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The mechanical properties of unidirectional 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' composites, with a large temperature processing window ($>30\,^{\circ}$C) and a high volume fraction of highly oriented PP ($>90\%$). These composites show little deviation of mechanical properties with compaction temperature. This paper introduces all-polypropylene composites and reports the tensile and compressive properties of unidirectional composites. These composites show good retention of tape properties despite the relatively high temperatures used in composite manufacture.
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1. Introduction

Isotropic polypropylene (PP) lacks sufficient mechanical properties for many engineering applications. The mechanical properties can be greatly improved by two main routes. Firstly, foreign fillers or fibres can be added to PP to produce PP matrix composites, and secondly the mechanical properties of PP can be improved to satisfy these criteria by molecular orientation achieved by drawing. The most common method to improve mechanical properties is through the addition of fillers such as talc or glass fibres. However, PP composites which contain foreign fillers suffer from reduced recyclability since the main obstacle for recycling composites is the separation of fibre and matrix, which each have very different recycling requirements. With recent European environmental legislation [1] the recyclability of polymer composites in applications such as automotive, has become a greater concern. This problem can be reduced or removed by using similar or identical materials for both fibre and matrix. Polymers are already commonly used to form the matrix phase of composites, and can also be oriented by drawing to yield high strength and high modulus fibres, while several studies have also investigated the use of polymers for both fibre and matrix phase of the same composite. This is not so straightforward because most traditional composite processing routes cannot be applied, and so novel methods have been devised to combine fibres and matrices from similar polymers. Ultimately, because these composites can be entirely thermoplastic, recycling is simplified to melting of the composite and reprocessing, although this may not always be economically viable.

In order to attain a direct comparison with fibre reinforced polymer composites, polymer fibre/polymer matrix composites have been investigated. Single polymer composites based on oriented polyethylene (PE) fibres were first described in the mid-1970s [2]. In order to employ the high mechanical properties of polyolefin fibres, numerous investigations were conducted to combine high modulus PE or PP fibres with a similar matrix. Most early studies concern PE since the ultimate modulus of a linear PE molecule (~250 GPa) is much greater than the crystal lattice modulus of the helical PP molecule (~40 GPa) [3–5]. This molecular modulus ultimately limits the maximum modulus achievable in a fibre of either material, and so higher ultimate properties are achievable with PE fibres, and therefore this polymer has been the prime candidate for most initial studies. The lower inherent molecular stiffness of PP is reflected in the smaller scope of research into 'all-polypropylene' (all-PP) composites.

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However, the lower glass transition and melting temperatures of PE mean that creep at room temperature can be problematic and maximum usage temperature is lower than that of PP, which also benefits from a slightly lower price and density.

High modulus polymer fibres are required to impart high mechanical properties on resulting all-polymer composites. Such fibres have been produced by a variety of production routes from a melt [6,7] or via (solution) gel spinning [8–11], and subsequent orientation by drawing. The high mechanical properties of the fibre are due to molecular orientation in the drawing direction [12]. However, the main difficulty of combining fibres and matrices of similar polymers to create an all-polymer composite is to retain the properties of the oriented polymer molecules in the final composite, since molecular relaxation of highly oriented fibres readily occurs during heating [13]. Initially, most studies focussed on traditional routes to create thermoplastic composites, such as melt, powder or solution impregnation of multifilament yarns. The combination of similar polymer grades by exploitation of different melting temperatures opens up many routes for the production of single polymer composites. The creation of single polymer composites based on PE was first suggested by Porter and co-workers in the mid-1970s [2,14], by exploiting the difference in melting temperature of HDPE fibres and conventionally crystallised LDPE. Because these composites possess relatively low fibre volume fractions, the mechanical properties of the composite system are poor. Short fibre composites using UHMW-PE fibres in a HDPE matrix, have also been created by placing random fibres between two HDPE sheets [15]. This system was then consolidated by heating above the melting temperature of HDPE, and applying pressure. Using this method, higher fibre volume fractions were achieved but similar mechanical properties were obtained. All-PE composites have also been created by filament winding UHMW-PE fibres between isotropic ethylene–butene copolymer films [16], or by combining UHMW-PE fibres with a LDPE matrix using a dry powder impregnation technique [17]. The impregnation of UHMW-PE fibres with a HDPE powder suspended in propanol has also been reported [18], although the lower fibre volume fractions achieved make this route unattractive. Each of these routes exploits the difference in melting temperature between the fibre and matrix. Any route to the creation of all-polymer composites based on a discrete fibre and matrix system limits the fibre volume fraction, and so ultimately limits the mechanical properties of the composites. With both film stacking and powder impregnation routes, it is likely that with increasing fibre volume fraction, adequate matrix impregnation also becomes more difficult during consolidation.

2. Hot compaction of single polymer composites

A novel method for the preparation of all-polymer composites, without the need for a separate matrix material has been created at the University of Leeds, UK [19,20]. By carefully controlling the temperature applied, it was seen that the exterior of polymer fibres could be melted and with the simultaneous application of pressure, this molten polymer flows to fill the interfibrillar voids. Upon cooling, this recrystallised material forms the matrix phase of the composite.

By carefully controlling the processing parameters, up to 90% of the tensile modulus of the fibre has been retained in a PE/PE composite, but only by using a small optimum processing temperature window of a few degrees [19]. For unidirectional composites, a sharp decrease in longitudinal modulus and an increase in transverse modulus are seen on either side of this optimum compaction temperature as at higher temperatures, fibre properties begin to be lost by molecular relaxation. The transverse strength increases with compaction temperature, ultimately reaching the strength of isotropic PE when all orientation is lost. The hot compaction process was subsequently successfully applied to alternative grades of UHMW-PE [21], and PET [22], PMMA [23,24] and PP [25–29] fibres, in each case using a melted part of the fibre to act as the matrix. Because of the continuity between the fibre phase and the newly crystallised matrix phase, an excellent interfibrillar adhesion is seen in most of these cases. However, there is a fine temperature balance between melting a sufficient volume of the fibre to provide interfibrillar adhesion and melting an excessive volume of the fibre leading to a lower volume fraction of reinforcing fibre [30].

