Insert injection molding of polypropylene single-polymer composites

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ABSTRACT

A process for making polypropylene (PP) single-polymer composites (SPCs) by insert injection molding was investigated. PP SPCs with relatively good tensile and interfacial properties were prepared with a relatively short cycle time. Melt-spun PP fibers from the same resin as the matrix were treated in heated silicone oil, with and without tension, to study the changes of fiber properties upon exposure to high temperature. PP SPCs containing 36 wt% lab-made fabric achieved a tensile strength of 70 MPa, 2.3 times that of neat PP. Polarized light microscopy of PP SPCs with uniaxial fibers indicated that different crystalline regions formed from partial melting of fibers along the fiber surface. Differential scanning calorimetry results showed that most fibers could maintain the thermal properties after molding. Voids were not observed in the scanning electron microscopy images of the fracture surface after tensile failure indicating good bonding between the matrix and fibers.

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1. Introduction

Single-polymer composites (SPCs), proposed by Capiati and Porter [1] in 1975, refers to composites with the reinforcement and the matrix made from the same polymer type. SPCs can be recycled by simple thermal processing which arouses increasing interest. They are classified into two-component or one-component SPCs depending on the chemical structure of the reinforcement and the matrix [2,3]. Two-component SPCs involve the same type of polymer with the same chemical composition but different chain configurations. Examples are low-density polyethylene (LDPE)/high-density polyethylene (HDPE) SPCs [4], LDPE/ultra-high molecular weight polyethylene (UHMWPE) SPCs [5], and polypropylene (PP) copolymer/PP homopolymer SPCs [6–8]. The advantage for two-component SPCs is that a relatively wide temperature window can be established due to the considerably large melting temperature difference between the matrix and reinforcement. The wide melting point contrast arises from the different chain structures of the two components. However, the different structures result in a compromised compatibility between the two constituents and reduced recyclability.

One-component SPCs are composed of polymers with the same chemical composition and chemical structure. Such polymers include but not limited to PP [9–11], UHMWPE [12], polyethylene terephthalate (PET) [13], and polyamide 6 [14]. It was reported that the interfacial shear strength for one-component HDPE SPCs was 17 MPa, 7.5 MPa greater than two-component LDPE/HDPE SPCs [15]. Further, when the two constituents are made from the same polymer, it is expected that the compatibility and recyclability can be improved. Loos and Schimanski [16] illustrated this feasibility by means of embedding constrained PP fibers into films made from exactly the same resin as the fibers. Transcrystallization layers were observed along the fiber surface due to the good lattice match between the composite materials and the highly favorable energetics. Currently, the primary process for manufacturing one-component SPCs involves hot compaction of fibers or tapes, including PE [17], PP [18], and PET [19]. Strong bonding between the matrix and the reinforcement is promoted. This is supported by the fact that the fracture surface in hot-compacted SPCs often proceeds through the oriented fibers rather than the interface between the matrix and fibers [20]. However, this method still suffers from the poor processability due to the narrow processing temperature window and the relatively long cycle time on the order of minutes.

Insert injection molding offers the potential for high-volume production of SPCs. A study of PP SPCs using resins with different molecular weight by insert injection molding has been reported [21]. In this study, an insert injection molding method was developed to process one-component SPCs with relatively short cycle time. The feasibility was investigated with PP as a model system.
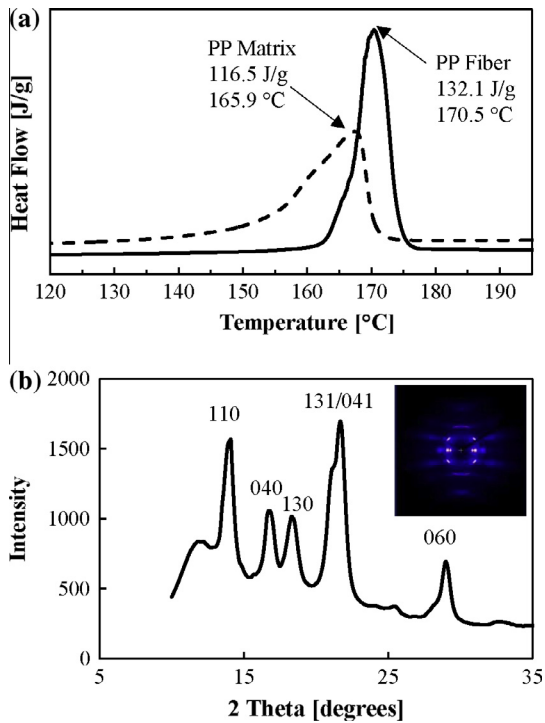
Fig. 1. Properties of melt-spun PP fibers: (a) DSC thermograms and (b) WAXD pattern.

Melt-spun fibers made from the same resin as the matrix were heat treated to experimentally simulate the change of fiber properties during the molding process. Two types of PP SPCs (with lab-made fabric and with uniaxial fibers) were prepared by insert injection molding.

