

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 ($> 30\text{ }^{\circ}\text{C}$) 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 ($\sim 250\text{ GPa}$) is much greater than the crystal lattice modulus of the helical PP molecule ($\sim 40\text{ GPa}$) [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*

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