It is clear that a change in processing temperature of just a few degrees leads to a big decrease in longitudinal mechanical properties of the composite. Since highly oriented thermoplastic fibres are very sensitive to thermal relaxation, it is possible that a loss in fibre modulus will be seen well below the melting temperature of the fibre. As all the processing routes described in this paper involve consolidation by heating, it is expected that some molecular relaxation is possible in all processing routes.

3. Current research

The methods described so far, for preparing single polymer composites can be classified into two groups:

(i) combination of a discrete fibre and matrix by exploiting the difference in melting temperature of two grades of the same polymer, and
(ii) hot compaction of fibre bundles to selectively melt fibre exterior in order to form a matrix phase.

Both of these methods are feasible but have inherent limitations that reduce their viability; combining fibre/film or fibre impregnation is limited to *low fibre volume fractions,* which ultimately limit composite performance, while using a hot-compaction route is limited by a *very*
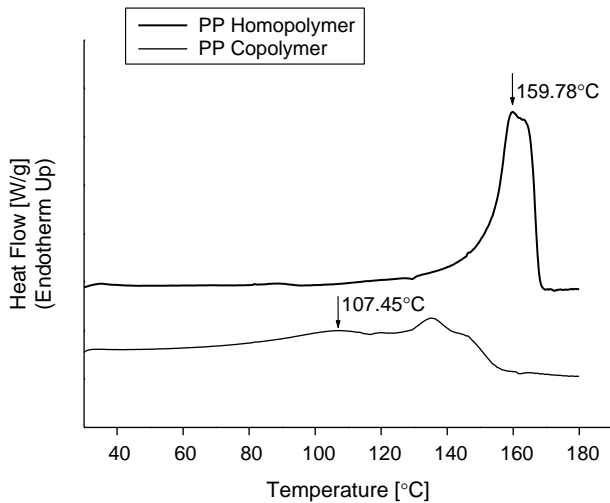
Fig. 1. DSC melting endotherms of PP homopolymer and PP copolymers used for the core and skin components, respectively, in the co-extrusion of all-PP precursor tape.

narrow temperature processing window, which complicates processing. Despite the obvious attraction of fully-recyclable composite materials which possess comparable mechanical properties to glass reinforced thermoplastic composites, these processing limitations have reduced the success of single polymer composites. The aim of the research reported in this paper is to develop a route to produce single polymer composites based on PP, which can compete with glass fibre reinforced PP, by combining a high volume fraction of reinforcement together with a large temperature processing window. A new processing route is now proposed that satisfies both of these criteria.

It has been reported [13,31] that high modulus polyolefin fibres could effectively be 'overheated' above their melting temperature by constraining the fibre, i.e. physically fixing the fibre ends. To allow relaxation of a highly oriented polymer fibre, a macroscopic decrease of length in the drawing direction must occur. Physically constraining the fibre ends prevents shrinkage and thus prevents the molecular reorientation, which causes the loss of modulus and, ultimately, a return to the isotropic state. It is possible to use this overheating by constraining process to create an 'artificial' difference in melting temperature between two structures of the same grade polymer, by constraining one but not the other [32].

This 'overheating' or 'superheating' as it is often termed, can be applied to further expand the temperature processing window of the hot compaction methods described above, and is affected by the application of lateral pressure by the press. This artificial expansion of the temperature-processing window is most interesting for combining the same grades of a polymer. This constraining method has recently been successfully applied to investigate the feasibility of creating a composite of high modulus PP fibre within a matrix of the same grade of PP [25,32–35], and model composites have been prepared by using this constraining route to produce single-fibre model composites.

Our studies have investigated alternative methods to further increase this temperature processing window. This new development uses high modulus PP homopolymer tapes, which are co-extruded with a coating of a PP copolymer [36–40]. Using a tape geometry PP was seen to be preferential to circular cross section fibres as the reduced thickness of the tape led to a lower crimping in the woven fabric, and so a more efficient reinforcement. The mechanical properties of woven tape all-PP composites have been assessed [39] and will be the subject of a separate publication. The PP copolymer coating used in this study is propylene based and contains 6% w/w ethylene. It has a broad melting behaviour in which melting (and so sealing during consolidation) initiates at 107 °C compared to the narrow melting peak of the PP homopolymer ($T_m \sim 160$ °C). Fig. 1 shows DSC curves of the PP homopolymer core layer compared to the PP copolymer skin layer, illustrating the large difference in melting behaviour. These co-extruded tapes can then be oriented in a mould and heated to above the melting temperature of the copolymer, while constraining by the application of lateral pressure in a hot press leads to a processing temperature window of > 30 °C. Besides constraining, the pressure also acts to consolidate layers of tape into a laminated composite, and this process is shown schematically in Fig. 2.

