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Measuring the pressure dependent viscosity at high shear rates using a new rheological injection mould

Summary — To shorten the development time for injection moulded parts, process simulations in different developing steps as well as quick and accurate measurements of rheological data even for the increasing variety of special compounds are necessary. For simulation of thin wall injection moulding accurate viscosity data measured at shear rates up to 800.000 s^{-1} and more are important, but not available in any commercial material database. Therefore, within the EU project "*Pro4Plast*", an easy-to-handle rheological measurement system was developed by the Institute of Polymer Processing, Montanuniversitaet Leoben, Austria. The rheological mould is constructed like a standard injection mould with interchangeable dies. It can be mounted onto any conventional inection moulding machine and enables operators (manufacturers, especially SMEs) to measure viscosity in time on their own machines at practically relevant shear rates (from 100 s^{-1} to $2 \cdot 10^6$ s^{-1}). A special feature allows measuring the pressure dependency of viscosity using closed-loop counter pressure control. Experimental data are evaluated taking into account the melt temperature rise due to dissipative heating.
Keywords: rheology, viscosity, pressure dependency, high shear rate, dissipative heating.

BADANIE ZALEŻNOŚCI LEPKOŚCI OD CIŚNIENIA W WARUNKACH DUŻEJ SZYBKOŚCI ