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Shear viscosity measurement of highly filled polycarbonate melts using a slit-die rheometer

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Viscosity measurement and the flow behavior prediction of reinforced polymer melts have been considered crucial for quality consistency in commercial products where highly filled polymer composites are used. In this study, a slit-die rheometer mounted on an injection molding machine was employed to measure the shear viscosity of a reinforced polymer melt by monitoring the pressure drop in the slit and the ram movement for various melt temperatures and shear rates. Mold filling experiment was also conducted using an injection molding process with two plaque-shaped parts of different thicknesses. Polycarbonate with 40 weight % of glass fibers was investigated in the experiment. A comparative numerical simulation was conducted as well using the measured viscosity in the current study. The pressure drops from the simulation and the experiments were compared, which reveals that the viscosity measured using a slit-die rheometer is relatively accurate.

***Keywords*:** slit-die rheometer, shear viscosity, glass fiber reinforced polycarbonate, flow analysis, cavity pressure

1. Introduction

Most of today's multifunctional mobile products such as laptop computers, cellular phones, personal media players, *etc*., have evolved considerably small-sized and light-weighted over the past few years, with consequent reduction in part thicknesses. In order to compensate for the reduced thicknesses, polymers with high portions of reinforcement fillers are used in these products. Therefore predicting melt flow behaviors and the measurement of the viscosity are of great importance to ensure the production of thin and compact injection-molded products with consistent quality. Although there have been a lot of researches on the measurement of the viscosity in injection molding (mainly focused on neat polymers), one needs a robust viscosity measurement technique particularly well suited for highly filled polymer melts in injection molding applications.

Traditionally, the shear viscosity of an unreinforced polymer melt is measured by capillary rheometers and slit-die rheometers (Laun, 1983; Langelaan *et al*., 1994) for the injection-molding application. Commercial capillary rheometers allow the measurement of shear viscosity of unreinforced polymer melts at high shear rate regions, but the process is time-consuming. Moreover the Bagley correction is necessary to account for the entrance effect caused by the contraction of the cross-sectional area from the reservoir into the capillary die (Bagley, 1957). Also, to measure the viscosity of a short- or long-fiber reinforced polymer, the fillers may not come out easily through the capillary die with the polymer melt, due to their large volume fractions or long lengths (as in long fiber thermoplastics (LFT)), which makes it hard to measure the shear viscosity accurately. Moreover, unlike actual injection molding processes where fillers are often broken by a screw, fiber fillers in capillary rheometers largely remain unbroken and the resulting fluidity or the viscosity may become significantly different from actual processing conditions (Thomasset *et al*., 2005). Amano *et al*. (2000) employed a capillary-type rheometer which was mounted on an injection molding machine to measure the viscosity of a reinforced polymer. However, it has turned out not straightforward to get the viscosity accurately, unless the pressures at the die entrance and the exit were measured accurately and the Bagley correction is performed correctly, which is particularly difficult to be done accurately for multi-phase materials with fillers such as glass fibers with non-linear Bagley plots (Han, 1981).

On the other hand, slit-die rheometers do not require the Bagley correction (Macosko, 1994). They are mostly used in connection with conventional extruders in measuring the shear viscosity in low shear rate regions (Han, 1974). In this study, a slit-die rheometer mounted on an injection molding machine is employed to measure the shear-dependent viscosity of highly filled short-fiber reinforced polycarbonate. In order to verify the accuracy, the measured pressure data were compared with the prediction from the flow analyses using the measured viscosity. More specifically, the wall shear stresses of the reinforced polymer melt were calculated from the pres-

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Table 1. Thermal and mechanical properties of the test material

Property	Symbol	Value
Melt density	ρ	1347.0 kg/m^3
Thermal conductivity	k	0.312 W/m·^{o}C
Heat capacity	C_p	1825.0 J/kg.^{o}C

sure drop obtained by the pressure transducers in the slit-die; the shear rates were estimated by the flow rates; and the injection speed information was used to obtain shear-rate dependence of the viscosity. For the same materials, the measured viscosity from the slit die was compared with that from the capillary-type rheometer mounted on the injection molding machine. In addition, further validation of the measured viscosity from the slit die rheometer was made by comparing the experimental cavity pressure data during the injection molding process and the numerical cavity pressure distribution from the flow analysis.

2. Experiments

2.1. Materials

The polymer used in this study (LUPOY GN2403F, LG Chemical) is a commercial-grade polycarbonate (PC) with 40 weight % of glass fiber. This is frequently used in laptop computer housings because of its excellent flexural properties. The thermal and mechanical properties of the material are listed in Table 1. The thermal conductivity (k), melt density (ρ) and heat capacity (C_p) of the material were assumed constant within the range of temperatures in the slit-die.

2.2. Experimental setup

In order to measure the shear viscosity of the reinforced polymer, a slit-die system was mounted on an injection molding machine (Battenfeld 75 ton, screw diameter of 35 mm) as shown in Fig. 1. The slit-die system, directly connected to the barrel of the injection molding machine, was designed as an independent structure to minimize heat loss. The slit-die system consists of five sections: (i) a cylindrical slit-die divided into two halves (upper and lower parts), (ii) an adaptor to the injection molding machine, (iii) a barrel, (iv) band heaters surrounding the slit-die and (v) a temperature controller for the heaters.

The total length of the slit-die was 200 mm, as shown in Fig. 2. The length of the transition region between the cylindrical barrel and the flat slit is 40 mm. The rectangular slit is 2 mm in thickness and 20 mm in width. Five Dynisco transducers (TPT4636-1MK-12/30) for temperature and pressure measurements were installed along the slit path, each of them 30 mm apart. The sensors can measure the pressure from 30 to 100 MPa and the temperature up to 350^{o}C. The maximum sampling frequency of the sensor was 1000 Hz. The position of the ram (or the screw) with time is obtained directly from injection molding machine. The temperature, the pressure and the ram position are displayed on the computer monitor through the data acquisition system (DAQ) with Dataflow from Kistler. Flow rates were measured by

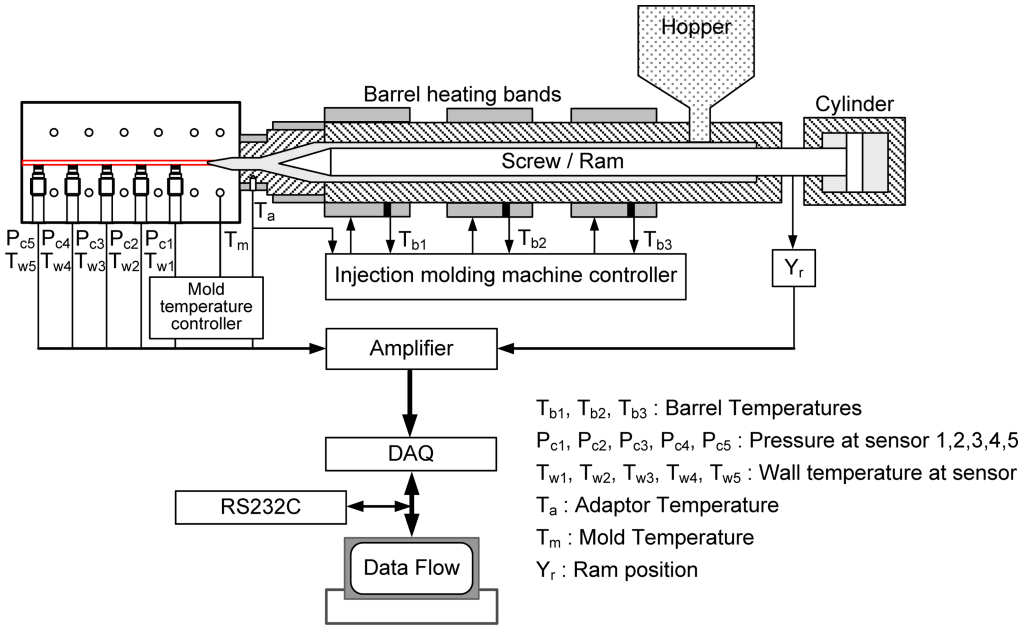

Fig. 1. (Color online) Schematic diagram of the slit-die system.

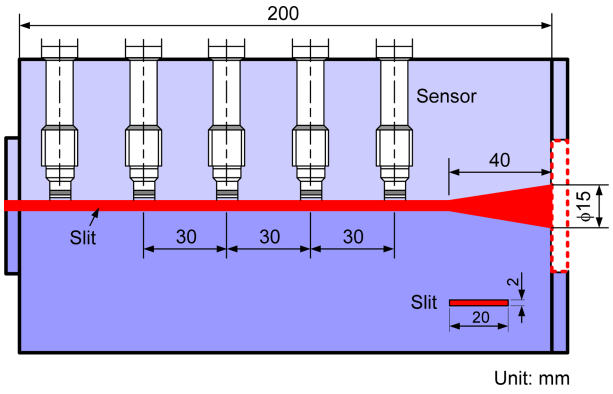

Fig. 2. (Color online) Slit-die geometry.

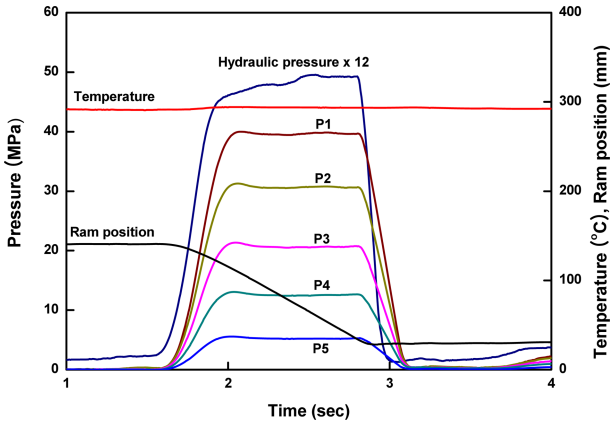

Fig. 3. (Color online) Change of pressure, temperature and ram position with time (290°C, 103 mm/s).

monitoring the ram speed during the injection molding cycle.

2.3. Viscosity calculation

The procedure for the viscosity calculation is described as follows: (i) calculation of the shear stress at the wall of the silt-die using the pressure value obtained from the sensors; (ii) calculation of the apparent shear rate from the measured flow rate in the barrel of the injection molding machine, (iii) analysis of the linearity of pressure profiles obtained at different sensor locations, (iv) estimation of the true viscosity through the Weissenberg-Rabinowitsch correction with the wall shear stress and the apparent shear rate calculated above, and (v) regression analysis for the true viscosity with the Cross-WLF equation (Cross, 1979; Williams *et al.*, 1955). The procedure for calculating the viscosity does not require a Bagley correction for the entrance effect.

The wall shear stress in a slit die with thickness of H and width W is calculated from the following equation (Walters, 1975):

$$\tau = -\frac{H}{2(1+W)}\frac{\Delta p}{L} \tag{1}$$

where Δp denotes the pressure drop along the distance of L.

