Thus, with increasing pressure, there will be a transition from a combined state of intertape voids and voids within the tape, to solely voids within the tape. At the highest pressure applied here, 12.4 MPa, a density of ~0.87 g cm^{-3} is achieved, which approaches the density of isotropic PP, ~0.91 g cm^{-3}. The copolymer skin layer is highly viscous even above the melting temperature and so requires some pressure to flow into the voids caused by the surface roughness due to the weave of the fabric.

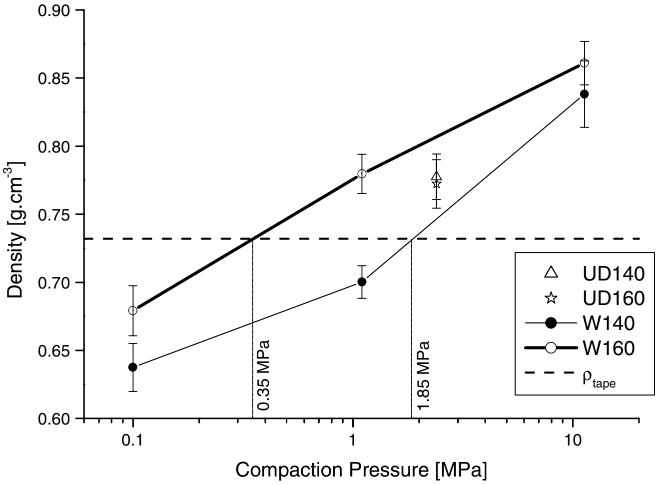


Fig. 11. The effect of processing parameters on plate density of unidirectional tape plates compacted at 140 °C and 160 °C (UD140 and UD160) [29] and woven tape plates compacted at 140 °C and 160 °C (W140 and W160) compared to the density of the precursor tape, ρ_{tape}. The woven tape composites show an increase in density with increasing compaction pressure.

From Fig. 11, the trends for compaction temperatures of 140 and 160 °C have an approximately constant difference suggesting that the density is predominantly dependant on compaction pressure, rather than temperature. An increased temperature will assist the filling of these voids by reducing the viscosity of the copolymer layer and encouraging copolymer flow, but compaction temperature in the range considered is not the dominant parameter. By assuming that filament wound unidirectional composites possess a perfect, void free tape stacking arrangement, the effect of compaction pressure would not be seen in unidirectional composites. Unidirectional specimens consolidated at 140 and 160 °C have very similar

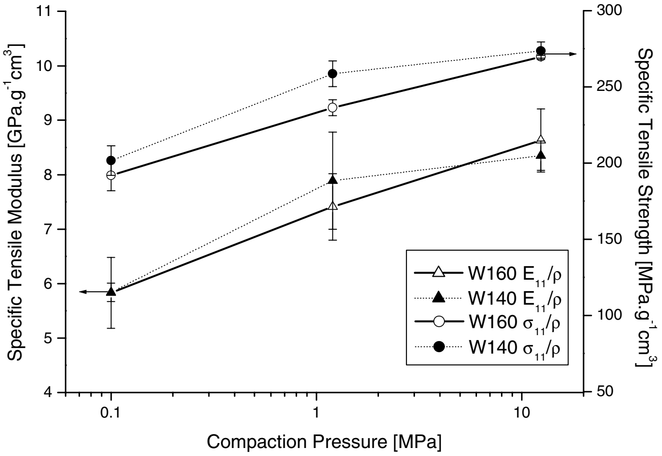


Fig. 12. Specific tensile moduli and strengths of woven tape all-PP composite laminates consolidated at two different temperatures (140 and 160 °C) and three different compaction pressures (0.1 MPa (Low), 1.2 MPa (Med.) and 12.4 MPa (High)), showing an increase in specific mechanical properties with increasing consolidation pressure.

density, ~7% greater than that of the tape [2,29]. The small difference in density between the tape and the unidirectional specimens is probably due some closure of voids within the tape during consolidation. The presence of intertape voids in woven tape fabric must strongly affect the mechanical properties of woven tape composites since the voids can make no load bearing contribution to the composite structure in tension. However, in some composite systems, the presence of intertape voids increases the strain to failure, and so the energy absorbed during failure of the composite, via a crack arresting mechanism [39]. Since tensile strength and modulus are inversely proportional to the cross-sectional area subjected to loading, the results presented in Figs. 5 and 6 can be reassessed by normalising the mechanical properties for variations in density. This suggests that providing that molecular relaxation is prevented, tensile properties of all-PP composites increase with increasing compaction pressure and temperature. The specific mechanical properties in the [0°/90°] direction as a function of compaction pressure are shown in Fig. 12.