Table 1 shows some mechanical properties of unidirectional composites produced using constrained co-extruded tapes compared to unidirectionally oriented PP composites reported in literature [26,41,42]. As with the other composite systems, described previously, it is essential that fibre properties are retained in the final composite component, and so processing conditions are crucial. This retention is now simplified because
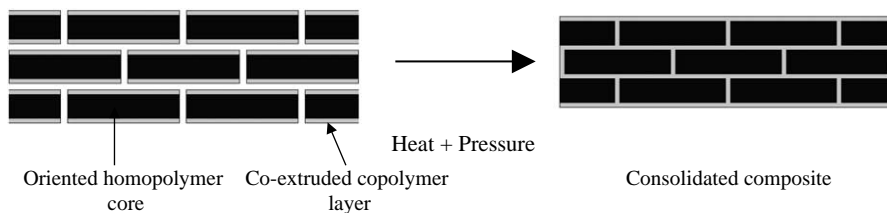

Fig. 2. Schematic of consolidation of co-extruded tapes into a coherent load bearing structure, showing that the use of tapes with a thin co-extruded skin layer can lead to the creation of composites with extremely high volume fractions of reinforcement phase (oriented homopolymer). The tape direction is into/out of the plane of the paper.

Table 1

Mechanical properties of unidirectionally oriented PP sheets manufactured via different processing routes

Method	Draw ratio [–]	E_I [GPa]	σ_I [MPa]	E_{II} [GPa]	σ_{II} [MPa]	Reference
Roll-drawn billets	20	20	450	2	50	[42]
Drawn sheets	14	7.5	250	–	–	[41]
Hot compacted PP fibres	–	3.7	87	–	7	[26]
UD co-extruded all-PP tape composite laminate	17	13	385	1.5	4.5	This paper

the temperature-processing window has now become much wider by combining constraining and co-extrusion coating of the tapes, and processing can occur at lower temperatures, thus reducing the risk of loss of fibre orientation. The mechanical properties of unidirectional all-PP composites produced by consolidating co-extruded and physical constrained PP tapes are presented in this paper.

4. Specimen preparation

The tape used throughout this section is a co-extruded three layer tape, with an A:B:A (copolymer:homopolymer: copolymer) structure, that was manufactured at Lankhorst Indutech B.V., The Netherlands, using a co-extrusion and tape drawing line. The tape has a thickness of 60 μm and a width of 2.15 mm. The mechanical properties of the as drawn, co-extruded tape are summarised in Table 2. The density of the highly drawn PP tapes ($\rho=0.732$ g cm^{-3}) is less than that of isotropic PP ($\rho=0.91$ g cm^{-3}) due to the phenomenon of microvoiding that occurs within the tape structure during drawing [43]. The presence of these microvoids reduces the density of the tapes but may also reduce the transverse strength of the tapes.

Unidirectional all-PP composite laminates were created from this tape via a filament winding process, by winding tape from a bobbin onto a thin, flat steel plate using a custom built winding machine (see Fig. 3). This frame is then placed in a matching mould and the tapes are compacted into a unidirectional composite sheet by the application of heat and pressure. The composite is placed in a purposed built mould (see Fig. 4), which is positioned between the platens of a 500 kN hot press. After the desired compaction temperature has been achieved and held for consolidation, the press is rapidly cooled. Pressure is maintained in

the press throughout this process. The temperature inside the mould is monitored externally using thermometer probes, inserted in upper and lower halves of the mould.

The time-temperature and time-pressure profiles for both methods are shown in Fig. 5. The specimen is placed in the hot press 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 compaction 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 hot press used and cannot necessarily be scaled up to larger process equipment. The compaction time (t_3-t_2) is kept constant at 10 min, while the entire cycle from insertion of the mould into the hot press 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. After consolidation and cooling, this sheet is cut from the frame to give two unidirectional laminates (one from either side of the winding plate). A schematic of the winding plate and mould is shown in Fig. 4. The unidirectional composite sheet formed using this winding method is approximately 120 mm×120 mm. These laminates can be cut into tensile testing specimens as described in ASTM 3039, and the dimensions of the tensile testing specimens are given in Table 4.

All specimens were cut from these plates by hand, using a razor blade to rectangular specimens of dimensions listed

Table 2

Mechanical properties of PP precursor tapes for the production of all-PP composites

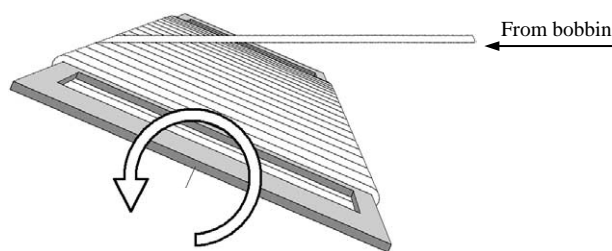|	
Width	2.15 mm
Thickness	60 μm
Density, ρ	0.732 g cm^{-3}
Draw ratio, λ	17
Composition, [A:B:A]	5.5:89:5.5
Tensile modulus	15 Gpa
Tensile strength	450 MPa
Strain to failure	7%

Fig. 3. Schematic of filament winding plate to manufacture unidirectional all-PP composites.
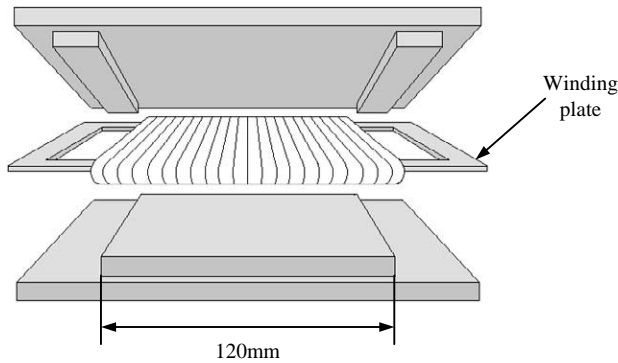
Fig. 4. The filament wound plate (shown in Fig. 3) is subsequently placed in a closed fitting mould and heat and pressure are applied to consolidate the composite.

in Table 4. The dimension of the specimens was limited in some cases by the dimensions of the moulds used to manufacture the specimens. The 'unidirectional' samples can be considered slightly off axis due to the winding process, but since the angle of winding is <0.5°, this is ignored. The notation used to describe the range of tensile and compressive specimens is described in Table 4. Unidirectional specimens (UD140 and UD160) were cut at a range of angles to tape winding direction ($\theta = 0°$, 10°, 30°, 45°, 60°, 90°) from plates compacted at one of two temperatures: 140 and 160 °C.

5. Tensile and compressive testing of unidirectional all-PP composites

Tensile test specimens were tested using an Instron 5584 tensile testing machine, equipped with a 5 kN load cell and data acquisition software. Specimens were placed in composite grips, which allow 10° free rotations to reduce the effect of moments in the off-axis specimens. Tensile
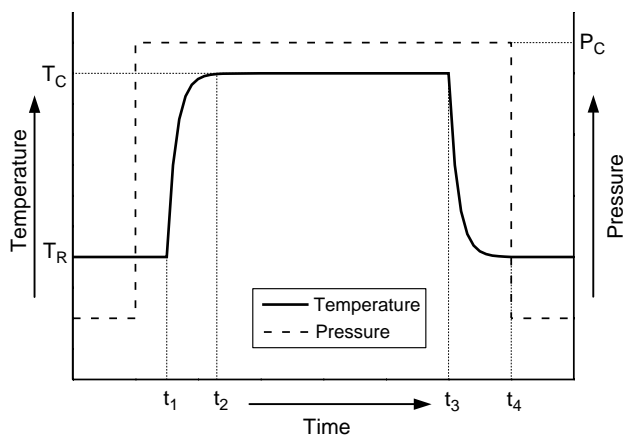

Fig. 5. Time-temperature and time-pressure profiles during consolidation of all-PP composites. Note that pressure is applied before heating and maintained until cooling is achieved to minimise molecular relaxation during heating [35].

Table 3
Compaction parameters for the production of unidirectional all-PP composites

Compaction temperature, T_C [°C]	140	160
Release temperature, T_R [°C]	40	40
Compaction pressure, P_C [MPa]	2.4	2.4
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

tests were performed at 2 mm min^{-1} with a small preload (~ 1 N). To fully characterise the material, 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 Young's modulus (E-modulus) for each specimen, strain was measured using 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. Each test was performed 5 times 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 specimens cut from plates with a thickness of 5 mm. 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. The machine is operated in a tensile mode but the geometry of the compression cell results in a compressive load being applied to the specimen. The specimen geometry is shown in Table 4. Tests were performed in accordance with ASTM D695 applying compressive loading at a rate of 1.3 mm min^{-1}. The recorded load increases with compression until ultimately it tends to reach a uniform stress. This plateau of constant stress is recorded as

Table 4
Dimensions and tape orientation for tensile and compressive testing specimens of consolidated all-PP laminates

Test	Specimens	Tape orientation	Dimensions[a] [mm]
Tension	UD140/UD160	0°, 10°	120×12×1
Tension	UD140/UD160	30°, 45°, 60°, 90°	100×25×2
Compression	UD140C/ UD160C	0°, 90°	15×5×5

[a] Dimensions given as length×width×thickness.

the compressive strength. As before, five repetitions of each test were performed to provide an accurate measure of the compressive strength. Tests were performed along the longitudinal [0°] and transverse [90°] directions of uni-directional laminates.

6. Laminate theory

Creating unidirectional composite plates from highly oriented polypropylene tapes will clearly lead to highly anisotropic composite structures. In order to collect this information, unidirectional composites are loaded at various angles, θ, to the tape direction.

The tensile modulus, E_θ, of a unidirectional laminate loaded at an angle, θ, to the tape direction, can be predicted by Eq. (1)

$$\frac{1}{E_\theta} = \left[\frac{1}{E_{11}}\right]\cos^4\theta + \left[\frac{1}{G_{12}} - 2\frac{\nu_{12}}{E_{11}}\right]\cos^2\theta\,\sin^2\theta + \left[\frac{1}{E}\right]\sin^4\theta \tag{1}$$

where G_{12} = shear modulus, ν_{12} = major Poisson ratio, E_{11} = longitudinal tensile modulus, E_{22} = transverse tensile modulus, and the 1 and 2 direction refer to the tape direction and perpendicular to the tape direction out of the plane of the tape. E_{11}, E_{22}, and ν_{12} can be determined experimentally by performing longitudinal ($\theta = 0°$) and transverse ($\theta = 90°$) tensile tests on unidirectional composites. If $\theta = 45°$, then Eq. (1) can be rearranged to give:

$$\frac{1}{G_{12}} = \left[\frac{4}{E_{45}}\right] - \left[\frac{1-2\nu_{12}}{E_{11}}\right] - \left[\frac{1}{E_{22}}\right] \tag{2}$$

Thus shear modulus, G_{12}, can be determined by testing unidirectional composites loaded at $\theta = 45°$.

