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Microcellular polypropylene single-polymer composites prepared by insert-microcellular injection molding

Jian Wang *, Dongjie Chen

School of Chemical Engineering and Environment, Beijing Institute of Technology, Beijing 100081, China

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ABSTRACT

An insert-microcellular injection molding process was performed on an injection molding machine equipped with a supercritical fluid system. The prepared microcellular polypropylene (PP) single-polymer composites (SPCs) combine the advantages of SPCs with benefits of microcellular plastics, they hold the promise for further reduced weight, improved fiber-matrix interface and enhanced recyclability. In comparison with the solid PP, the weight reductions of the tensile and impact microcellular PP SPCs (MPPSPCs) could be up to 12.9% and 3.3% respectively, the tensile and impact strengths of the MPPSPCs were improved by 59% and 1799% respectively. Based on the tensile properties, the injection temperature of 220 °C and injection speed of 70 mm/s were the optimum processing for the tensile MPPSPC samples. The typical morphology structure of the MPPSPC sample includes five different layers: sandwiched fabric layer, transition layer between fabric and core, center core layer, transition layer between skin and center core, skin layer.

1. Introduction

Microcellular foamed plastics represent a group of lightweight materials with heat preservation, acoustic insulation and good processing properties. However, the applications of neat polymer foams are limited by inferior mechanical strengths compared to their solid counterparts due to the significant density reduction and cell structures [1]. Reinforcement materials such as glass fibers, carbon fibers, natural fibers, metal fibers are usually used in polymer composites for enhancing the mechanical properties, accordingly microcellular fiber reinforced polymer composites have been developed to achieve not only reduced weight but also good mechanical properties [2–9]. Especially, the natural fiber reinforced polymer composites or biocomposites have attracted the attention of researchers due to density reduction and rapidly increasing environmental awareness [3–8]. Recently, most research efforts have focused on polymer nanocomposite foams, the combination of functional nanoparticles and supercritical fluid foaming technology has a high potential to generate a new class of materials that are lightweight, high strength and multifunctional, moreover, nanoparticles are suitable for microscaled reinforcement achieving the macroscopic mechanical enhancement [9,10]. The microcellular nanocomposite samples exhibit smaller cell size and uniform cell distribution as well as higher mechanical strength compared to the corresponding base microcellular samples [10–14]. The mechanical properties normalized by weight ratio of the microcellular samples produced under the corresponding optimal processing conditions were increased significantly. However, actually the polymer composite foams have heavier weight compared with the neat polymer foams, and some of them even might have heavier weight than their solid counterparts since the glass/carbon/metal/natural fibers or nanoparticles all have higher density than the polymer. Furthermore, good compatibility of fiber and matrix helped the dispersion of fiber in matrix and improved melt strength, as it was beneficial to stabilize the cellular structure and improve the mechanical properties, whereas poor interfacial adhesion might show a negative influence [15]. Polymer composites usually fail at the weak fiber/matrix interface, resulting from their chemical incompatibility [16]. Interfacial adhesion might be a problem for both the fiber reinforced polymer composite foams and the polymer nanocomposite foams. Moreover, recycling is one of the biggest challenges posed by these above materials. The related recycling techniques must carry out a complete evaluation in terms of environmental impact, efficiency and commercial viability [17]. It is hard to fully recycle or reuse these kinds of composites or composite foams as a result of different compositions between reinforcement and matrix.

* Corresponding author at: 1024 5th Building, 5 Zhongguancun S St, Haidian, Beijing 100081, PR China.
E-mail address: wjj_0107@163.com (J. Wang).

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Single-polymer-composites (SPCs), whose reinforcement and matrix materials are from identical type of polymers, are an emerging class of composite materials with superior interfacial properties, reduced weight and fully recyclability [18]. Therefore, foaming of SPCs has emerged as a novel means to expand the accessible range of foam morphology and produce novel materials without those problems of microcellular polymer composites. Although a great deal of effort has been focused on SPCs, to our knowledge the microcellular SPCs have never been considered and manufactured. There would be many advantages for the microcellular SPCs. Firstly, polymer fiber has the lower density than the fibers based on glass, carbon, flax, wood, etc., and then microcellular processing can produce SPC foams with further density reduction. Besides, strong and stable interfaces could be naturally produced since the two phases are of identical chemistry. Furthermore, the enhanced end-life recyclability can be achieved by using the same polymer for both fiber and matrix phases.

Most of the microcellular foams we mentioned above were produced via microcellular injection molding. Microcellular injection molding has been successfully commercialized by Trexel, Inc. [19,20], thus it can be applied to produce SPC foams. In addition, it is intuitive that the major task when producing SPCs is to widen the temperature range between the matrix and reinforcement with respect to their softening and melting [21]. Hot compaction is still the principle method to produce SPCs, its disadvantage is the long preparation cycle. Therefore, an intriguing challenge for SPCs is to develop processing techniques sufficiently versatile to be scaled up at an industrial level [22]. In our recent publications [23,24], it was shown that the insert injection molding process could realize the production of SPCs with a wide processing temperature window and a short cycle time. Therefore, insert injection molding combined with microcellular injection molding could be conducted to realize the production of microcellular SPCs. On the other hand, the microcellular injection molding can benefit the permeability and improve the interfacial adhesion due to the polymer-gas solution has lower viscosity and the cell growth could create pressure in the cavity.

