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# Shear induced fiber orientation, fiber breakage and matrix molecular orientation in long glass fiber reinforced polypropylene composites

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ABSTRACT

Long fiber reinforcement is well known to offer thermoplastic materials with high performances. But little work has been done to systematically investigate the effect of shear on the structures and properties of long glass fiber reinforced thermoplastics. The purpose of this work is to investigate the effect of shear on fiber orientation, fiber breakage and matrix molecular orientation in long glass fiber reinforced polypropylene composites, and to construct the structure–property relations. A so-called dynamic packing injection molding (DPIM) technique, which imposed oscillatory shear ($10\,s^{-1}$) on the gradually cooled melt during the packing solidification stage, was used to prepare the dynamic samples, and optical microscope (OM), scanning electron microscope (SEM), micro-Fourier transform infrared spectroscopy (FTIR) and differential scanning calorimetry (DSC) were used to characterize the samples. The results show that despite the well-published enhanced fiber and matrix orientation, shear will cause a remarkable fiber breakage. Combined effect of fiber breakage and orientation on the mechanical properties of the composites was qualitatively investigated and tensile properties of the dynamic samples, the conventional injection molded samples and the short glass fiber reinforced samples were compared. It is demonstrated that shear will induce more severe fiber breakage in long glass fiber reinforced polypropylene than in short glass fiber reinforced polypropylene, and that compromising the decreased strength caused by the severe fiber breakage is very difficult.



## 1. Introduction

In recent years, thermoplastics/fiber composites have been intensively investigated in both industrial and academic fields, due to the advantages of these systems such as ease of processing, enhanced mechanical and thermal properties and an unlimited shelf life. Within polymer/fiber composites, glass fiber is the most preferred reinforcement material despite its limitations. This is mainly because of its availability in various forms, its capability to be economically processed by different manufacturing routes, the production of high specific strengths, and its use in a corrosive environment. Glass fiber reinforced thermoplastics can be divided into the following four categories based on the fiber aspect ratio [1].

- Short fiber reinforced thermoplastics for injection molding, delivering an average fiber length of <1 mm in the molded part.
- Long fiber reinforced thermoplastics for injection and extrusion compression molding, delivering an average fiber length in the 1–25 mm range in the molded part.
- Random, in-plane, fiber-reinforced thermoplastics known as Glass Mat Thermoplastic (GMT) for compression-flow molding, with fiber lengths in the 10–50 mm range.
- Continuous fiber reinforced products based on pre–preg or other technologies for compression molding.

Moving down this list generally means moving to systems which can offer higher performance but usually involves more complex, time-consuming manufacturing processes. So long (but discontinuous) fiber reinforced thermoplastics possessing both high performance and mass processibility has recently received much attention. A variety of techniques [2–8] producing long fiber reinforced thermoplastics such as wire coating, crosshead extrusion, impregnation and thermoplastic pultrusion have been developed and several articles [1,9–20] investigating the effect of fiber length on the mechanical properties of composites are also available. The results from Thomason's work [1,9–11,20] and Fu's work [14–17] show that final fiber length plays an important role in mechanical properties of the composites, and that long fiber reinforced PP delivers significantly longer fibers to the molded composite in comparison to the extruded compounds. Many factors influence the final fiber length in the molded composites such as the fiber instinct [17], flow stress during processing [12,21], mold

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geometry [22], processing conditions [21,23,24] and fiber content [12,17,19,21,24,25].

Besides the fiber length or aspect ratio, other variables such as fiber content, base resin properties, fiber strength, fiber dispersion, fiber orientation and interfacial adhesion are also of prime importance to the final balance of properties exhibited by molded thermoplastic composites [1,9–17]. A number of studies are concerned with fiber orientation and interfacial adhesion in the composites [12,22,26–40]. It is well known that short fiber reinforced thermoplastics produced by conventional injection molding show complex fiber orientation distributions, which vary through the sample thickness and with different locations in the molding [8,20,22,26]. The resulting fiber orientation can be manipulated by the material flow type during the molding filling, the mold geometry and the processing conditions [12,22,29–32]. Several methods have been reported to better orient the glass fibers in the injection molded parts, such as the open-ended injection molding [27,28], the sandwich injection molding/co-injection molding [29], injection into a rotation mold [30] and dynamic packing injection molding [32]. Most of the results show enhanced mechanical properties when more glass fibers are aligned along the flow direction because of the shear stress [22,29,30,32]. But it is worth noting that all the fibers used in the above-mentioned work on fiber orientation are short fibers processed from extrusion molding, and few researches have focused on the effect of shear on microstructures and properties of long fiber reinforced thermoplastics to the best of our knowledge.