Fig. 12 further reinforces the concept that tensile properties in the [0°/90°] direction are not very sensitive to changes in compaction temperature in the range 140–160 °C. It is likely that a set of optimum consolidation conditions will be seen with increasing compaction temperature and pressure, but ultimately these will encourage plastic flow, allowing molecular mobility and a loss of mechanical properties. To fully understand the changes in density seen in these composite plates, cross-sections of

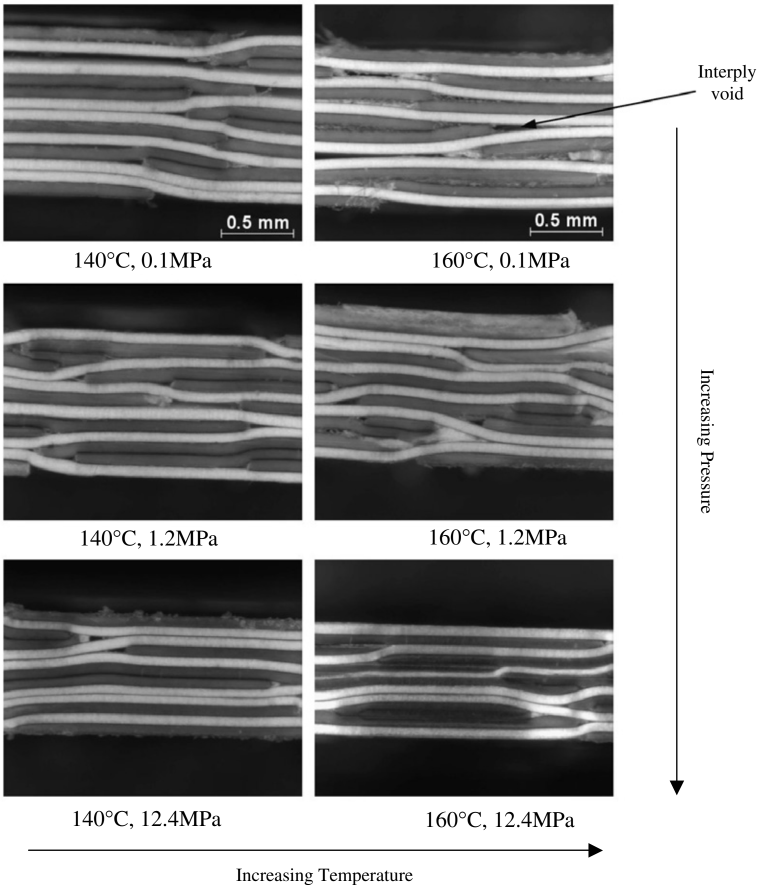

140°C, 0.1MPa
160°C, 0.1MPa
140°C, 1.2MPa
160°C, 1.2MPa
140°C, 12.4MPa
160°C, 12.4MPa

Fig. 13. Cross-sections of woven tape composites compacted with increasing temperature and pressure illustrating the effect of processing conditions on intertape voids and degree of tape crimping.

woven tape composite plates were viewed using optical microscopy. These images are presented in Fig. 13. Although some degree of crimping is seen in the compacted tape, the degree of crimping is reduced by the use of a thin tape geometry reinforcement, rather than a traditional glass fibre yarn or roving, which typically leads to a much higher degree of crimping and an associated decrease in mechanical performance of the composite [40].

In Fig. 13, light and dark areas are clearly visible in the composite cross-sections. The light areas are tapes travelling along the width of the specimen from left to right on the page. The dark areas are tapes travelling out of the plane of the page. The reason for the differences in brightness between these tapes, is that the tapes oriented out of the plane of the paper are much rougher than those cut along the plane of the paper, and so these tapes reflect less light and appear darker. The increase in compaction temperature from 140 °C (left-hand side) to 160 °C (right-hand side) shows little affect, but increasing pressure (top to bottom of page) has a noticeable effect. Even though all cross-sections are presented at the same scale, the increase in density is clearly seen as the thickness of the laminates decreases noticeably with increasing pressure. Also seen in specimens compacted at 0.1 MPa is the conspicuous presence of intertape voids, proving their existence. These intertape voids are also seen in specimens compacted at 1.2 MPa at both temperatures but are absent from those compacted at 12.4 MPa and 160 °C. Therefore, it would seem that the optimum pressure to eliminate these intertape voids is between 1.2 and 12.4 MPa, but also the exact value of this "optimum" pressure may depend on compaction temperature since following consolidation at 140 °C, even 12.4 MPa is insufficient to totally close all the intertape voids. This means that even though Fig. 11 showed that the composite plates processed at 140 and 160 °C would exceed the density of the tape if compacted at 0.35 and 1.85 MPa, respectively, Fig. 13 shows that intertape voids are still present in composites. The composite processed at 140 °C and 12.4 MPa has a density of $\sim$0.83 g cm^{-3}, and so the presence of voids means that the density of the tape must be above this value; voids within the tape must be closing before complete closure of intertape voids. This complicates a direct comparison between specific modulus of tapes and specific modulus of composites.

