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Material characterisation

Influence of fiber orientation and length distribution on the rheological characterization of glass-fiber-filled polypropylene

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ABSTRACT

We report on the characterization of glass-fiber-filled polypropylene compounds identified by means of rotational rheometry in parallel plate mode. The small gap between the plates and the length of the glass fibers cause considerable wall and initial orientation effects, which we minimized by applying various degrees of pre-shearing, optimizing both duration and level of shear deformation. The results were verified using X-ray computed tomography to investigate the dispersion of glass fibers in the melt. In addition, extrusion experiments confirm the validity of our results.

1. Introduction

Thermoplastic materials such as polypropylene random copolymers are often filled with glass fibers to improve properties such as stiffness, strength, and heat distortion temperature compared to their unfilled counterparts. As they are processed in the molten state, their rheology is of technical relevance. In contrast to geometrically isotropic fillers, maximum reinforcement is obtained only when the fibers are properly oriented. During processing, the fibers can adopt complex patterns of orientation which are retained in the final component. Crowson et al. [1] reported observations of the fiber orientation under conditions of converging, diverging, and shearing flows, which they investigated using contact microradiography (CMR).

However, because large aspect ratio fibers are dispersed in a polymer matrix, the rheological properties are complex. It is well known that a thermoplastic polymer filled with glass fibers with initially isotropic orientation exhibits, in the molten state, a viscosity overshoot when sheared in the parallel plate geometry of a rheometer. Sepehr et al. [2] developed a model based on the Folgar-Tucker equation for fiber motion and the Lipscomb constitutive equation to simulate viscosity and normal stress overshoots.

Generally, glass fibers used for reinforcing thermoplastic materials can be classified into short and long fiber types. Short fibers have a diameter of about 10 μm and are generally less than 1 mm long, while long discontinuous fibers are typically more than several millimeters long. Reinforcement with these long fibers is more effective, but introduces new problems such as fiber breakage during compounding. A number of studies [3,4] have shown that average fiber length decreases in processing.

Most of the results reported by other researchers were obtained using a constant flow rate capillary rheometer. Crowson and Folkes [5] investigated the effect of fiber concentration, fiber length, and temperature on shear

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viscosity and die swell of several short glass-fiber-filled thermoplastics. Both fiber length and volume fraction hardly affect viscosity at high shear rates, but were found to have a considerable effect at low shear rates, which was attributed to fiber clustering.

Similar investigations were reported by Laun [6], who compared the results to experiments with unfilled and glass-bead-filled samples. In addition to the aforementioned capillary rheometer, Laun used a Weissenberg rheogoniometer in combination with X-ray microradiography to investigate the effect of fiber orientation in simple shear flow. When compression-molded samples were replaced with samples prepared by injection molding, differences were observed that were attributed to changes in initial fiber orientation during measurement.

Thomasset et al. [7] also chose injection-molded samples in their study of long-glass-fiber-filled polypropylene when they developed a model of shear viscosity that describes solid-like to fluid-like behavior at low to high shear stresses, respectively, and takes fiber content and orientation into account.

Analysis of contraction flows in capillary rheometers provides a well-established technique for estimating the elongational viscosity of polymer melts. Binding [8] confirmed in his studies that the filler causes a relatively small increase in shear viscosity (of the order of twice the viscosity of the base polymer) but a 1000-fold increase in extensional viscosity.

The general consensus appears to be that, for a material filled with a high-volume fraction of large-aspect-ratio fibers, entrance pressure increases markedly with increasing volume fraction. Due to the strong correlation between entrance pressure and extensional deformation of a polymer melt, elongational viscosity increases, which was shown in uniaxial extension by Kamal et al. [9].

Mutel and Kamal [10] employed parallel plate rheometry to study the steady shear viscosity and normal stress behavior of glass-fiber-reinforced polypropylene melts. For fiber loadings above 10 percent, both viscosity and normal stress difference at a given shear rate increase with increasing fiber loading. The effect of fiber loading diminishes at higher shear rates, which is in agreement with the findings of other researchers.

However, if the distance between two plates (1 mm) and fiber dimensions (diameter 10 μm and maximum length around 2.5 mm) are considered, it is clear that wall and initial orientation effects will be significant. The aim of this study was to investigate the steady and dynamic rheological properties of glass fiber-filled polypropylene melt using a rotational rheometer in parallel plate mode. In contrast to previous contributions, we varied the duration of different levels of pre-shearing in order to achieve optimal orientation of the fibers parallel to the plates.

Shear measurements using a slit die in combination with a laboratory scale extruder equipped with a melt pump were in good accordance with the rotational rheometer results. In addition, we investigated fiber length distribution and fiber orientation using X-ray computer tomography (XCT). Elongational measurements using a converging slit provide a sensitive means of determining fiber length.

2. Experimental

2.1. Materials

A Borealis RA130E polypropylene random copolymer intended for plumbing and heating applications was used as a base polymer, and two types of glass fiber were investigated. The fiber dimensions as stated by the manufacturer are summarized in Table 1. The glass fiber compounds were prepared by means of a Coperion ZSK 70 industrial twin-screw extruder with a length of 42 times the diameter and three venting units (two atmospheric and one under vacuum) in combination with underwater pelletizing.

Three types of compounds were prepared and investigated in this study: compounds with (i) 100% short fibers, (ii) 20% long and 80% short fibers, and (iii) 70% long and 30% short fibers. The weight proportion of glass fibers was 24% in each compound.

Each compound was available in the form of cylindrical pellets with a diameter of 2.5 mm and a length of about 4 mm. The material was compression-molded at 180 °C into disc-shaped specimens with a diameter of 25 mm and a thickness of about 1.5 mm at 180 °C. Molding time was approximately half an hour.

2.2. High-temperature gel permeation chromatography (HT-GPC)

Weight-average and number-average molecular weights (M_w and M_n) and polydispersity index (PDI) were determined by gel permeation chromatography with 1,2,4-trichlorobenzene (Acros Organics, >99%) as solvent and 250 mg/L 2,6-di-tert-butyl-4-methylphenol (Sigma Aldrich) as stabilizer using the following configuration: flow rate 1 mL/min, mixed bed separation columns GMHXL-HT from Tosoh, and detector for refractive index (RI), right-angle light scattering (RALS) and low-angle light scattering (LALS) combined with a differential viscometer. Polystyrene 99k standards from Viscotek were used for calibration.

The PP-GF samples were filtered using extraction thimbles, and the soluble fraction was analyzed. Two random samples were taken, and reproducibility was within the usual range, although a margin of error of about 5% must be taken into account.

2.3. X-ray computed tomography (X-CT)

A Phoenix X-ray Nanotom 180NF CT device was used for quantitative sub-μm analysis of fiber length and diameter distributions and for determining the orientation in three spatial dimensions. Data quality has to be superior for length and diameter determination and can be slightly worse for determination of orientation. Better resolution leads to better data quality but also to smaller analysis

Table 1
Types of glass fiber used in the study.

Type of glass fiber	Coding	Diameter	Max. length
OTTE OK-2301 KF	Short fibers	10–19 μm	1.5 mm
ChopVantage® HP 3299	Long fibers	14 μm	4.5 mm