The tensile strength, σ_θ, of a unidirectional composite loaded at an angle, θ, to the tape direction can be predicted using anisotropic failure criteria models. Two models will be considered here: the maximum stress criterion and the Tsai-Hill criterion. The maximum stress criterion dictates that there are three discrete failure modes, one of which will be dominant depending on the angle of loading, θ, and the weakest of these strengths will thus determine the failure mode of the composite laminate. For small angles, typically $0° < \theta < 10°$, composite strength, σ_θ, is controlled by failure in the fibre direction, σ_{11},

$$\sigma_\theta = \frac{\sigma_{11}}{\cos^2\theta} \tag{3a}$$

As θ increases, σ_θ is dominated by intralaminar shear strength, τ_{12},

$$\sigma_\theta = \frac{\tau_{12}}{\cos\theta\,\sin\theta} \tag{3b}$$

As θ approaches 90°, σ_θ is dominated by transverse strength σ_{22},

$$\sigma_\theta = \frac{\sigma_{22}}{\sin^2\theta} \tag{3c}$$

These criteria consider that the three failure modes act in isolation and when the threshold of the weakest failure mode is reached, the specimen fails in this failure mode. However, an adaptation of the Von Mises yield criterion has also been applied to transversely isotropic composites. If a unidirectional composite is loaded in the 1 direction, the Tsai-Hill criterion states [44]:

$$\left(\frac{\sigma_{11}}{\sigma_{11}^*}\right)^2 + \left(\frac{\sigma_{22}}{\sigma_{22}^*}\right)^2 + \left(\frac{\tau_{12}}{\tau_{12}^*}\right)^2 - \frac{\sigma_{11}\sigma_{22}}{(\sigma_{11}^*)^2} = 1 \tag{4}$$

where σ_{11}, σ_{22} = Tensile stress in 1 and 2 directions, τ_{12} = Shear stress in 12 direction and * indicates ultimate stress (strength). In the case of a unidirectional composite loaded at an angle, θ, to the reinforcement direction, this criterion can be adapted to predict the failure stress based on the longitudinal, transverse and shear strengths, and the loading angle:

$$\sigma_\theta = \left[\frac{\cos^2\theta(\cos^2\theta - \sin^2\theta)}{\sigma^2} + \frac{\sin^4\theta}{\sigma_{22}^2} + \frac{\cos^2\theta\,\sin^2\theta}{\tau_{12}^2}\right]^{-1/2} \tag{5}$$

This equation describes a single failure prediction for $0° > \theta > 90°$. The longitudinal and transverse strengths can be simply established experimentally, and the intralaminar shear strength, τ_{12}, of a unidirectional composite can be approximated by loading a 10° off-axis specimen, and applying Eq. (3b) [45]. In principle, these laminate formulae can be applied to whole range of composite materials but they have also been successfully applied to highly anisotropic polypropylenes oriented by roll drawing which, due to the microfibrillar nature of highly oriented PP, can be considered as a unidirectional fibre laminate [46–48].

7. Results and discussion

7.1. Tensile performance of unidirectional laminates

The density of the unidirectional laminates ($\rho = 0.770\ \mathrm{g\ cm^{-3}}$), as measured using the Archimedes principles, is slightly greater than the density of the tapes ($\rho = 0.732\ \mathrm{g\ cm^{-3}}$). This means that consolidation between adjacent tapes is excellent with no intertape voiding, and there is some closure of intratape microvoids during compaction. This is reinforced by Fig. 6, which shows an optical micrograph cross section of a unidirectional laminate in which the excellent consolidation of tapes can be clearly seen. The compacted tapes are visible running into the plane of the page, and the copolymer layer can be seen as a thin, grey line between the tapes, as highlighted in the Fig. 6.

The tensile moduli and tensile strengths of these specimens are presented in Figs. 7 and 8, respectively. Very little difference was seen in E_{11} between UD140 and
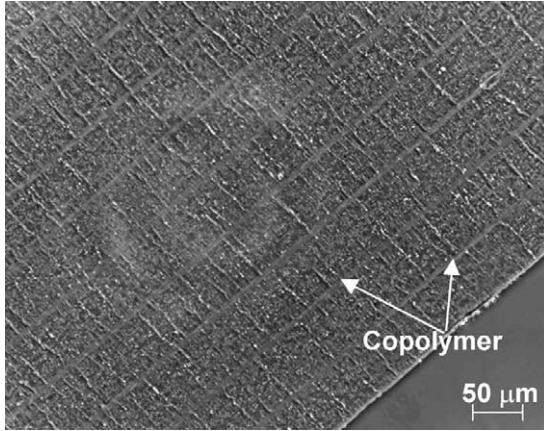
Fig. 6. Optical micrograph of a cross section of unidirectional all-PP laminate showing the skin core morphology of the original tapes still present in the consolidated plate.

UD160, and a greater but still small variation in E_{22} was seen. It is clear that unidirectional all-PP composites produced in this way show increased moduli close to the direction of drawing while the transverse modulus is not significantly greater than isotropic PP (1–1.5 GPa). Fig. 7 shows the variation of modulus with loading angle for UD160, compared to the modulus predicted by Eq. (1).

It can be seen there is very small standard deviation and Eq. (1) describes the behaviour of UD160 very well. Since moduli are very similar for UD140 and UD160, it suggests that this temperature processing window provides uniform stiffness, and also E_{11} (13 ± 0.8 GPa) is $87 \pm 6\%$ the modulus of the tape (15 GPa) showing a good retention of tape modulus in the composite. The off-axis mechanical properties of these unidirectional composites are also shown in Table 5. A small decrease in tensile modulus due to composite consolidation is seen. The anisotropy of these unidirectional composites is similar to that reported for
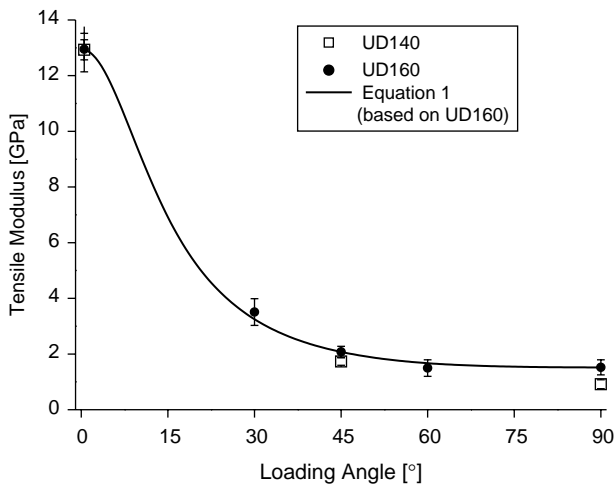