The apparent shear rate is calculated by

$$\dot{\gamma}_a = \frac{6Q}{WH^2} \tag{2}$$

The true shear rate can be obtained from the apparent shear rate using the Weissenberg-Rabinowitsch correction (Rabinowitsch, 1929):

$$\dot{\gamma} = \frac{2n''+1}{3n''}\cdot\dot{\gamma}_a \tag{3}$$

where

$$n'' = \frac{d(\log\tau)}{d(\log\dot{\gamma}_a)} \tag{4}$$

There are several viscosity models to represent the fluid behaviors of polymer melts in simulation of injection molding process. In this study, in order to fit the measured viscosity results, the Cross-WLF model was chosen, which is widely used in many commercial softwares. The Cross-WLF model has the form:

$$\eta = \frac{\eta_0}{1+(\eta_0\dot{\gamma}/\tau^*)^{1-n}} \tag{5}$$

where $\dot{\gamma}$ is the shear rate, n the power-law index, τ^* the shear stress at the transition between Newtonian and power law behaviors, and η_0 the zero shear viscosity. The zero shear viscosity η_0 encompasses the temperature dependent viscosity behavior with the WLF equation as follows:

$$\eta_0(T,p) = D_1\exp\left\{-\frac{A_1(T-T^*)}{A_2+(T-T^*)}\right\} \tag{6}$$

where T^* might be taken as the pressure-dependent glass-transition temperature.

$$T^* = D_2 + D_3 p \tag{7}$$

$$A_2 = \tilde{A}_2 + D_3 p \tag{8}$$

3. Results and Discussions

3.1. Linearity of pressure profiles in slit-die

In order to find the shear viscosity of the reinforced polycarbonate, the pressure inside the slit-die was measured at the melt temperatures of 290°C, 305°C and 320°C and for various injection speeds. Fig. 3 shows the measured pressure, the hydraulic pressure, the temperature and the ram position with time at the injection speed of 103 mm/s. A slight overshoot in the pressure immediately after the start of injection and subsequent pulsation can be observed. This becomes more evident as the injection speed increases. Fig. 4 shows the measured pressure values at

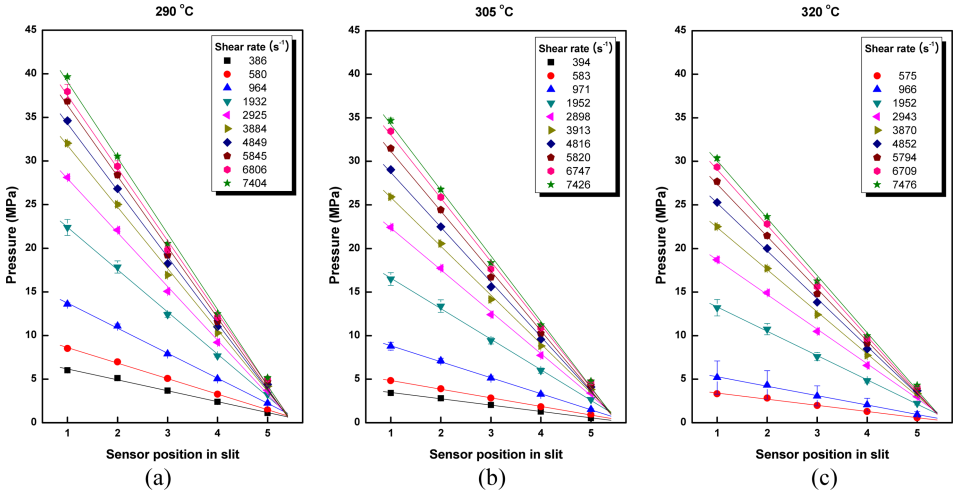

Fig. 4. (Color online) Measured pressure values at several senor locations at various shear rates and at the melt temperatures of (a) 290°C, (b) 305°C and (c) 320°C.

several sensor locations at different melt temperatures and shear rates. Dots represent the measured pressures and the solid lines represent the fitting. For a given temperature, the slope of the pressure versus distance along the slit increases with the shear rate which indicates increase in the wall shear stress. At the same time, one can observe that, for a given shear rate, the slope of the pressure versus distance decreases with increasing the melt temperature, indicating the reduction in the viscosity.

