



# The influence of process parameters on the properties of hot compacted self-reinforced polypropylene composites



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## ABSTRACT

Hot compacted self-reinforced polypropylene composites have good tensile properties and excellent impact resistance, but they have a limited processing window. Therefore, the influence of compaction temperature, dwell time and the application of interleaved films on the tensile and impact properties was assessed. Increased compaction temperature allows more molecular relaxation, thereby melting more matrix and creating a stronger interlayer bonding. This results in reduced 0° tensile properties and penetration impact resistance, while the 45° tensile properties and non-penetration impact resistance are maintained or improved. The dwell time only has minor influences on tensile and impact properties, while interleaved films have a similar influence as increased compaction temperature. These films increase the interlayer bonding, which increases the tensile strength and non-penetration impact resistance, but reduces penetration impact resistance. This paper demonstrates a wide property range depending on the processing parameters, helping in future tailoring of self-reinforced composites to specific applications.

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## 1. Introduction

Polymers are increasingly replacing traditional materials such as wood, ceramics and metals. Their relatively low stiffness and strength hampers their applicability to compete in many applications. These properties can be improved by reinforcement fibres, such as carbon and glass fibres. The interface strength between the fibre and polymer, however, is a traditional problem. An elegant solution was proposed by Capiati and Porter [1], who invented the first self-reinforced composite, also referred to as single polymer composites or all-polymer composites. The reinforcing phase, which is an oriented polymer fibre or tape, is combined with an unoriented matrix phase from the same polymer. The molecular orientation of the reinforcing phase imparts excellent mechanical properties to these self-reinforced composites.

Several processes have been invented to create self-reinforced composites. Capiati and Porter's initial process [1] stacked films of low molecular weight PE in between the layers of high molecular weight PE fibres. The difference in melting point and orientation of both PE grades was exploited to create the first self-reinforced

composites. In 1993, Hine et al. [2] reported a new approach, which starts off from an oriented homopolymer fibre. Their process, called hot compaction, exploits the difference in melting temperature between the outer sheath and the inner core of the fibre. This process has a narrow, yet viable temperature window. An alternative process is co-extrusion, developed by Peijs and co-workers [3–6]. This process uses bi-component tapes, in which the outer layer is a lower-melting point copolymer and the inner layer is a homopolymer. This increases the processing window from a few degrees [7–9] to a few tens of degrees Celsius [3]. The main advantage is that co-extrusion works at a lower temperature and can therefore maintain higher mechanical properties. Some other processes have also been investigated, such as injection moulding [10] and continuous extrusion [11], but have not yet led to commercial exploitation. The most successful self-reinforced polymer is polypropylene (PP), as it excels in impact performance and processability [12–15].

Self-reinforced PP composites have better tensile properties than isotropic polypropylene [12], but they are weaker and more compliant than traditional fibre-reinforced composites. Therefore, most of the current applications exploit its excellent impact resistance [12]. Alcock et al. [5] demonstrated the influence of the processing conditions on the impact resistance of co-extruded self-reinforced PP. They established delamination, tape fracture, and tape debonding as the main energy absorbing mechanisms and

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proved that the penetration impact resistance decreased with increased compaction pressure or temperature. This is mainly the case at low compaction temperatures where insufficient surface welding occurs to achieve proper bonding. At higher compaction temperatures, the influence seems to level off. The decrease in penetration impact resistance with increased temperature was also confirmed by Bárány et al. [16].

Further investigation into the effects on impact velocity and test temperature were performed by Alcock et al. [17]. Aurrekoetxea et al. [18] performed repeated impact tests and established an energy threshold below which no plastic deformation occurs. Swolfs et al. [19] proved that there is only a small influence of the weave architecture on the penetration impact resistance, while Abraham et al. [20] demonstrated the importance of the type of PP polymorph on the penetration impact resistance in self-reinforced PP.

The addition of interleaved films is another way to optimise the processing and mechanical properties of self-reinforced composites. While many studies have investigated interleaved films in traditional fibre-reinforced composites [21,22], their main goal was to increase fracture toughness and damage resistance in carbon fibre-reinforced composites. In contrast, the main goal of interleaved films in self-reinforced composites is to achieve wider processing window. These films also increases the peel strength and hence the interlayer bonding [19,23,24] by creating more matrix in between the fabric layers in hot compacted self-reinforced PP composites. Unfortunately, these films are known to reduce the penetration impact resistance [19], but it is not clear whether this is valid at other compaction temperatures and dwell times. The influence of the interleaved films on non-penetration impact resistance is currently unknown.

