Rheological and thermal analysis of the filling stage of injection moulding

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Abstract. Flow conditions are different in the cavity of the injection mould from the capillary flow of a laboratory rheological instrument. An injection moulding slit die rheometer (Rheo-mould) was designed with a series of slit and orifice dies. Four pressure sensors were built in the stationary side of the mould, therefore the pressure could be measured at four different places. A changeable slit die insert was designed in the moving side. The shear stress and the shear rate can be calculated from the pressure gradient and from the flow rate of the melt, respectively. Flow curves of low density polyethylene were determined using Bagley, Rabinowitsch and Mooney corrections. The results were compared to the flow curves determined by Göttfert and Haake capillary equipments. It was found that the agreement between the methods is excellent.

Keywords: rheology, injection moulding, cavity pressure, mould filling

1. Introduction

Determination of rheological properties of molten polymers is a considerably studied field of Polymer Physics. Knowledge of real viscosity of polymer melt in the injection moulding process is extremely important. The flow of polymers is a complex process, as the viscosity of plastics, apart from temperature and pressure, depends on the molecular mass, molecular mass distribution and on the deformation rate. These make plastic processing rather difficult. During the flow in nozzle, runner system and in thin walled cavities of injection moulds, both shear and elongation deformation processes are developed, and the deformation rates are generally high. Many laboratory instruments can be found on the market but only a few of them are suitable for measuring the flow properties of polymeric material in the processing environment. On-line extrusion viscometers are wide-spread instruments, but injection

moulding machines are hardly used for rheological tests [1–6].

One of the main goals of our research is to develop a special injection mould which is suitable to determine the rheological properties of polymers at a high deformation rate. In the first stage of the research, two pressure sensors were built into a standard injection mould. Basic measurements and calculations were carried out by it. Then a spiral injection mould was designed, and after some success, it was upgraded to a changeable slit die system. In the final set-up, most of the important parameters can be controlled [7, 8].

The importance of the results is to get reliable data for the simulation of the injection moulding process. Reliability of filling simulations depends mainly on the following four factors [7]: input data, process parameters, software applications and numerical models. In particular, among the processing param-

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eters, the rheological characteristics of polymer melts have the largest influence on the filling simulation.

2. Theoretical background and calculations

There are two approaches to describe flow behaviour of a rheologically complex fluid: continuum mechanics and molecular approach. The molecular approach is rather complex, as molecular structure and molecular motions are considered to describe the relationships. Considering the material as a continuum, stress-strain relationships can be calculated by mathematical equations based on the theories of continuum mechanics. In principle, there are six independent components of stress that must be specified to completely describe the state of stress. However, in simple steady shear flow, fewer number of stress components is necessary. For example, stress components in a cube are shown in Figure 1, and the corresponding stress and deformation rate tensors are shown in Equation (1) and Equation (2), respectively:

$$\tau = \begin{bmatrix} \tau_{zz} & \tau_{zy} & 0 \\ \tau_{yz} & \tau_{yy} & 0 \\ 0 & 0 & \tau_{xx} \end{bmatrix} \quad (1)$$

$$\dot{\gamma} = \begin{bmatrix} 0 & \dot{\gamma} & 0 \\ \dot{\gamma}_{yz} & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix} \quad (2)$$

where τ is the shear stress tensor, $\dot{\gamma}$ is the shear rate tensor, x , y and z shows the directions. τ_{zz} means the shear stress acting on the surface perpendicular to the z axis and showing in z direction.

Different kinds of measuring systems can be used for rheological measurements. Mostly simple shear flow is carried out in capillary or in slit die. In steady shear flow there are no more than three independent material functions needed for correlating the stress quantities of rheological significance to the shear

rate. These viscosity functions are given by Equations (3–5):

$$\tau_{12} = \tau(\dot{\gamma}), \quad \eta(\dot{\gamma}) = \frac{\tau(\dot{\gamma})}{\dot{\gamma}} \quad (3)$$

$$\tau_{11} - \tau_{22} = N_1(\dot{\gamma}), \quad \xi_{12}(\dot{\gamma}) = \frac{N_1(\dot{\gamma})}{\dot{\gamma}^2} \quad (4)$$

$$\tau_{22} - \tau_{33} = N_2(\dot{\gamma}), \quad \xi_{23}(\dot{\gamma}) = \frac{N_2(\dot{\gamma})}{\dot{\gamma}^2} \quad (5)$$

in which $\eta(\dot{\gamma})$, N_1 and N_2 are called material functions. $\eta(\dot{\gamma})$ defines the shear dependent viscosity, ξ_{12} defines the first normal stress function, ξ_{23} defines the second normal stress function [9].