The total closure of the intertape voids requires an excess of molten copolymer to flow out from between adjacent tapes into the voids. However, the particular copolymer selected for the skin layer was chosen to create a large enough temperature processing window as possible. The melting behaviour of the copolymer determines the minimum consolidation temperature during processing and the maximum application temperature of the all-PP composite product. The use of a copolymer with a lower melting temperature would expand the temperature processing window to even lower temperatures, but would limit the upper operating temperature of the composites; applying the composites in these higher temperatures would lead to delamination and rapid composite failure, since the copolymer would soften and no longer be able to facilitate load transfer between the tapes.

However, if the compaction temperature is too low, the viscosity of the copolymer melt may not be low enough to enable flow into these intertape voids. In this case the voids will remain in the final composites. As well as affecting the apparent mechanical properties of all-PP composites, the presence of these voids may have implications for applications such as automotive in which the composites will be at risk of fluid ingress. The specimen compacted at 160 °C and 12.4 MPa shows total closure, flattening the composites and forcing the tapes into alignment with the loading direction. This reduced crimping should definitely increase the performance of the composite especially in view of the extremely poor off-axis properties of the tape, and is reflected in the increased k_2 efficiency factor for composites processed at higher temperatures, as described earlier.

A summary of the mechanical properties for woven tape fabric plates is presented in Table 7, with a comparison to commercial glass reinforced PP composites: a typical glass mat reinforced PP (GMT, quadrant composites), a continuous, woven glass fibre reinforced PP (Twintex®, Saint Gobain–Vetrotex) and a random flax mat reinforced PP (NMT) [41]. It is clear that woven all-PP composites can compete with the GMT properties presented here, due to the comparable mechanical properties together with the weight advantage of all-PP. The truly isotropic nature of random short (glass or natural) fibre reinforced PP has to be considered when comparing the apparently inferior off-axis ($\theta = \pm 45°$) properties of woven tape all-PP composites. A variety of stacking arrangements (e.g., $[0°/90°/\pm 45°]_s$) can be applied to achieve quasi-isotropic properties in an all-PP laminate, if this is required.

Woven all-PP composites can compete with woven glass reinforced PP presented here in tensile strength applications if density is taken into account, but cannot compete in terms of tensile modulus due to the significantly greater stiffness of glass fibres. While the ultimate mechanical properties of all-PP composites can be compared to GMT, the damage process during tensile deformation of all-PP composites seems to be very much different. The number of acoustic emission (AE) hits can be used to provide valuable information about the onset of damage in glass fibre reinforced composites [42–44]. Fig. 14 shows the AE hits per unit volume recorded during tensile deformation of all-PP composites consolidated at 140 and 160 °C, compared to GMT and woven glass reinforced PP. GMT and woven glass reinforced PP emit acoustic events from very low stresses due until failure. This is due to the low strain to failure of glass fibres, combined with the relatively large scatter of glass fibre strength as well as the poor interfacial strength giving a combination of interfacial failure and fibre breakage. Conversely the all-PP composites reveal a much smaller number of AE hits due to the greater strain to failure of the PP tapes, together with an excellent interfacial bonding. Also contributing to

Table 7
Mechanical properties of all-PP woven tape laminates compared with commercially available alternatives (see also [45])

Material	All-PP woven tape composite						PP/random glass	PP/woven glass	PP/random flax
Temperature (°C)	140 °C			160 °C			GMT	(Twintex®)	NMT
Pressure (MPa)	0.1	1.2	12.4	0.1	1.2	12.4	50%W_f[a]	60%W_f[b]	40%W_f [41]
Density [g cm^{-3}]	0.638	0.700	0.838	0.679	0.780	0.861	1.31	1.50	1.15
E_{11} [GPa]	3.73	5.53	7.00	3.96	5.78	7.43	9.2	15	4.6
E_{11}/ρ [GPa g^{-1} cm^3]	5.85	7.90	8.35	5.82	7.41	8.63	7.02	10	4
E_{45} [GPa]	1.88	2.28	2.84	1.93	2.41	3.34	9.2	–	4.6
E_{45}/ρ [GPa g^{-1} cm^3]	2.94	3.26	3.39	2.84	3.09	3.88	7.02	–	4
σ_{11} [MPa]	129	181	229	130	185	232	145	350	55
σ_{11}/ρ [MPa g^{-1} cm^3]	202	259	273	191	237	269	111	233	48
$\sigma_{\theta=45}$ [MPa]	25	64	117	79	112	149	145	–	55
$\sigma_{\theta=45}/\rho$ [MPa g^{-1} cm^3]	39	91	140	116	144	173	111	–	48
σ_{11C} [MPa]	–	34.4	–	–	43.2	–	105 [45]	160	–
ν_{12}	–	0.07	0.15	–	0.08	0.15	–	0.10	–

[a] GMT, quadrant composites.
[b] Saint Gobain–Vetrotex.

