



Properties of polypropylene single-polymer composites produced by the undercooling melt film stacking method



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ARTICLE INFO

Article history:

Received 10 October 2013

Received in revised form 16 June 2014

Accepted 7 December 2014

Available online 12 December 2014

Keywords:

A. Polymer-matrix composites (PMCs)

B. Mechanical properties

B. Interfacial strength

D. Scanning electron microscopy (SEM)

ABSTRACT

Polypropylene (PP) single-polymer composites (SPCs) were produced by the undercooling melt film stacking method within a large usable processing temperature window from 125 to 150 °C. Samples with good stiffness and strength could be made using 145 ± 5 °C. The undercooling compaction temperatures were all lower than the hot compaction temperatures in traditional processing. Tensile and peel tests were carried out. The effects of compaction temperature, holding time and compaction pressure on mechanical properties of the PP SPCs were investigated. The compaction temperature is the main factor in influencing the properties of the PP SPCs. At different compaction temperature, both holding time and compaction pressure have different influences. Especially, the holding time at undercooling temperature showed different effects compared with the traditional SPCs processing. The morphological properties of the PP SPCs were also examined, good interfacial adhesion were shown.

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1. Introduction

Single-polymer composites (SPCs) refer to the class of composites in which the matrix and the reinforcement come from the same polymer. Compared with traditional polymer composites, SPCs offer the promise of superior mechanical properties and reduced weight, but also provide a real world solution for improving recyclability. The future of SPCs looks promising due to continuing improvement in their preparation and properties, market growth, and recyclability [1]. Polypropylene (PP) is usually reinforced with glass, carbon, aramid fibres or fabrics. Although excellent mechanical properties have been achieved, limited recyclability and interface problem of traditional PP composites have been restricted [2]. PP SPCs seems to be an alternative in this aspect. However, because the reinforcement also comes from PP, traditional preparation methods are not suitable. The difference between the melting temperatures of fibre and matrix determines the processing window. Abo El-Maaty et al. [3] were the first to prepare PP SPCs. Since then, different methods have been developed for preparing PP SPCs, including hot compaction of fibres/tapes [4–7], film stacking [8–17], combination of hot compaction and film stacking [18], and compression of co-extruded tapes [19–22].

PP SPCs have been investigated for a long time. The originality of our investigation is the undercooling melt film stacking method. Undercooling refers to cooling a substance below a phase-transition temperature without the transition occurring [23]. The matrix is firstly melted and cooled fast into undercooled state, then the temperature lower than melting temperature of the matrix was used to compact the composites. Compared with the other methods [3–22], the processing temperature window of this undercooling melt film stacking method depends on the difference between the melting onset temperature of the fibre and the crystallization onset temperature of the matrix, which is much wider than the melting temperature difference between the fibres and matrix. A processing temperature window of 25 °C was established in the PP SPCs manufacturing [23]. Although a narrower temperature range is applied for a usable modulus and strength, it is still larger than using the hot compaction technique. In order to further obtain the mechanism of the undercooling melt film stacking method, PP SPCs were produced in different conditions. The effects of compaction temperature, holding time and compaction pressure on tensile properties of the PP SPCs were examined. Interfacial and morphological properties of the PP SPCs were also analyzed.

2. Experimental

2.1. Materials

PP granules were supplied by Phillips Sumika Polypropylene Company, with a density of 0.905 g/cm^3 at room temperature

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and a melt flow rate of 3.8 g/10 min at 230 °C. The PP fibre cloth was supplied by Innegrity LLC (Simpsonville, SC). The PP cloth was weaved in a plain structure and each yarn is consisted of 225 continuous filaments with a diameter of 48 µm. The density of the warp and weft is 4.3 threads/cm and 6 threads/cm, respectively. The tensile strength (560 MPa) and modulus (6.6 GPa) of the PP fibre were measured by a universal tensile test machine (5166 Series, Instron Corp.) with a load of 30 kN and test speed of 5 mm/min.

2.2. Specimens preparation

The processing route was described in the previous publication [23]. In this study, different compaction temperatures, holding times and compaction pressures were used. Table 1 summarizes the processing parameters applied in the creation of PP SPCs.

In order to investigate the morphological properties of the PP SPCs, two specimens with only one single fibre were created. In the preparation of the single-fibre specimen, PP granules were pressed into a film with 0.1 mm in thickness firstly. The film was set on a glass slide and heated to 200 °C, then quickly transferred to the second station at an undercooling temperature of 140 °C. At last a single fibre from the PP cloth was inserted into the film and held for 10 min, then cooled to room temperature.