In the measurements by capillary rheometers, the flow rate (shear rate) is controlled and the pressure drop (shear stress) is measured. The apparent values can be calculated from the primary data, but some corrections must be made for the determination of real functions.

In the calculations, apparent values are used. The apparent shear rate, shear stress and viscosity can be calculated if slit die is used by Equations (6–8):

$$\dot{\gamma}_{app} = \frac{6Q}{h^2w} \quad (6)$$

$$\tau_{app} = \frac{h\Delta p}{2l} \quad (7)$$

$$\eta_{app} = \frac{\tau_{app}}{\dot{\gamma}_{app}} \quad (8)$$

where Q is the volumetric flow rate, h is the height of the die, w is the width of the die, l is the flow length, Δp is the pressure drop, τ is the shear stress, $\dot{\gamma}$ is the shear rate.

In order to obtain accurate experimental results, the height-to-width ratio (h/w) of the slit die should be near to zero ($w \gg h$), because the existing theoretical equations converting Q and $\Delta p/l$ to shear rate and shear stress, respectively, are based on the assumption that the velocity does not depend on lateral position. Approaching w/h ratio to zero, the edges make negligible contribution to the pressure drop. In this case the corrected shear rate and shear stress are calculated by Equations (9–11) [10]:

$$\tau_w = \frac{\Delta p h}{2l} \left(\frac{1}{\frac{h}{w} + 1} \right) \quad (9)$$

Figure 1. Shear flow in a slit die. (w : width of the die; h : height of the die, l : flow length)

where h is the height and w is the width of the die. Δp is the pressure drop, l is the measured flow length (Equation (10)):

$$\dot{\gamma} = \frac{2}{3} \left(\frac{6Q}{h^2} \right) \left(\frac{b}{f} + \frac{a}{f} \frac{1}{n} \right) \quad (10)$$

where Q is volume rate, a , b , f are correction factors, n is the power law index (Equation (11)):

$$n = \frac{d \log \tau_f}{d \log \dot{\gamma}_f} \quad (11)$$

Some material might slip on the wall of the die. Figure 2 shows the velocity profile of the melt if it slips. Using different slit die cross sections but same flow length, the velocity of the wall slip can be determined [11].

The total volumetric flow rate (Q_T) can be calculated by Equation (12):

$$Q_T = \frac{h^3 w}{6} \dot{\gamma} \quad (12)$$

The volumetric flow rate by the slip (Q_{slip}) is given by the Equation (13):

$$Q_{\text{slip}} = whv_{\text{slip}} \quad (13)$$

where v_{slip} is the velocity of wall slip.

The apparent shear rate can be plotted as function of $1/h$ and the wall slip velocity can be calculated from the slope of the curve described in Equation (14):

$$\dot{\gamma}_{\text{app}} = \dot{\gamma}_{\text{true}} + \frac{6v_{\text{slip}}}{h} \quad (14)$$

According to Equation (7), the pressure gradient is a linear function of flow length. If it is not valid, Bagley correction should be applied.

The viscosity of molten polymer is really high, basically it is between 10...10.000 Pa·s in injection moulding or extrusion process. Due to the high viscosity, the dissipation heat and the effect of pressure

are not negligible. The average temperature rising (ΔT) can be calculated by Equation (15):

$$\Delta T = \frac{\Delta p}{\rho c_p} \quad (15)$$

where Δp is the pressure drop, ρ is the density, c_p is the specific heat of the material. Equation (15) assumes that there is no heat exchange between the material and the mould (wall of the die). In real cases there is some heat transfer between the mould and the polymer melt. This transfer depends on the temperature difference, therefore changing the mould temperature it is possible to influence the heat exchange rate. Dimensionless numbers (Graetz-, Nahme- Brinkman-number) are used to characterise the thermal conditions of the flow of polymer melt. Graetz-number (Gz , Equation (16)) [12] compares the heat transfer in the flow direction to the perpendicular direction:

$$Gz = \frac{2\rho c_p Q h}{\lambda w l} \quad (16)$$

where ρ is the density of the material, c_p the specific heat capacity, λ is coefficient of thermal conduction. If $Gz \gg 1$, the material temperature in the calculation and the incoming material temperature are the same [13].