In this work, the insert-microcellular injection molding was performed to investigate the processing benefits and property improvements of the microcellular SPC foams. PP was used as an example to demonstrate the novel material and its processing. Sample weight and mechanical properties were all compared among the solid PP, PP SPC, microcellular PP, and microcellular PP SPC (MPPSPC) samples. The effects of injection temperature and speed on the mechanical properties were studied. The morphology of the MPPSPC was also characterized.

2. Experimental

2.1. Materials

PP granules (model number: Marlex HGZ-1200, Phillips Sumika Polypropylene Company) with a density of 0.907 g/cm^3 at room temperature were used as the matrix. A plain woven fabric supplied by Innegrity LLC (Simpsonville, SC) was selected as the reinforcement. The woven fabric had an areal density of 742 g/m^2 (4 threads/cm in warp direction and 6 threads/cm in the weft directions) and a thickness of about 0.86 mm. Each bundle consisted of 225 individual fibers. The fiber had a diameter of 48 μm, tensile strength of 560 MPa and tensile modulus of 6.6 GPa measured by a universal tensile test machine (5166 Series, Instron Corp.).

2.2. Composites preparation

The MPPSPC samples were prepared by combining the insert injection molding and microcellular injection molding. The injection molding machine (PT130, L.K. Machinaery Co., Ltd.) was modified and equipped with a commercially available supercritical fluid system (Beijing CHN-TOP Machinary Group Co., Ltd.). Supercritical nitrogen was used as the physical blowing agent. Fig. 1 shows a schematic of the insert-microcellular injection molding and the mold cavities. During the process, the PP woven fabric was firstly set in the mold cavity like an insert and clamping force was used to fix it, then the PP melt with gas droplets was pushed to fill and permeate the fibers from both sides of the fabric.

The operating conditions for the injection molding are shown in Tables 1 and 2. In microcellular injection molding, the pack/hold stage was absent due to the homogeneous packing pressure that resulted from the cell nucleation and growth. In order to investigate the effects of two important parameters, injection temperature and injection speed, seven groups of experiments were performed. For experimental No. 1, 2, 3 and 4, the injection temperature was changed as 200, 220, 240 and 260 °C with a fixed injection speed of 50 mm/s; for experimental No. 5, 2, 6 and 7, the injection speed was changed as 40, 50, 70 and 90 mm/s with a fixed injection temperature of 220 °C.

2.3. Weight measurement

The weight of each sample was measured by an electronic balance (JA3003B) with an accuracy of 0.001 g (made by Shanghai Yueping Scientific Instrument Co., Ltd.). The weight reduction of the microcellular foams was calculated by the following formula

$$\text{Percentage of weight reduction} = (m_{solid} - m_{foam})/m_{solid} \times 100\% \quad (1)$$

where m_{solid} is the weight of the samples without foaming, m_{foam} is the weight of the microcellular samples.

The fiber weight fraction of the PP SPC and MPPSPC samples was calculated using the formula

$$\text{Fiber weight fraction } (\text{wt.}\%) = W_f/m = (\rho_A \times A_s)/(m \times 100) \quad (2)$$

where W_f is the fabric weight (g), ρ_A is the areal density of the woven fabric (g/cm^2), and A_s is the area of the sample shape (mm^2), and m is the sample weight (g). The fiber melting and shrinkage were not considered in the calculation.

2.4. Mechanical tests

The tensile tests were carried out on a universal testing machine (XWW-20Kn, Beijing Jinshengxin Testing Machine Co., Ltd.). The tensile sample was a dumbbell shaped testing specimen with a length of 165 mm, a width of 19 mm and a thickness of 3.2 mm under the guidelines of ASTM D638. The crosshead speed was 5 mm/min with a gauge length of 115 mm. Izod impact tests were conducted using a conventional pendulum-type Izod impact tester (XJUD-22, Chengde Kebiao Testing Instrument Co., Ltd.). The specimens on the face of the notch (based on the Test Method A in the ASTM D256) were rectangle shape with a length of 63.5 mm, a width of 12.7 mm and a thickness of 6.4 mm. Each test was performed at room conditions, and at least five samples were tested for each group.

2.5. Morphological observation

Scanning electron microscopic (SEM) pictures were taken on a microscope (JSM-6301F, JEOL Ltd.) with an accelerating voltage of 15 kV. Observations were performed after sputtering the samples with gold. The failed fracture surface of the tensile MPPSPC samples was firstly examined, then the interface between the fab-