Fig. 7. Tensile modulus vs. loading angle for unidirectional all-PP composites consolidated at 140 and 160 °C, respectively, showing similar mechanical properties for both processing temperatures and a high degree of anisotropy in either case.
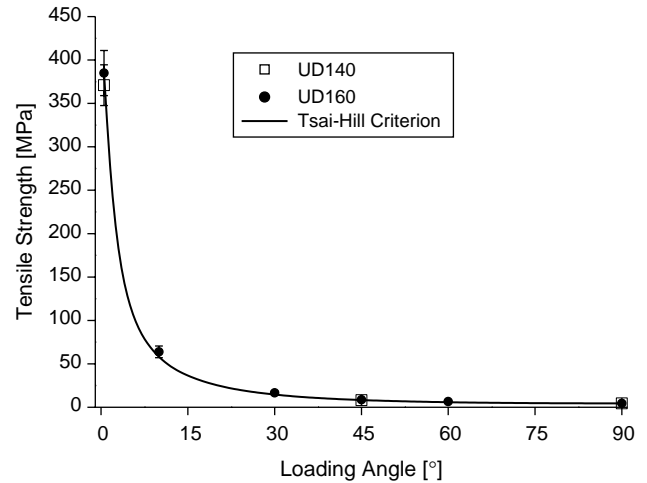

Fig. 8. Tensile strength vs. loading angle for unidirectional all-PP composites consolidated at 140 and 160 °C, respectively, also showing similar mechanical properties for both processing temperatures and a high degree of anisotropy in either case.

polyethylene reinforced polyethylene composites [16] based on ethylene–butene copolymer films reinforced with commercial UHMW-PE fibres. Despite the fact that these UHMW-PE fibres initially possess much greater tensile modulus ($E_{11} = 70$–110 GPa [21,49]) than the PP tapes used in this study ($E_{11} = 15$ GPa), the lower weight fraction of fibre present (65%) and possible molecular relaxation in the UHMW-PE fibre during composite processing means that tensile performance ($E_{11} = 20.6$ GPa, $\sigma_{11} = 700$ GPa) is only 60–80% greater than that of the all-PP composites ($E_{11} = 13$ GPa, $\sigma_{11} = 385$ MPa) presented in this study. Nevertheless, the high performance of UHMW-PE fibres indicates there are clear potential advantages of using an all-PE composite for applications, which will not be exposed to elevated temperatures, such as biomedical applications [50–52].

The tensile strength as a function of loading angle is shown in Fig. 8, based on UD140 and UD160. Similarly to off-axis moduli, off-axis strengths, σ_θ, are very similar for

Table 5
Tensile properties of unidirectional all-PP composite specimens with varying loading angle

Loading angle [°]	Compaction temperature [°C]	Tensile modulus [GPa]	Tensile strength [MPa]	Strain to failure [%]
0	Uncompacted tape	15	450	7
0	140	13	371	8
45	140	1.7	8.2	1
90	140	1	4.5	1
0	160	13	385	8
10	160	–	64	2
30	160	3.5	17	1
45	160	2.1	8.7	1
60	160	1.5	6.5	1
90	160	1.5	4.5	1

Table 6
Degree of anisotropy of unidirectional all-PP composites compared to unidirectional glass-PP composites [53]

Composite system	σ_{11} [MPa]	σ_{22} [MPa]	σ_{11}/σ_{22} [–]
All-PP	385	4.5	85.6
Glass/PP	1025	4.5	277.8

UD140 and UD160. The unidirectional composites are clearly very anisotropic, and while Fig. 7 shows that while $E_{\theta=30} \approx 16\% E_{11}$, $\sigma_{\theta=30} < 5\% \sigma_{11}$. The degree of anisotropy can be described as the ratio of σ_{11} to σ_{22} and in the case of these all-PP composites, this ratio is 85.6. This is almost three times that seen in unidirectional glass fibre reinforced polypropylene [53], but this is due to the greater σ_{11} value of glass/PP composites since σ_{22} is similar (see Table 6). Therefore, while the transverse modulus (E_{22}) of these composites is comparable to the modulus of isotropic PP, the transverse strength (σ_{22}) is much less than the tensile strength of isotropic PP.

The transition from tensile failure to transverse failure of tapes can be predicted by Eq. 3(a)–(c), and is shown in Fig. 9. These maximum stress criteria suggest that at $\theta \approx 2°$, failure mode operates via a shearing mechanism and this dominates until $\theta \approx 23°$, and these criteria describe the experimental data very well, as does the single Tsai-Hill criterion shown in Fig. 8. The failure of unidirectional specimens is dominated by tape strength at $\theta = 0$, but changes to transverse tape strength as $\theta > 25°$.

Fig. 10 shows a photograph of failed unidirectional specimens tested at $\theta = 0°$, 10° 60° and 90°. The specimen loaded at $\theta = 0°$ shows a large amount of fibrillation and no single fracture surface. This is to be expected since the transverse strength of the tapes is so poor that cracks rapidly propagate along the tape direction. It is also clear that a combination of inter-tape debonding and intra-tape fibrillation leads to fibrils being pulled from the composite. This

Fig. 10. Photograph of failed unidirectional specimens (left to right: $\theta = 0°$, 10°, 60° and 90°) illustrating a transition from a highly fibrillar failure ($\theta = 0°$) to transverse failure ($\theta > 10°$).

reduces load transfer and it is likely that what is being seen here, is not true tensile failure but instead failure by delamination. This may explain the difference between the tensile strength of a single tape (450 MPa), in which definite tape tensile failure occurs, compared to the tensile strength of a unidirectional composite (380 MPa) which shows fibrillation and some fibril pull-out. Fig. 11 shows a composite optical micrograph of a unidirectional all-PP specimen loaded at $\theta = 0°$, in which the complete fibrillation of the composite can be seen.