The pressure changes linearly with the distance in many cases and this may imply that the effect of viscous heating on the viscosity is not so significant inside the slit die. That is, the viscosity reduction at high shear rates appears to be small in this typical setup. And also, it seems that the combined effects of viscous heating and pressure on the viscosity approximately cancel out and the linearity appears. These are two competing effects (Ansari *et al.*, 2012; Syrjälä and Aho, 2012). One can observe slightly concave data (dots) in Fig. 4 (a-c), in comparison with the solid lines from the linear fit, particularly above the shear rate 3000 s^{-1}. The difference between the measured and fitted data has turned out more evident as the shear rate increases. This discrepancy is mainly due to the viscosity reduction with viscous heating which scales with the square of the shear rate (Mitsoulis *et al.*, 1998; 2003), which cannot be avoided in a long slit die. Furthermore, the polymer melt, having experienced a change in its morphology while passing through the transient area of the slit die, shows the steady and fully developed flow from the first sensor, from which the performance of the proposed design of the slit-die rheometer can be partly assessed.

3.2. Calculation and validation of the shear viscosity of the reinforced polymer

In this study, the wall shear stress is obtained from the measured pressure values at the pressure sensors (Eq. 1). The flow rate is estimated by the injection speed which is obtained from the ram position history of an injection molding machine. From these data, one can obtain the true shear rate and the apparent shear rate (Eqs. 2-4). The apparent shear rate in this experiment ranges from 300 s^{-1} to 8,000 s^{-1}. Fig. 5 shows the apparent viscosity obtained at three different temperatures. Solid lines represent the curve fitting with the Cross-WLF equation. Fig. 6 shows the true viscosity with the Weissenberg-Rabinowitsch correction. The viscosity of the same material obtained from Autodesk Moldflow Plastics Labs, which is the data with the Weissenberg-Rabinowitsch correction and the Bagley correction, is presented as well for comparison (Autodesk testing report, 2011). Autodesk Moldflow Plastics Labs uses two capillary-type dies mounted on an injection molding machine (Arburg Allrounder 270S, screw diam-

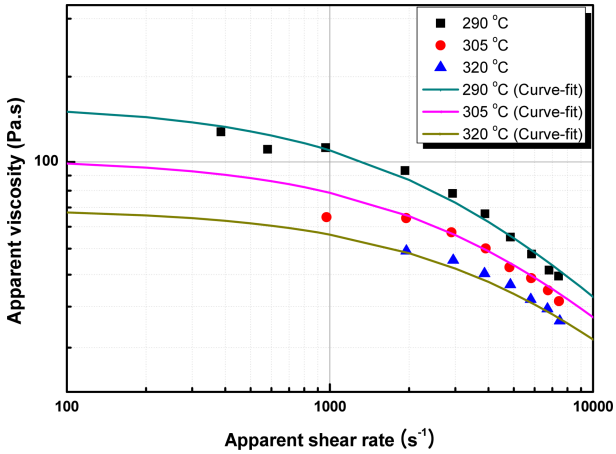

Fig. 5. (Color online) Apparent viscosity of the polymer obtained at various shear rates and temperature.

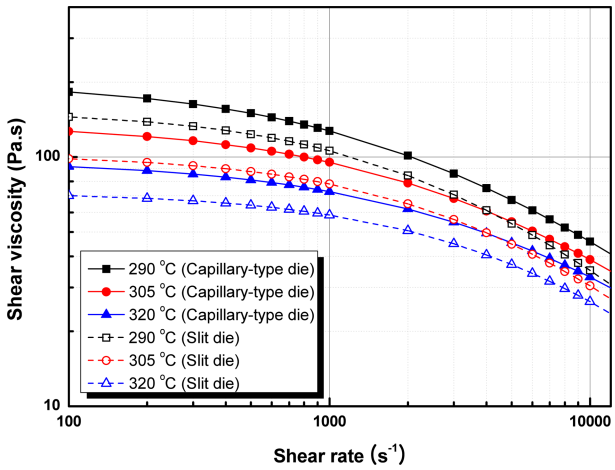

Fig. 6. (Color online) True viscosity of the polymer obtained at various shear rates and temperature.