This study assesses the influence of the process parameters for hot compacted self-reinforced PP composites. This influence has already been investigated for co-extruded self-reinforced PP composites by Alcock et al. [5]. The hot compaction process is much more sensitive to the process parameters, making it even more important to understand those influences for hot compaction. Furthermore, the study of Alcock et al. used strain mapping to determine the degree of plastic deformation, which does not measure the damage after a non-penetration impact event. Instead, this study uses ultrasonic C-scans, which leads to a more direct measurement of the extent of the damage [25].

## 2. Materials and methods

### 2.1. Materials

Propex Fabrics GmbH provided balanced twill 2/2 PP tape weaves with an areal density of 130 g/m<sup>2</sup>. These PP tapes had a stiffness of 6.9 ± 1.2 GPa and a strength of 589 + 24 MPa [26]. The weaves were overfed, a feature which is discussed in detail in Swolfs et al. [19]. Propex Fabrics GmbH also provided 20 µm thick films of the same homopolymer PP grade as the tapes. These films have melting temperature of 163 °C.

### 2.2. Hot compaction

The weave was cut into layers of 320 × 320 mm under 0° and 45°. The 45° layers were used to produce samples for 45° tensile tests. A total of eight weave layers were stacked on top of each other to obtain samples without interleaved films. In other layouts, a single film was interleaved in between each of the weave layers.

This assembly was placed between two 1 mm thick aluminium cover plates, which was then inserted into a Fontijne LabPro400 press. This press was preheated to the correct temperature, within

the range of 180–194 °C. A pressure of 39 bar was applied throughout the process. The assembly was held at the pre-set temperature for 2, 5 or 15 min. After the dwell time, the assembly was cooled down to 40 °C in 5 min. Unless otherwise mentioned, the standard dwell time and compaction temperature were 5 min and 188 °C.

### 2.3. Differential scanning calorimetry

Differential scanning calorimetry (DSC) samples were cut from the middle of the hot compacted plates, as that location has the most reproducible temperature profile during hot compaction. The samples with a 2 mg nominal weight were tested in a TA instruments Q2000. These low sample weights are needed to obtain accurate DSC measurements in self-reinforced composites. Despite the low sample weights, the DSC thermograms were reproducible, showing that the relative amount of tapes and matrix was similar in all cases.

The samples were heated from room temperature to 200 °C at 10 °C/min, with a constant flow of 50 ml air/min. At least four samples were tested for each configuration. The melting temperature was determined at the maximum of the heat capacity versus temperature.

### 2.4. Tensile tests

Tensile tests were performed on an Instron 4505 tensile machine, with hydraulic clamps and a 100 kN load cell. The load cell was electronically scaled to 10 kN to improve its accuracy. The tensile samples measured 250 by 25 mm and were tested at a gauge length of 150 mm, according to ASTM D3039. A minimum of 5 samples was tested for each processing condition. Sand paper was used in all cases to avoid slipping in the clamps. The strain was measured by digital image correlation of images of a speckle pattern on the sample surface, taken every 250 ms. The modulus was calculated in the strain range 0.1–0.3%. The strength was determined as the maximum stress in the tensile diagram and the corresponding strain is defined as the failure strain.

### 2.5. Impact tests

Two types of impact tests were performed: penetration and non-penetration. This terminology is used throughout, as it reflects more directly the difference between both types of impact than the terminology high and low energy impact. Falling weight impact tests were performed on a Fractovis CEAST 6789 machine, according to ISO 6603-2. In both cases, a hemispherical striker with 20 mm diameter was used to impact 100 × 100 mm samples. All samples were clamped with a pressure of 9 bar and were tested at room temperature.

For the penetration impact tests, a 26.17 kg striker was dropped from 1 m height, resulting in an impact energy of 257 J which was sufficient to cause penetration. These samples were clamped by a support ring with an inner and outer diameter of 40 mm and 60 mm respectively. The load was registered by a 20 kN load cell in the striker tip, while the displacement was measured using a laser. Six samples were tested for each configuration. The penetration impact energy was calculated according to ISO 6603 as the area underneath the force–displacement diagram until the load has dropped below half of the peak load. This value was divided by the sample thickness.

For the non-penetration impact tests, a 3.17 kg striker was dropped from the same machine. The drop height was adjusted to achieve 5 J and 15 J impacts, both of which are energies high enough to induce damage but low enough to avoid penetration. The clamp had an inner and outer diameter of 80 and 100 mm respectively. This was deliberately chosen larger than for

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