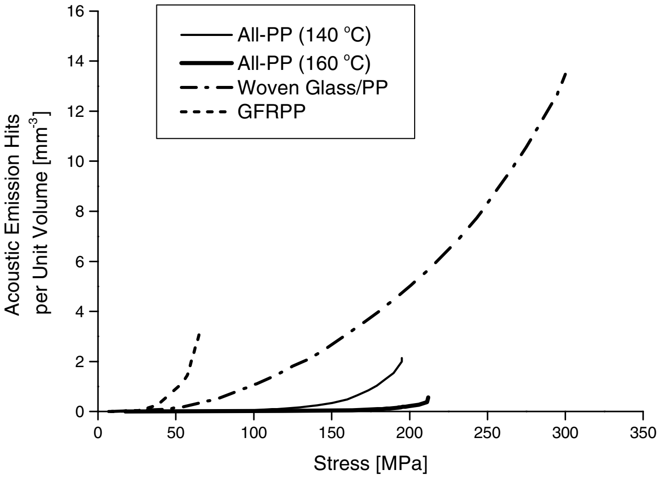


Fig. 14. Acoustic emission of all-PP composites (compaction temperature shown in brackets) loaded in tension to failure compared to two commercial glass reinforced polypropylenes, indicating the excellent damage resistance of well-compacted all-PP laminates.

this lower number of AE hits of all-PP composites compared to glass reinforced composites is the lower number of larger reinforcing elements (tapes) in all-PP composites compared to the high number of smaller, reinforcing elements (glass fibres) present in either GMT or woven glass reinforced PP. The cross-sectional area of a PP tape (2.15 mm wide × 65 μm thick) is more than 500 times that of a typical glass fibres (diameter 18 μm), and so the same volume fraction of reinforcement is achieved with a much greater number of discrete glass fibres. Therefore, a much larger number of AE hits due to fibre failure in the GMT would be expected, and this is seen in Fig. 14.

The difference in number of AE hits between all-PP specimens compacted at 140 and 160 °C is indicative of the greater interfacial strength of all-PP composites consolidated at higher temperatures, since Fig. 5 revealed that these specimens have very similar tensile strengths. Therefore, the decreased number of AE hits seen for the specimen compacted at 160 °C seems to be due to a reduction in AE hits caused by a reduced number of interfacial failures.

5. Conclusions

It has been shown in this paper that the parameters applied to consolidate all-PP composite specimens determine the mechanical properties. Despite the high temperatures involved during the consolidation process, the high strength and stiffness of a highly oriented polymer tape can be retained in a composite laminate, and all-PP composites agree with a rule of mixtures prediction of mechanical performance, as shown in Table 6. Furthermore, the tensile mechanical properties in the [0°/90°] direction are not sensitive to deviations in process temperature since mechanical properties proved approximately constant within the temperature processing window applied (140–160 °C), while tensile mechanical properties in the [±45°] direction are sensitive to both consolidation temperature and pressure. The mechanical properties measured in this paper show that due to their high reinforcement fraction of around 90%, all-PP composites can compete or outperform a popular type of glass fibre reinforced PP (GMT), and shows only slightly inferior tensile properties to a commercial woven glass reinforced PP, such as Twintex®.

The original reason for applying pressure during the consolidation of woven tape all-PP composites was to prevent shrinkage and relaxation by lateral constraining. Subsequently, it became obvious that pressure is also required to provide good surface contact by making the opposing surfaces, which are uneven due to the tape weave, meet. The compaction temperature and pressure can affect the mechanical properties of these composites by altering both the mesostructure of the composite (i.e., the structural composition), and the microstructure (i.e., the molecular orientation) of the tape material, but by choosing suitable parameters, loss of mechanical properties can be minimised. Increasing compaction temperature increases the interlaminar properties of all-PP composites [2], as indicated here by off-axis tensile testing together with acoustic emission studies, and this will be presented in more detail in a subsequent paper. This study has clarified the relationship between these parameters and the mechanical performance of the composite although further study may reveal absolute optimum processing conditions.

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