Table 2. Cross-WLF coefficients from the slit- and the capillary-type die

Coefficients	Slit-die	Capillary-type die
n	1.2348 E-01	2.3125 E-01
τ^* (Pa)	3.8315 E+05	4.1185 E+05
D_1 $(Pa.s)$	1.7392 E+09	5.3195 E+10
D_2 $(°K)$	4.1715 E+02	3.9715 E+02
D_3 $(°K/Pa)$	0.0000 E+00	1.0000 E-07
A_1	2.1981 E+01	2.5425 E+01
A_2 $(°K)$	5.1600 E+01	5.1600 E+01

eter of 30 mm). The lengths (L/D) of the two capillary dies are 32 mm (16) and 7.97 mm (3.98), respectively. The Cross-WLF coefficients measured from the slit-die and the capillary-type die are summarized in Table 2. At all temperature ranges, the measured viscosities from the capillary-type rheometer are found higher than those

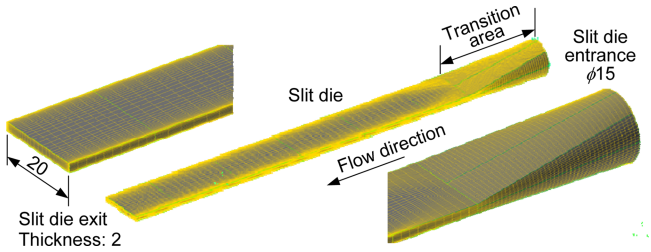

Fig. 7. (Color online) The details of the mesh used for CFD simulation at the entrance and the exit of the slit-die.

obtained from the slit-die rheometer as can be seen in Table 2. It seems that the melt preparation affects the polymer behavior and the measured properties.

3.3. Comparison of the pressure drop with flow simulations

For comparison and validation of the shear viscosity obtained from the slit-die rheometer, the flow inside the slit-die has been simulated using a commercial CFD software, Fluent. The finite element mesh in the simulation is shown in Fig. 7. The finite element mesh has 140,000 tetrahedral elements. As for the boundary conditions, the flow rates are assigned at the entrance of the die. Zero pressure condition is assigned at the die exit. No-slip condition is applied on the remaining boundary. To assess the accuracy of the viscosity prediction, pressures are calculated using the viscosity obtained from the slit-die rheometer or that from the capillary-die. The calculated pressures are compared with the experimental results.

Fig. 8 (a) shows the measured pressure data and the calculated values using the viscosity model parameters from the slit-die rheometer. Fig. 8 (b) shows the measured pressure data and the calculated values using the viscosity from the capillary-type rheometer. A good agreement is found between the measured (dots) and the predicted pressures (solid lines) with the viscosity parameters from the

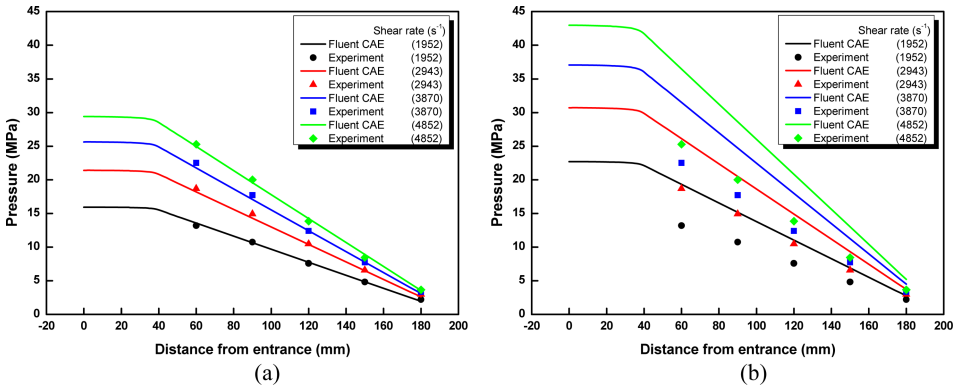

Fig. 8. Comparison of the experimental data with the predicted results with the viscosities from (a) the slit-die rheometer and (b) the capillary-type rheometer at the melt temperature of 320°C.