Nahme-number (Na , Equation (17)) compares the temperature rising by dissipation heat to the temperature influence to the viscosity:

$$Na = \frac{\eta Q^2 \alpha_\eta}{4kh^2 w^2} \quad (17)$$

where α_η is temperature coefficient of viscosity.

If $Na > 1$, the dissipation heat has significant effect on the viscosity.

Brinkman-number (Br , Equation (18)) compares the dissipation heat to the perpendicular heat transfer:

$$Br = \frac{\eta Q^2}{4kh^2 w^2 |T_{\text{in}} - T_{\text{wall}}|} \quad (18)$$

where T_{in} is the incoming material temperature, T_{wall} is the temperature of the wall of the cavity.

If $Br > 1$, the dissipation is the major process, therefore the temperature drop of the material is negligible. If the mould temperature and the material temperature are the same, the Brinkman-number is infinite.

Figure 2. Velocity profile of the melt

According to Laun studies [13–14], the real pressure drop (allowing for the dissipation heat and the effect of the pressure) can be calculated by Equation (19):

$$\Delta p \approx \Delta p_0 \exp(\beta_\eta p_1) \left[1 - (\beta_\eta + \alpha_\eta \varepsilon_p) \frac{\Delta p_0}{2} \exp(\beta_\eta p_1) \right] \quad (19)$$

where Δp_0 stands for the pressure loss without pressure and temperature effects, β_η pressure coefficient of viscosity α_η temperature coefficient of viscosity, Δp is the measurable pressure difference ($\Delta p = p_1 - p_2$).

$$\varepsilon_p = \frac{1}{\rho_0 \left(c_p + \frac{\Lambda}{\dot{m}} \right)} \quad (20)$$

Equation (20) represents a dissipative heating coefficient (ε_p) (Λ lump heat transfer coefficient summarising all types of heat loss out of the melt). The adiabatic case (insulated walls) is got by setting $\Lambda = 0$, $\dot{m}$ represents the mass flow rate, ρ_0 is the melt density [13].

3. Experimental

3.1. Materials

TIPOLEN FA2210 (Tisza Chemical Group PLC, Tiszaújváros, Hungary) low-density polyethylene material was used for the measurements. This material is recommended for blow moulding technology, so the viscosity of the material can be measured easily during shear and the deformation caused by elongation. The characteristics of the PE are summarised in Table 1.

Table 1. Main properties of the material

Properties	Value
MFI (190°C; 2,16 kg)	0.3 g/10 min
Density (23°C)	0.922 g/cm ³
Maximum viscosity (190°C)	90 000 Pa·s
Apparent activation energy of flow	31.5 kJ/mol

3.2. Measuring system

An ARBURG Allrounder 270 C 350-70 standard injection moulding machine was used in the experiment. The most important machine parameters are listed in Table 2.

There are some instrumented injection moulds [1–5] but the one developed by us is basically different

Table 2. The most important machine parameters

Parameters	Value
Injection rate (Q)	5...90 cm ³
Injection pressure (pinjection)	1500 bar
Material temperature (T_{mat})	180...220°C
Mould temperature (T_{mould})	30...220°C
Screw rotation speed (n)	150 mm/s
Back pressure (p_{back})	20 bar
Switch-over point (s_{switch})	1 cm ³
Packing time (t_{pack})	0 s

Figure 3. Active plates and the accessories of the system

from them. The material enters in the centreline of the mould from the sprue bush and goes to the parting line. There is a quick-change slit die system in the moving side. The material flow through the die and came out at the end of it. Three pressure sensors (Kistler 6157BD) and one P-T combo sensor (Kistler 6190A) were built into the fixed side. The mould temperature was controlled by cooling fluid to 90°C, and an electric heating system was used above 90°C. The drawing of the mould can be seen in Figure 3.

Figure 4 shows the positions of the sensors. The dimensions of the dies used in the experiments are listed in the Table 3. The width of the dies was the same, 15 mm.

A virtual instrument was developed by LabView software environment for data processing. It can control the database and calculate the rheological properties from the measured pressure in Windows XP environment. The applied sampling rate was 0.001 sec in the measurements. This relatively high sampling rate is necessary to study the melt fracture effect. The schematic drawing of the electronic system is shown in Figure 5.