2.3. Mechanical tests

Tensile tests in the warp and weft directions were carried out using a universal tensile test machine (Instron Universal Testing Machine 5166 Series, Instron Corp.). The test specimen was cut from the molded sample using a metallic mold according to DIN-53504. The load was 30 kN, and test speed was 5 mm/min. The measurements were repeated with five specimens in the same conditions.

The universal tensile tester was also used for T-peel test to determine the interfacial strength. The specimens, 10 mm wide and 100 mm long, were tested at a crosshead speed of 5 mm/min. The peel strength was calculated in the range from 30 to 70 mm. An additional Teflon film was employed in the middle of the specimen to create an unbonded region for peeling testing.

2.4. Determination of the fibre weight fraction

In order to determine the fibre weight fraction, the PP cloth was cut into the tensile sample geometry by the metallic mold according to DIN-53504, and then an electronic balance was used to obtain the weight of the cloth (0.205 g) in the tensile SPC sample. The weight of each tensile specimen was also measured by the electronic balance. The fibre weight fraction equals to the weight

of reinforcement divided by the average weight of the tensile specimens.

2.5. Observation of micromorphologies

The microstructure of PP SPCs samples made at 125, 135 and 150 °C was observed by a metallomicroscope (CMM-30E, Sunguang Optical Instrument Co., Ltd). The fracture surfaces of the peeled specimens were examined by a scanning electron microscope (SEM, S-800, Hitachi, Japan) with an accelerating voltage of 8 kV. The observation of the interfacial morphology of the first single-fibre specimen was conducted by a polarized optical microscope (PLM, DM4500P, Leica, Germany). The scanning electron microscope was also used to observe the surface structure of the pulled-out fibre from the second single-fibre specimen.

3. Results and discussion

3.1. Effects of compaction temperature on tensile properties of PP SPCs

According to the thermal analysis [23], the undercooling compaction temperatures are from 125 to 150 °C. They are below the melting onset temperature of the fibres (152 °C) but above the crystallization onset temperature of the matrix (124 °C), so the matrix can keep its liquid state while the fibres will not melt. During processing, the undercooling compaction temperature is one of the most important factors to maximize the adhesion and to keep the original fibre structure. The effects of compaction temperature on the tensile strength and modulus of PP SPCs could be seen obviously in Figs. 1 and 2, though different holding times and compaction pressures were used. The tensile strength and modulus in the weft direction were all higher than that in the warp direction, because the weft density is higher than the warp density. As shown in Fig. 1a and b, the tensile strength in both the warp and weft directions increased with the increase of temperature. At low compaction temperature, lower tensile strength reflects incomplete compaction and bad adhesion property. Samples with good stiffness and strength could be made using 145 ± 5 °C. The tensile strength could be up to a maximum value of 172 MPa in the warp direction and 208 MPa in the weft direction when the compaction temperature increased to 150 °C. The highest tensile strength indicates the improvement of fibre wetting with increasing temperature. However, the tensile modulus increased first then decreased (Fig. 1c and d). The maximum modulus emerged at an intermediate temperature of 135 °C, when matrix fully infiltrated the void spaces between the fibres and no fibres were melted. The subsequent decline in the modulus with increasing temperature is primarily due to the orientation loss of the specimen. The modulus is the interdependence between stress and strain. The melting temperature for PP fibre is around 150 °C. The application of higher temperature around 145 °C causes the skin layer of the fibres in the fabric to soften and molecular interdiffusion occurs between the skin layers of adjacent fibres, the polymer chains start to have some relative movement and cohesiveness may go down leading to a higher value of strain corresponding to the stress developed. Thus the modulus happens to fall down. Fig. 2 shows the similar variation of the tensile strength and modulus with different temperatures.

The variation of the tensile strength of the specimens is also related to the fibre weight fraction in the specimen. When the compaction temperature increased, the viscosity of the matrix was deduced thereby a thinner sheet was obtained at a given compaction pressure. Because the fabric was the same, the film melt was pressed to the around space under a higher compaction temperature, then the fibre weight fraction became higher. Table 2

Table 1
Compaction parameters for the production of PP SPCs.

Compaction temperature (°C)	Holding time (min)	Compaction pressure (MPa)
125	5, 10, 15	9
130	5, 10, 15	9
135	5, 10, 15	9
140	5, 10, 15	9
145	5, 10, 15, 20	9
150	5, 10, 15, 20	9
125	10	6, 9, 12
130	10	6, 9, 12
135	10	6, 9, 12
140	10	6, 9, 12
145	10	6, 9, 12
150	10	6, 9, 12

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