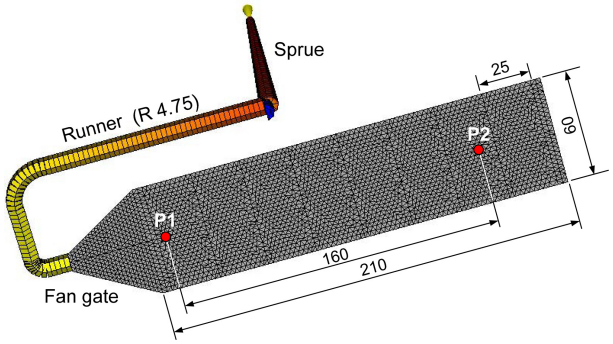

Fig. 9. Rectangular cavity geometry and mesh configuration.

slit-die rheometer, as shown in Fig. 8 (a). However, the simulation result with the viscosity obtained from the capillary-type rheometer in Fig. 8 (b) is found unsatisfactory: The pressure drop is higher than the corresponding experimental data in this case, which is consistent with the higher viscosity obtained the capillary rheometer in Fig. 6 and Table 2.

3.4. Flow analysis during mold filling phase

Having validated the accuracy of the viscosity obtained from the slit-die rheometer, a flow analysis during the filling phase of the injection molding process has been performed using the measured viscosity. We remark that this flow analysis is much more complicated than the slit-die flow, as the working fluid is short-fiber filled polymer melts. Especially, fiber orientation distributions along both the thickness direction and the in-plane direction in diverging flows affect local viscosity distribution. Of course there are other factors such as non-isothermal effects due to cooling and solidification. However, the objective of this work is the assessment of the applicability and the limitation of the measured viscosity model in predicting polymeric flow behaviors during the filling processes.

MPI 2012 from Autodesk was used for the flow analysis using the viscosity obtained from the slit-die rheometer and that provided by Autodesk Moldflow Plastics Labs. The other material properties for the flow analysis are shown in Table 1. Two rectangular cavities with different thicknesses (2 mm and 3 mm) are chosen as the test model. The geometry of the cavity is shown in Fig. 9 along with location of the fan gate, a runner and sensor positions. The total length is 210 mm and the width is 60 mm. Two pressure sensors (Kistler 6190, 6195) are installed in order to measure the pressure inside the cavities. The total number of finite elements for the simulation is 5319. For the flow simulation, the flow rate, the melt temperature and the mold temperature are 50 cm^3/s, 300°C and 80°C, respectively.

Figs. 10 and 11 show the pressure obtained from experiment and simulation for the 2 mm and 3 mm thickness specimens. The dots represent the pressure profiles from the simulation and the solid lines from the experiment. In the simulations, the viscosity model data from both the slit-die and the capillary-type die provided by Autodesk are employed. Higher pressure values are observed from the numerical filling simulations of both viscosity model data than that from the experiment, particularly in case of the data from the sensor near the gate (P1). The discrepancy between simulations and measured data is smaller with the slit die viscosity data than that with the capillary-die data. Specifically, for the 2 mm-thick cavity, the pressure measured near the gate (P1) was 32.39 MPa at the end of the filling, while the pressures calculated