Figure 4. Slit die insert

Table 3. Geometry of the dies

No. of slit die	1.	2.	3.	4.	5.	6.
Height (h) [mm]	2	3	4	2	2	2
Flow length (l) [mm]	80	80	80	45	65	80
Width (w) [mm]	15	15	15	15	15	15

Figure 5. Signal transducer system

The measured four pressure curves and the pressure differences are plotted as a function of time in Figure 6. It can be seen that the cavity filling has two different phases. The material flows inside of the cavity in the first stage. The pressures increase continuously until the melt front reaches the outlet point.

Figure 6. Measured pressure curves

The molten polymer flows out freely from this time and the measured pressures are roughly constant. These constant pressures were used in the calculations. The pressure drops close to the end of the cavity between p_3 and the p_4 pressure were smaller than the former pressure drops. It can be explained by the end effect of the die.

Minimum three injections were made by each set up and the pressure curve was recorded if the standard deviation was less than 5%. The average deviation was less than 1%.

4. Results and discussion

4.1. Investigation of the flow

The measuring range was 500–9000 1/s. Three different temperatures were investigated. The calculated apparent flow curves are shown in Figure 7. These curves correspond to the curves of pseudo plastic materials: increasing the shear rate, the viscosity of the melt decreases.

Correction of cross section showed that the apparent shear rate increased (Rabinowitsch correction), and the apparent shear stress decreased (Figure 8).

Figure 7. Effect of the cross section correction

Figure 8. Flow curves calculated with and without the wall effect

Figure 9. Mooney plots

The Mooney correction was calculated by the measurement of die inserts No 1,2,3. It was found that the slope of the Mooney lines were negative so there is no slip on the wall (Figure 9)

The Bagley correction was calculated by the measurement with inserts No. 4, 5 and 6 (Figure 10). It can be seen that the smallest l/h ratio was 15. It is rather far from the ideal l/h range (close to 0), however the regression constants of the lines are above 0,98. The calculated values are summarised in Table 4.

The apparent values were corrected and the real viscosity curves are shown in Figure 11. For comparison, the material was tested by two other kinds of

Figure 10. Bagley plot ($T_{\text{mat}} = 180^\circ\text{C}$)

Table 4. Constants of the Bagley plots

$\dot{\gamma}$ [1/sec]	Slope of Bagley plots [Pa·s]	Extrapolated pressure drop [Pa]	Linear regression (R^2)
500	265 175	2 415 699	0.9878
1 000	290 716	3 046 552	0.9877
2 000	318 000	3 809 722	0.9874
3 000	335 000	4 327 469	0.9873
5 000	357 000	5 065 246	0.9870
8 000	378 000	5 838 593	0.9868
10 000	389 000	6 240 761	0.9866
15 000	408 000	7 034 650	0.9864

Figure 11. Real viscosity curves

Table 5. The constants of the viscosity curves

Instrument	μ_1	n
Haake	29 706	0.250
Göttfert	30 349	0.246
Rheo-mould	29 091	0.270

instruments. Haake extrusimeter (Miskolc University) and Göttfert Rheograph 25 were used. Correlation of the results of the different instruments is excellent.

The Equation (21) was fit to the calculated real viscosity curves, where μ_1 is a constant without real physical meaning. The constants of the equation are listed in the Table 5 (Equation (21)).

$$\eta = \mu_1 \dot{\gamma}^{n-1} \quad (21)$$

4.2. Investigation of thermal condition by dimensionless quantities

Calculations were made by the dimensionless quantities (Equations 16–18) using data of the minimum and the maximum flow rate. The mould and material temperature and the cross section of the slit die were changed, too. The results are summarised in Table 6 and Table 7.

Table 6. Dimensionless quantities ($T_m = 180^\circ\text{C}$)

Mould	T_{mould}	Flow rate	Graetz-number	Nahme-number	Brinkman-number	p_1 [bar]
Die 1 $h = 2$ mm $w = 15$ mm	30°C	min.	183	25	3	300
		max.	2198	1353	42	350
	90°C	min.	183	29	1.23	173.8
		max.	2198	681	28.6	345.4
	180°C	min.	–	–	–	149
		max.	–	–	–	290
Die 2 $h = 3$ mm $w = 15$ mm	30°C	min.	274	35	1.6	147.9
		max.	4946	992	44	232.7
	90°C	min.	274	34	2.1	136.8
		max.	4946	1388	64.2	234.4
Die 3 $h = 4$ mm $w = 15$ mm	30°C	min.	366	29	1.3	90.6
		max.	6595	807	36.6	144.8
	90°C	min.	366	29	1.9	88.7
		max.	6595	859	55	148.6