The low transverse strength is partially due to the extremely poor transverse strength of the tape due to high orientation, which limits the transverse composite strength, but also due to the tape geometry. Since the width of the tape is $\sim 36\times$ greater than the thickness of the tape, a large overlap in the transverse direction is present providing excellent load transfer between tapes [54]. Thus transverse
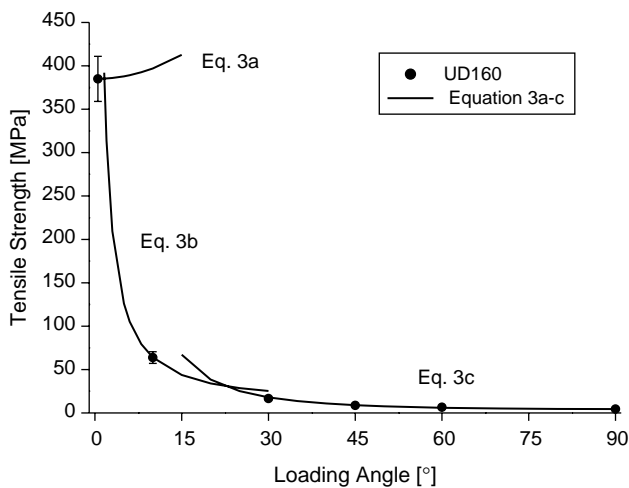

Fig. 9. Tensile strength vs. loading angle for unidirectional all-PP composites with predicted failure mechanism described by the maximum stress criterion (Eq. (3a–c)).
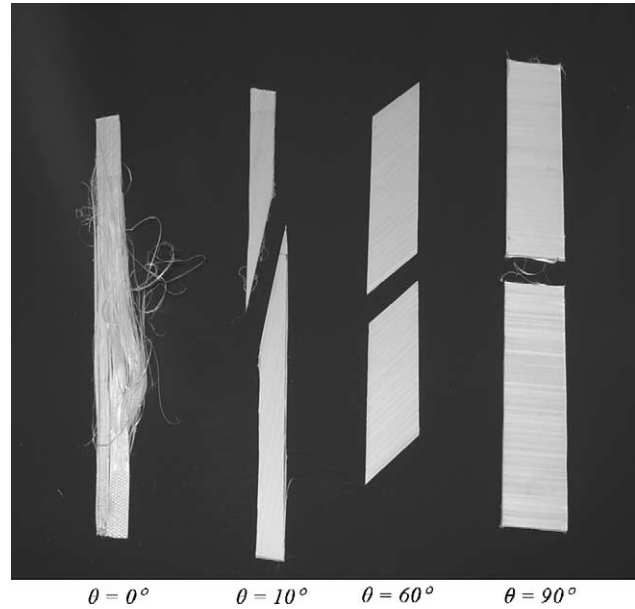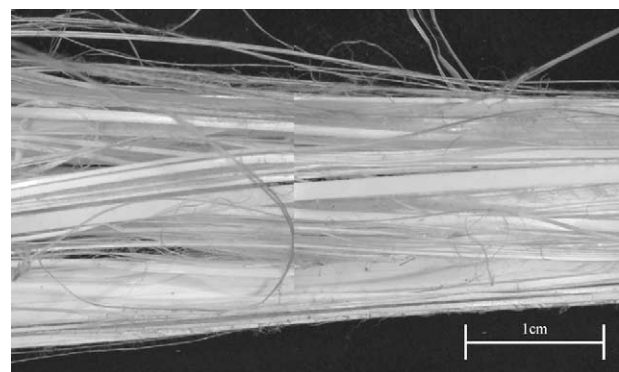

Fig. 11. Composite optical micrograph of the fibrillated surface of a unidirectional all-PP composite loaded in the tape direction ($\theta = 0°$).
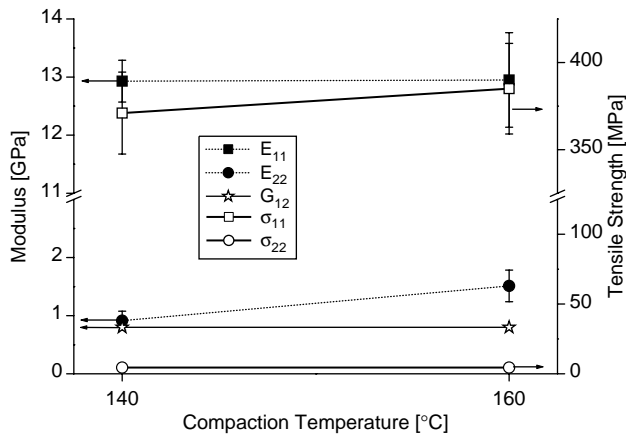
Fig. 12. Summary of unidirectional composite tensile properties vs. compaction temperature showing approximately constant mechanical properties with processing temperature, validating the large temperature processing window which can be achieved by using co-extruded tape technology.

failure of a unidirectional composite by an interfacial failure mode would theoretically occur by pull-out of the tape width from the surrounding tapes, but this is very unlikely since the transverse strength of the tape is so poor compared to the shear strength of the interface. The interlaminar shear strength of the composite can be obtained by rearranging Eq. (3b). At $\theta = 10°$, τ_{12} can be predicted since $\tau_{12} = \sigma_{\theta=10}$

$$\frac{\tau_{12}}{\sigma_{11}} \approx 0.171 \qquad (6)$$

From Table 5, it can be seen that Eq. (6) gives $\tau_{12} = 65.8$ MPa. This agrees well with the experimentally determined value for $\sigma_{\theta=10}$ of 64 MPa. The shear modulus, G_{12}, determined by Eq. (2), has a value of ~ 0.8 GPa for specimens compacted at both temperatures. Fig. 12 summarises these mechanical properties as a function of compaction temperature. This shows that increasing the compaction temperature between 140 and 160 °C, has only a small effect on mechanical properties. The transverse modulus and longitudinal strength actually increase slightly with increasing compaction temperature, although the increase in longitudinal strength is within the experimental scatter of the results, and the transverse modulus at either temperature is too low to be of major interest.

