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Process-induced fiber matrix separation in long fiber-reinforced thermoplastics

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ABSTRACT

This work explores the fiber migration during injection molding of long glass fiber-reinforced polypropylene. It was found that the simplified assumption of uniform fiber concentration distribution is inaccurate, and the process causes substantial variations in the fiber concentration along the flow path and through the thickness of injection molded parts. This was tested for a simple plate geometry molded at varying nominal fiber concentrations. The fiber concentration was measured by pyrolysis to obtain a global concentration, and using micro computed-tomography for a through-thickness analysis. Additionally, the fiber concentration at the melt front of partially filled moldings was investigated. A new measurement protocol using micro computed-tomography and digital image processing is proposed to calculate the through-thickness fiber concentration. The results of this study show substantial heterogeneity of the fiber concentration throughout the molded plates. Fibers agglomerated in the core layer with volume fractions up to 1.5 times the nominal fiber concentration.

1. Introduction

Injection molding using long fiber-reinforced thermoplastics (LFTs) is a widely-used process to manufacture parts with enhanced mechanical properties. Particularly in the automotive industry, LFTs have gained importance due to their exceptional lightweight properties and cost-efficient manufacturing processes. With favorable specific stiffness and strength, LFT materials can potentially replace metals for structural applications and play a key role in reducing the overall weight of automobiles. However, the local properties and global performance of the molded part greatly depend on the final state of the fibers [1,2]. During mold filling, the configuration of the fibers changes significantly, reflected in mechanisms referred to as fiber attrition, fiber orientation, fiber jamming and fiber matrix separation [3–5]. In particular, the phenomenon of fiber matrix separation, which describes the process-induced variation of fiber concentration, has not been fully understood. Fiber migration during processing occurs on two scales - variation in fiber *local concentration through the thickness* of a molded part and *global concentration gradient* along the flow path.

Previous studies have addressed the process-induced concentration gradients throughout molded parts for filled thermoplastics, including short fiber (aspect ratio <100) and long fiber (aspect ratio >100) reinforced composites. Toll and Andersson [6] published results showing an increase in fiber concentration from nominal 30 wt.% in the raw material to 36 wt.% at the end of the flow path of a film-gated plate for glass fiber-reinforced polyamide 66 (PA66). In a similar study, O'Regan et al. [7] observed an increase of 3–4 vol.% at the end of the flow path in injection molding of fiber-reinforced PA66. Lafrance et al. [8] showed an increase from 39 wt.% at the gate to 51 wt.% at the end of the flow path for a fan-gated plate. Using a spiral mold, Kubat and Szalanczio [9] studied the filler migration effects of a glass sphere-filled low-density polyethylene. Along a total length of up to 1950 mm, they measured the filler concentration and found substantial migration of the particles towards the end of the flow path. Their results suggest a relative increase of up to 25% filler concentration at the tip of the spiral for glass spheres with diameters between 53 and 105 μm.

Hegler and Menning [10] published work on the filler separation effects during injection molding of glass bead and glass fiber-filled thermoplastics using dumbbell and rectangular box specimens. Their results show that the separation effects were more pronounced with beads than with fibers indicating an influence of the shape and dimensions of the fillers. Furthermore, they conclude that mold geometry and filler concentration are key parameters determining the degree of filler migration in injection molding. Other processing parameters, such as mold temperature, injection speed, matrix material and screw speed did not

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substantially impact the filler-matrix separation phenomena in their experiments.

Ogadhoh and Papathanasiou [11] studied particle migration during mold filling of glass bead-filled polystyrene with an average bead size varying from 50 to 500 μm. Their results agree with previous studies, showing the filler matrix separation increases substantially with larger beads with the largest relative increase of 15% more fillers for 500 μm glass beads at the end of the flow. Another major outcome of their work was the observation that particles appear to accumulate at the free surface. For the largest particle sizes used in the study, the concentration of particles near the free surface was almost double than in the feed material. Their results on filler migration strongly indicate a dependency of particle size and nominal concentration.

Mondy et al. [12] conducted fiber migration experiments in a wide-gap Couette flow for low aspect ratio nylon fibers in glycol. Their measurements show a migration of fibers to the low-shear regions. While the authors did not find an impact of aspect ratio on the migration, they found an influence of fiber concentration suggesting that the effect of shear-migration increases at elevated concentrations.

The common conclusion of all studies is the outcome that the fiber concentration increases towards the end of the flow path, suggesting that the last filled location carries an accumulated amount of filler. Furthermore, the processing conditions appear to play a smaller role for the degree of fiber matrix separation. The influencing factors are reported to be both the concentration and the shape of the filler since the migration effect is more pronounced for longer fibers than it is for shorter fibers [6].