Table 7. Dimensionless quantities ($T_m = 220^\circ\text{C}$)

Mould	T_{mould}	Flow rate	Graetz-number	Nahme-number	Brinkman-number	p_1 [bar]
Die 1 $h = 2$ mm $w = 15$ mm	30°C	min.	183	26	2	260.5
		max.	2198	570	44	318.3
	90°C	min.	183	20	1.36	150.9
		max.	2198	520	34	215.4
	220°C	min.	–	–	–	106
		max.	–	–	–	270
Die 2 $h = 3$ mm $w = 15$ mm	30°C	min.	274	24	1.41	127.4
		max.	4946	563	33	170.3
	90°C	min.	274	29.2	1.26	114.8
		max.	4946	879	38	196.7
Die 3 $h = 4$ mm $w = 15$ mm	30°C	min.	366	23	0.92	74.4
		max.	6595	608	24.4	115.0
	90°C	min.	366	15	0.7	46.3
		max.	6595	700	17	119.6

It can be seen that the Graetz number is larger than 100 in every set up. As the calculated Nahme numbers are much larger than 1, the cross heat flow is much smaller than the axial heat flow. However, it should be emphasised that on the cavity wall a frozen polymeric layer was developed affecting the efficient cross-section of the flow. This thickness is proportional to the pressure drop; the higher the pressure drop is, the thicker the frozen layer is. According to the measured and calculated values (Table 6, Table 7), increasing the height of the cavity the effect of the mould temperature on the pressure becomes smaller meaning that the frozen layer has less effect on the flow. Because of this frozen layer, the flow cross section must be smaller, and it has effect on the pressure drop and the dissipation heat, too. Applying the thinnest die ($h = 2$ mm) and

cooled mould temperature, the pressure drop was the highest.

Brinkman number was larger than one in every setup (except two measurements with die 3 at 220°C). This means that the dissipation heat has a larger effect on the viscosity than the perpendicular heat transfer.

The measured pressure drop between two sensors ($\Delta p_{\text{measured}}$), the average temperature rising by dissipation heat (ΔT_{dissip}), the real viscosity (η_{measured}), the viscosity decrease by dissipation heat (η_{dissip}), the viscosity decrease by pressure (η_{pressure}) and the corrected pressure drop ($\Delta p_{\text{corrected}}$) are summarised in Table 8.

The effect of the pressure and the temperature ($\Delta p_{\text{corrected}}$) were calculated by Laun. $\Delta p_{\text{measured}}$ and $\Delta p_{\text{corrected}}$ are in good correlation. The average dif-

Table 8. Effect of the dissipation heat and the pressure on viscosity

	T _{material} [°C]	180	180	220	220
	T _{mould} [°C]	30	180	30	220
		Q			
$\Delta p_{\text{measured}}$ [MPa]	min.	10.67	12.54	11.42	8.53
	max.	15.95	28.24	14.9	20.65
$\Delta T_{\text{dissip.}}$ [°C]	min.	5.4	6.33	5.7	4.31
	max.	8	14.27	7.5	10.4
η_{measured} [Pa·s]	min.	267	313.5	285.6	213.3
	max.	20	35.3	18.59	25.8
η_{dissip} [Pa·s]	min.	232	265.8	245.8	190.7
	max.	16.1	24.35	15.3	19.67
η_{pressure} [Pa·s]	min.	277.6	328.2	297.8	220
	max.	21.2	39.14	19.7	27.8
$\Delta p_{\text{corrected}}$ [MPa]	min.	11.4	13.47	12.23	9.03
	max.	17.2	28.1	16	22.04

ference is less than 6%. This good correlation means that the dissipation heat equalises the effect of the pressure on the viscosity of the melt.

5. Conclusions

A new type of rheological measuring system has been designed and manufactured. The special injection mould can be used to determine the flow curve of molten polymers during the injection moulding process. If the mould is heated up to the processing temperature, the flow condition is close to adiabatic, especially by higher flow rate. It was found that the mould temperature has a smaller effect on the flow condition if the cross section is bigger than 2 mm.

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