



Failure behaviour of self-reinforced polypropylene at and below room temperature



Y. Swolfs*, W. Van den fonteyne, J. Baets, I. Verpoest

Department of Materials Engineering, KU Leuven, Kasteelpark Arenberg 44, Belgium

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ABSTRACT

Self-reinforced polypropylene is a very tough material. It is even thought that its impact resistance increases with decreasing temperature. This was investigated by examining the constituent tapes and matrix. Tensile tests on both drawn polypropylene tapes and self-reinforced polypropylene were similar: the stiffness increased and the failure strain slightly decreased at low temperatures. The matrix, however, embrittled below room temperature due to the glass transition. In contrast with literature data on Izod impact resistance, the penetration impact resistance did not increase at low temperatures. At lower temperatures, the damaged area after non-penetration impact was significantly reduced. This was caused by a change in the damage mode from tape–matrix debonding to matrix cracking, as the matrix went through its glass transition. These conclusions provide the first understanding of the failure behaviour of self-reinforced polypropylene below room temperature, and can be exploited to further optimise the excellent impact resistance of self-reinforced polymers.

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1. Introduction

Fibre-reinforced polymers are known for their exceptionally high specific stiffness and strength, but often lack toughness. A promising solution is the use of self-reinforced polymers, in which reinforcement and matrix are made from the same polymer. The commercially most important example is self-reinforced polypropylene (SRPP) [1–3].

The starting material for SRPP is highly drawn polypropylene (PP) fibre or tape. The drawing process enhances the molecular orientation of the PP, which improves the mechanical properties in the drawing direction [4,5]. These fibres or tapes are then woven in the preferred configuration [6–8] and compacted at elevated temperature and pressure into a self-reinforced sheet. Several techniques exist for creating the matrix. The most important techniques are film stacking [9,10], co-extrusion [7,11,12] and hot compaction [1,6,13–17]. The focus here is on the hot compaction process. In this process, a homopolymer tape is used. By selecting the correct combination of temperature and pressure, it succeeds in selectively melting the outer sheath of the drawn tapes. This molten material then recrystallises to create the matrix [1,18].

The final SRPP properties depend on the properties of the two components, just as in classical fibre-reinforced polymers [13]. The properties of the oriented PP tapes before compaction strongly depend on the conditions of the drawing process, whereas the properties of the PP matrix and thus the tape-to-tape and interlayer bonding in the compacted sheet depend on the conditions of the hot compaction process [5,6,12].

The tape properties are mainly influenced by the draw ratio and temperature. In general, the higher the draw ratio, the higher the stiffness and strength of the final tapes will be. There is, however, a maximum attainable draw ratio, which is determined by the nature of the bonds and the chain extension in the polymer [19]. This maximum draw ratio depends on the molecular weight of the polymer [5,20]. The yield stress and the failure strain of the tapes depend on the drawing temperature. A lower drawing temperature results in a lower yield stress and a larger failure strain, as the drawing temperature determines the crystal structure variations during the drawing process. This has been investigated in detail by Schimanski et al. [5]. Furthermore, annealing treatments have been shown to affect the thermal and mechanical properties of drawn PP tapes [20].

The compaction temperature, pressure and time determine the degree to which the tape properties are maintained as well as the tape-to-tape and interlayer bonding strength in the final SRPP [6]. In case of woven SRPP, the mechanical properties in the [0°/90°] direction are determined by the tape properties after hot

* Corresponding author. Tel.: +32 16 37 36 16; fax: +32 16 32 19 90.

E-mail addresses: yentl.swolfs@mtm.kuleuven.be (Y. Swolfs), pywvdf@leeds.ac.uk (W. Van den fonteyne), joris.baets@mtm.kuleuven.be (J. Baets), ignaas.verpoest@mtm.kuleuven.be (I. Verpoest).

compaction, while the properties in the $[\pm 45^\circ]$ direction are dominated by the consolidation quality and the matrix fraction. A higher processing temperature and/or pressure leads to a higher interfacial strength [7]. A well-chosen processing temperature leads to full consolidation while retaining the good mechanical properties of the PP tapes. The processing also anneals the tapes, which has been proven to further refine the crystal structure [6].

The processing parameters and hence the properties of the final self-reinforced polymer may be chosen differently in function of the application. As the dominant failure modes in impact are tape fracture, tape–matrix debonding and delamination, the penetration impact energy of SRPP benefits from a lower processing temperature and/or pressure, because this facilitates delaminations. SRPP that is optimised for penetration impact, however, may not possess adequate interfacial properties to serve as a viable structural component [12]. SRPP outperforms glass or natural fibre reinforced PP in terms of impact resistance. Commercial SRPP even shows an increase of 50% in notched Izod impact resistance at a temperature of -40°C , compared to at room temperature [1,21].