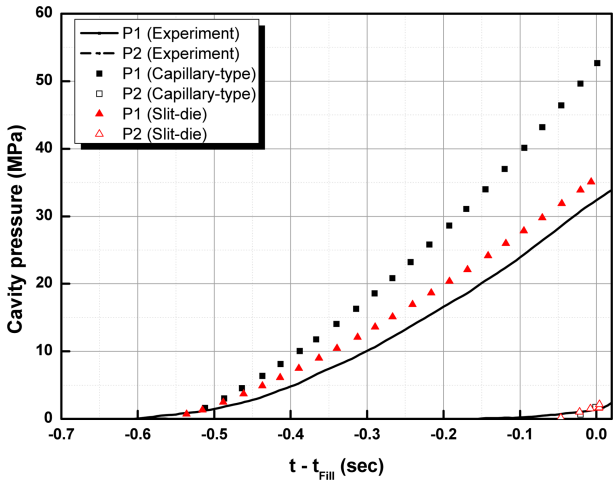

Fig. 10. Comparison of measured and calculated pressures at 2 sensor locations (cavity thickness = 2 mm, injection speed = 28 mm/s).

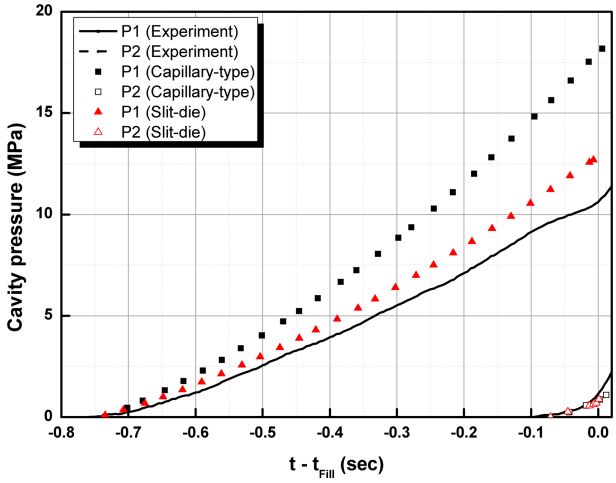

Fig. 11. Comparison of measured and calculated pressures at 2 sensor locations (cavity thickness = 3 mm, injection speed = 28 mm/s).

using the viscosity obtained from the slit-die was 35.07 MPa and that from the capillary rheometer was 52.66 MPa as shown in Fig. 10. For the 3 mm-thick cavity, the pressures obtained by experiment and the simulation using the viscosity from the slit-die and the capillary-type die were 10.8 MPa, 12.68 MPa and 17.53 MPa, respectively. The analysis using the viscosity from the slit-die viscosity data (Fig. 8) is 8 to 17 percent higher than the actually measured pressure. The accuracy of the pressure profile is directly related with the accuracy in the viscosity data and therefore the inaccurate high viscosity prediction of the capillary-type die seems to be responsible for the excessively high pressure prediction in filling simulations (Figs. 10 and 11).

4. Conclusions

In this study, a slit-die rheometer mounted on an injection molding machine was used to measure the viscosity of polycarbonate with 40 wt% of glass fiber. The measured viscosity was used to perform a flow analysis during the filling phase of an injection molding process and the predicted results were compared with the experiment results.

Below is the summary of this study.

1. Pressure sensors installed on the rectangular slit measured the pressure profile. The result showed a linear behavior in general, and this indicates that viscous heating in the flow inside the slit-die does not have significant impact on the shear viscosity for this case. And also, the distance between the end of the transition zone and the first pressure sensor of 20 mm is sufficient, to establish a stead and fully developed flow.
2. For the same material, the pressure predicted using the viscosity from the capillary rheometer is higher than that obtained from the slit rheometer, because the measured viscosity of the capillary-type die is higher.
3. The analysis results using the viscosity from the slit-die are 8 to 17 percent higher than the actual pressures measured by the pressure sensors installed on the rectangular cavity. The difference between the two results is bigger when the cavity thickness is 3 mm than when the cavity is 2 mm thick.

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