As to the interfacial adhesion, developing methods to control it has been the subject of considerable research efforts, because good adhesion results in efficient stress transfer from the continuous polymer matrix to the dispersed fiber and thus increases the ability of the material to absorb energy [33–35]. Several methods were used to enhance the adhesion, such as glass fiber sizing, fiber surface grafting and adding binding agent [33,34,36–38]. The effect of matrix molecular weight on interfacial properties was also investigated [35].

In our previous work [32,39,40], using the unique dynamic packing injection molding (DPIM) technique which imposed oscillatory shear on the gradually cooled melt during the packing solidification stage, effect of shear on the properties of short fiber reinforced polypropylene was investigated. We demonstrated that shear may enhance not only the fiber orientation but also the interfacial adhesion [32]. Meanwhile, a transcrystalline layer was observed to form on the interface [39]. In this work, long glass fiber reinforced polypropylene (LFPP) prepared with an impregnation device, which possesses higher mechanical properties, was used instead of short glass fibers reinforced polypropylene (SFPP). The tensile properties of the injection molded composites were investigated taking into consideration of the effect of shear on the fiber microstructures (dispersion, length and orientation) and the matrix supermolecular structures. For long glass fiber reinforced polypropylene (LFPP) composites, shear will cause not only a better orientation of fiber, but also a breakage of fiber. Thus shear induced fiber orientation, fiber breakage and matrix molecular orientation in LFPP will be determined and the different effect of shear on tensile properties of LFPP and SFPP will be explained.

## 2. Experimental

### 2.1. Materials

A commercially available isotactic polypropylene (iPP), purchased from Formosa Plastics Incorporation, with the melt flow index (MFI) of 15 g/10 min (230 °C, 2.16 kg), was adopted as basal resin. PP-g-MA (MA content = 0.9 wt%, MFI = 6.74 g/10 min) in which anhydride group is randomly grafted on a PP backbone was supplied by Chenguang Co. (Sichuan, China) and used as compatibilizer. The mass ratio of the basal resin and the compatibilizer was 98/2. Glass fiber roving was supplied by Jushi Group (Chengdu, China) and used as reinforcement. The fiber was treated with a silane coupling agent by the supplier before purchase.

**Table 1**
Processing parameters in dynamic packing injection molding.

| Parameters | Values |
|---|---|
| Injection pressure | 90 MPa |
| Injection speed | 80 $cm^3$/s |
| Oscillating packing pressure | 4 MPa |
| Oscillating frequency | 1.0 Hz |
| Holding time | ~4 min |
| Melt temperature | 205 °C |
| Hold temperature | Room temperature (about 25 °C) |

### 2.2. Sample preparation

#### 2.2.1. Impregnation

A self-designed extrusion–impregnation device, in which a co-rotating twin screw extruder (SHJ-35, Nanjing Haote Machinery Company) was connected with an impregnation device by a joint, was used for producing the LFPP continues strand. The schematic representation of this equipment was similar to that described by Yoon et al. [41]. The temperature of the extruder was maintained at 170, 195, 220 and 220 °C from hopper to die and the screw speed was about 90 rpm. The temperature of the impregnation groove was set at 225 °C.

The continuous strand was then chopped into pellets at a length of 10 mm for injection molding. Obviously, the length of glass fiber in the pellet is equal to the pellet size.

#### 2.2.2. Injection molding

After the extrusion–impregnation step, proper amount of polypropylene was added into the feedstock before injection molding to keep the glass fiber at the content of 0%, 3%, 6%, 10%, 20% and 30%, respectively. Then the PP/GF composites were molded through dynamic packing injection molding (DPIM), in which oscillatory shear (10 $s^{-1}$) was imposed on the gradually cooled melt during the packing solidification stage. The detailed experiment procedure has been described in Ref. [39,42]. Injection molding under static packing was also carried by using the same processing parameters but without shear for comparison. The samples obtained by dynamic packing injection molding are called dynamic samples, while the samples obtained by static packing injection molding are called static samples. The processing parameters are listed in Table 1.

### 2.3. Characterization and testing

#### 2.3.1. Determination of average fiber length

The injection molded bars of the composites were burned in a muffle furnace for a period of 2 h at 600 °C to isolate the fibers from the composites. Then the extracted fibers were dispersed on a glass slide for optical microscope observation. Length of at lease 500 fibers was recorded with a digital camera equipped on the microscope and measured with Image J software system. Number average fiber length and weight average fiber length were calculated using Eqs. (1) and (2), respectively:

$$l_n = \frac{\sum n_i l_i}{\sum n_i} \tag{1}$$

$$l_w = \frac{\sum n_i l_i^2}{\sum n_i l_i} \tag{2}$$