7.2. Compressive performance of unidirectional laminates

Fig. 13 shows the compressive strength of unidirectional composites tested in the longitudinal direction, $UD\sigma_{11C}$, and the transverse direction, $UD\sigma_{22C}$, compacted at 140 and 160 °C. There is no increase in the transverse compressive strength of the unidirectional composite but there is a noticeable increase in compressive strength in the longitudinal direction.

The drawing mechanism increases tensile properties by aligning the microstructure of the material in the drawing
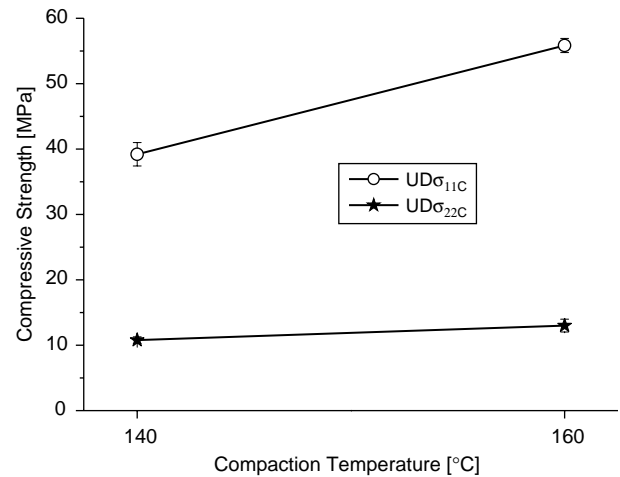

Fig. 13. Summary of unidirectional all-PP composite compressive strengths vs. compaction temperature showing a constant, but relatively low, transverse compressive strength and a longitudinal compressive strength which increases with processing temperature, but is also very low compared to tensile strength.

direction and so it is unlikely that any increase would be seen in compressive properties of tapes. A compressive strength of oriented polypropylene of ~ 40 MPa has been reported for draw ratios ≤ 5, and seems to be independent of draw ratio in this region [55]. Failure in these specimens is also by micro buckling. In fact, failure is reported to occur along shear planes, and therefore, a higher orientation structure may be more susceptible to buckling than an isotropic structure [56,57]. Even highly oriented polyethylene fibres, which show a much higher tensile strength (3 GPa) exhibit a very low compressive strength (70 MPa) [58]. This poor compressive strength must be taken into account when designing all-PP products, which will experience flexural loads. 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 likely that no benefit is gained by using this highly oriented PP rather than bulk PP for pure compressive applications.

The mechanical properties of unidirectional all-PP composite laminates reported in this paper are summarised in Table 7, together with the mechanical properties of a commercial unidirectional glass fibre reinforced PP roving containing 75% weight fraction of glass fibre (Twintex®, Saint Gobain-Vetrotex). The longitudinal mechanical properties of unidirectional all-PP composites are less than that of unidirectional glass fibre reinforced PP, but due to the low density of PP tapes reported in this paper, the specific tensile strength of unidirectional all-PP composites is comparable to that of unidirectional glass reinforced PP while the specific tensile modulus of unidirectional all-PP is slightly less than that of unidirectional glass reinforced PP.

Table 7
Mechanical and specific mechanical properties of unidirectional all-PP composites compared to a commercial unidirectional glass fibre reinforced PP

Material	All-PP		Commercial PP/Glass 50%V_f UD roving
Compaction Temperature [°C]	140 °C	160 °C	–
Compaction Pressure [MPa]	2.4	2.4	–
Density, [g cm^{-3}]	0.77	0.77	1.75
E_{11}, [GPa]	12.93	12.95	38
E_{11}/ρ [GPa g^{-1} cm^3]	16.79	16.82	21.71
E_{22} [GPa]	0.91	1.52	–
E_{22}/ρ [GPa g^{-1} cm^3]	1.18	1.97	–
σ_{11} [MPa]	371	385	800
σ_{11}/ρ [MPa g^{-1} cm^3]	482	500	457
σ_{22} [MPa]	4.5	4.5	–
σ_{22}/ρ [MPa g^{-1} cm^3]	5.84	5.84	–
σ_{11C} [MPa]	39.2	55.8	–
σ_{22C} [MPa]	10.8	13	–
ν_{12}	0.36	0.38	–
ν_{21}	0.06	0.06	–
G_{12} [GPa]	0.8	0.8	–

8. Conclusions

All-polypropylene composites can be successfully created with high fibre volume fractions ($>90\%$) possessing high tensile moduli and strengths. By using a combination of constraining and co-extrusion, the temperature window for creating these composites can be greater than 30 °C. Despite the high temperatures involved during the compaction process, the excellent mechanical properties of the oriented tapes are retained in the resulting unidirectional composites. Furthermore, the composite is not sensitive to deviations in the process temperature since mechanical properties proved approximately constant within the processing temperature window investigated here. The specific mechanical properties are comparable to those reported for a commercial unidirectional glass fibre reinforced PP, while the all-PP composites clearly have great advantages in terms of recyclability.

Acknowledgements

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