The through-thickness filler concentration gradient in injection molding has been addressed much less frequently. A first comprehensive study was conducted by Toll and Andersson [6] for 30 wt.% glass fiber-reinforced PA66 and simple plate geometry. They found an agglomeration of fibers in the core layer using a sectioning and microscopic measurement protocol. Their results suggest almost 40% more fibers in the core layer than in the shell for their long fiber PA66 (initial fiber length of 10 mm). For the short fiber grade (initial fiber length of 0.6 mm), the fiber agglomeration is much less pronounced with only 10% more fibers in core than in the shell region.

While other publications also mention the phenomenon of fiber agglomerating in the core of injection molded part, the observations are mostly side-effects in these studies; no comprehensive analysis exists nor have theories been formulated to explain the underlying mechanism. Velez-Garcia et al. [13] focused on fiber orientation measurements for short fiber-reinforced polybutylene terephthalate (PBT) by applying a newly developed sectioning procedure. From the obtained micrographs, they also reported a fiber concentration gradient through the thickness of the molded center-gated disk. Recently, Sun et al. [14] published an experimental study on using micro computed-tomography (μCT) to quantify the fiber orientation of an LFT injection molded instrument panel. While also focusing on orientation measurements, they incidentally analyzed the pixel fraction from μCT scans indicating a strong agglomeration of fibers in the core layer of the molded part.

Theoretical models to describe particle migration in concentrated suspensions have been proposed. Leighton and Acrivos [15] suggested a diffusive flux model, which describes the shear-induced particle migration as a result of irreversible particle collisions. This kinematic modeling approach expresses the migration as gradients of particle concentration and shear rate, but it neglects normal stress differences.

Nott and Brady [16] proposed a suspension balance model. This model is a two-phase approach describing the particle phase and suspension separately while incorporating a constitutive model to solve particle and suspension stresses. The particle transport is driven by gradients in these stresses. Morris and Boulay [17] expanded the suspension balance model to account for anisotropic migration. The latter model was recently implemented in a commercially injection molding simulation software (Moldex3D™, CoreTech Systems, Taiwan). Using this three-dimensional (3D) finite volume method (FVM) solver, Tseng et al. [18,19] simulated the fiber migration for a short fiber-reinforced PBT. Their results suggest that the model can qualitatively predict the tendency of the process-induced fiber concentration in the molded part. However, there are still quantitative discrepancies in the through-thickness prediction of the fiber concentration. Also, their work does not address the relevance of the suspension models fitting parameters nor how to obtain appropriate values for these parameters for fiber-reinforced thermoplastics.

Fiber matrix separation can also be caused by the part design and the features of the part. The separation effect in rib-filling has been studied by a few research groups, who describe the phenomenon of fibers accumulating in the base of the rib while the tip of the rib remains almost fiber-free [5]. While this is an important field of research, the impact of design features on fiber matrix separation is not part of this work and the reader is referred to a recent publication on rib-filling [20].

A major challenge has been, and remains to be, the availability of reliable measurement techniques that allow a comprehensive analysis of fiber concentration for sufficiently large samples in a timely manner. The full characterization of the 3D fiber microstructure for fiber-reinforced composites has not been standardized yet and differences in characterization protocols question the comparability of experimental studies. However, reliable experimental data is necessary to understand the underlying physics of fiber matrix separation, to develop new models to predict the process-induced fiber matrix separation in LFT injection molding and to validate predictive tools.

This work presents a comprehensive experimental study on the fiber matrix separation in LFT injection molding for a simple plate geometry at varying nominal fiber concentration. A measurement protocol using μCT and image processing is introduced for the characterization of the through-thickness fiber concentration. The analysis of the fiber concentration at the flow front was performed to obtain experimental data of the reorientation of the fibers in the fountain flow region and the corresponding impact on the final fiber concentration distribution.

2. Materials and methods

The material used in this study is a commercially available long glass fiber-reinforced polypropylene (PPGF) (SABIC® STAMAX™). Table 1 summarizes the main material properties.

The design-of-experiments (DoE) consists of nominal fiber concentrations varying from 5 wt.% to 60 wt.% Table 2 summarizes the trial label, the corresponding fiber volume and fiber weight concentration as well as the raw material used to achieve the respective nominal concentrations. PPGF20, PPGF30, PPGF40 and PPGF60 are provided as compounded pellets by the material supplier (coated long fiber pellets). PPGF05, PPGF10 and PPGF50 were achieved by mixing higher fiber concentrations with neat PP

Table 1
SABIC® STAMAX™ LFT material properties according to the material supplier [21].

Material property	Value
Nominal fiber length [mm]	15.0
Fiber diameter [μm]	19 ± 1
Density of fibers [g/cm^3]	2.55
Density of PP [g/cm^3]	0.905