2. Experimental

2.1. Material

PP pellets with trade name Marlex HGZ-1200 were provided by Phillips Sumika Polypropylene Company, with a density of 0.907 g/cm^3 and a melt flow rate of 115 g/10 min at 230 °C. The pellets were melt-spun into filaments and further woven into fabric.

2.2. Melt spinning and heat treatment of fibers

Fiber spinning was carried out by an Alex James and Associates piston extruder. PP pellets were fed into a bore preheated to 180 °C and equilibrated for 1 h. The molten PP was extruded through a 2.5 mm die orifice at 180 °C with a fiber extrusion speed of 1 m/min. The fiber was quenched in ambient air and collected onto bobbins at a speed of 1 m/min. The as-spun PP fibers were subjected to two stages of hot drawing through a heated glycerol bath with a draw ratio of 18× at 140 ± 2 °C and 1.2× at 145 ± 2 °C. After drawing, fibers were briefly rinsed with ethanol to remove the residual glycerol and then dried. The fiber diameter was 102.9 μm.

In order to study the influence of injection process on fiber properties, melt-spun fibers with and without tension were heat-treated in hot silicon oil to simulate the process of fibers contacting with hot melt during injection molding. Fibers were fixed on a metal frame which maintained constant tension during heat treatment. The fibers was submerged into the oil bath and then quickly removed from the bath. The contact time with hot oil was comparable to a short injection stage in molding process. The designed

temperatures of silicon oil, namely pretreatment temperature, were 130–180 °C for fibers with tension and 130–160 °C for fibers without tension. A lower temperature was chosen in the latter case due to the immediate melting of fibers without tension at 170 and 180 °C.

2.3. Composite sample preparation

Two types of one-component PP SPCs were made: SPCs with lab-made fabric for mechanical testing and SPCs with uniaxial fibers for microstructural observation. The fabric and fibers in these SPCs were made from the same PP resin as the matrix. Both the SPCs samples were made using a commercial reciprocated screw injection molding machine (SE-18D, Sumitomo Co.). The mold used in the experiments contained a rectangular cavity of dimensions 63.5 × 9.5 × 1.2 mm and was maintained at a temperature of 20–30 °C. The fibers or fabric were affixed onto the cavity wall along the melt flow direction using double-sided tape. The preparation time for the pre-placing fabric or fibers for each SPCs sample was less than 60 s. The molten PP was injected into the cavity after the mold was closed. Then the material in the cavity was cooled and solidified. After demolding, the double-sided tape was immediately removed from the sample surfaces. For both types of PP SPCs, the injection time and holding time were fixed at 1 and 10 s, respectively. Injection and holding pressures were 207 and 167 kPa, respectively. The cooling time was 10 s. Injection temperatures from 200 to 280 °C were chosen as the major variable to investigate the influence of temperature on the properties of SPCs. In order to analyze the temperature distribution of the melt in the mold cavity, Moldflow analysis was performed using the same experimental conditions that were used for molding the SPCs. Three injection temperatures were used: 200, 240 and 280 °C. The number of elements was 44,601, and 3D tetrahedral mesh was used.

One-component SPCs with lab-made fabric were made to investigate the mechanical property. The fabric was made in the laboratory by plain weaving using melt-spun PP fibers. The thickness of the fabric was about 0.2 mm. Each yarn had 20 filaments and each fabric had 6 warp yarns. The fabric thread count was 5 yarn/cm in both the warp and weft directions. The warp yarns of the fabric were parallel to the injection direction. Each composite sample has two layers of lab-made fabric with a fiber fraction of about 36 wt%. One-component SPCs with uniaxial fibers were made to observe the microstructure of SPCs. Each sample contains 20 layers of 200 fibers aligned along the injection direction in the mold.

2.4. Characterization

Differential scanning calorimetry (DSC) was performed on a TA Instruments Q200 DSC unit. PP granules and fibers were subjected to a heat-cool-heat cycle in the range from 40 to 200 °C at 10 °C/min. The thermal properties of PP SPCs were characterized at a heating rate of 1 °C/min. For comparison, neat PP bar after injection molding and original fibers were also examined at a heating rate of 1 °C/min. The DSC unit was purged with nitrogen at a flow rate of 50 ml/min.

Wide angle X-ray diffraction (WAXD) data were collected on a Rigaku Micro Max 002 (Cu Kα radiation, λ = 0.154 nm) operating at 45 kV and 0.65 mA using an R-axis IV++ detector. Exposure time was 30 min for each sample. The crystalline orientation factor was computed using the method developed by Wilchinsky [22]. The 110 and 040 equatorial diffractions were used to determine the orientation factor based on the monoclinic PP unit cell with dimensions a = 6.65 Å, b = 20.96 Å, c = 6.5 Å, and β = 99°8′. Total integration was used to calculate the crystallinity [23].