The influence of the drawing process parameters on the mechanical properties of the PP tapes and the influence of the processing parameters on the mechanical properties of the final SRPP were extensively studied [4,5,22–24]. However, these studies do not explain the impact behaviour of SRPP at low temperature. In particular, its increasing toughness at low temperature is unique for polymeric materials. In this paper, the SRPP failure behaviour at different temperatures is analysed with a focus on impact behaviour. The mechanical properties of the matrix and the drawn PP tapes are studied separately at and below room temperature, to analyse the influence of temperature on the different components of the SRPP. This leads to the first explanation of the impact resistance of SRPP below room temperature.

2. Materials and methods

2.1. Materials

Drawn polypropylene (PP) tapes and non-drawn PP films of the same PP grade were kindly provided by Propex Fabrics GmbH (Gronau, Germany). The non-drawn PP film is $20\ \mu\text{m}$ thick and can be considered to be the same material as the matrix in the final SRPP. The tapes were also provided in a twill 2/2 weave with an areal density of $130\ \text{g/m}^2$.

2.2. Hot compaction

The production method is a combination of hot compaction and film stacking [25,26]. Eight layers of the PP tape fabric alternated with PP film were stacked and placed in between aluminium cover plates. This assembly was inserted into a Fontijne Grotnes LabPro 400 press, which was preheated to 188°C . This temperature was maintained for 5 min, after which the press was cooled down to 40°C in 4 min. The pressure was kept constant at 40 bar. More information on how these processing conditions were optimised can be found in [27].

Samples for peel tests were made by stacking 4 layers of PP tape fabric alternated with PP films. A $13\ \mu\text{m}$ Upilex release film was inserted between the second and third fabric and the same processing conditions were applied.

To be able to compare the toughness of SRPP with that of isotropic PP, isotropic PP plates were produced from the non-drawn PP film. To produce these plates, the same hot compaction conditions were applied to a stack of PP films.

2.3. Annealing treatment

To study the evolution of the properties of the PP tapes inside SRPP during the hot compaction process, an annealing treatment was applied to the individual PP tapes. To avoid shrinkage of the tapes, they were kept under tension during the treatment. The tapes were drawn through a stainless steel die, and were in contact with the die for 10 s. The temperature was measured in the middle of the die with a thermocouple. Five different temperatures were used for annealing: 163°C , 176°C , 182°C , 186°C and 190°C . Higher temperatures were not possible without melting the tapes.

2.4. Tensile tests

Tensile tests were performed using an Instron 5985 at 20°C , -10°C and -40°C in a chamber cooled using liquid nitrogen. The tests at 20°C were performed at a deformation rate of 25%/min, using a load cell of 1 kN. Due to restrictions of the cooling chamber, a 30 kN load cell was used for the lower temperatures.

Three types of materials were tested: matrix, tapes and hot compacted SRPP. At least six samples were tested for each material type. For the matrix, dog bone samples with 60 mm gauge length, 10 mm width and 1 mm thickness were tested. For the tapes, the gauge length was 100 mm. Paper tape was wrapped around the tape in the clamping region to ensure proper gripping. For the SRPP, rectangular samples of $250 \times 25 \times 1.3\ \text{mm}$ were tested. The strain in the matrix samples was measured using a contact extensometer, while an optical extensometer was used for the tapes and SRPP samples.

The stiffness was calculated between 0.1% and 0.3% strain. The energy absorbed by the sample during the test was calculated from the integral of the stress in the sample over the strain from the start of the test until the stress drops to 40% of the maximum stress, resulting in an energy per volume.

2.5. Peel strength tests

To assess the interlayer bonding, T-peel tests were performed according to ASTM D1876. The samples were cut down to a width of 25 mm width, with an unbonded length of 76 mm width. Samples from three different plates were tested in a random fashion. The unbonded sample ends were pulled apart at a displacement rate of 254 mm/min in an Instron 5985 tensile machine, equipped with a 30 kN load cell. The tests were performed at 20°C , -10°C and -40°C in a cooling chamber. At least 11 samples were tested for each temperature. The peel strength was calculated as the average load per width of the sample for the first 127 mm of displacement after the initial load peak.

2.6. Falling weight impact tests

The impact behaviour of SRPP was investigated by performing falling weight impact tests on a Fractovis CEAST 6789. This device has a hemispherical striker with a 20 mm diameter. The sample was clamped at a pressure of 9 bar. The tests were performed at 20°C , -10°C and -40°C using liquid nitrogen cooling. At least eight samples were tested for each combination of temperature and impact energy.

For penetration impact tests, the striker was set to a height of 1 m and the inner clamping diameter was 40 mm. The mass of the striker was 8.17 kg. The energy absorption was calculated from the surface underneath the load–displacement curve, until the load dropped to half of the peak load.

The non-penetration impact tests were performed using an 80 mm inner clamping diameter. The larger diameter compared to penetration tests facilitates the registration of the damaged area

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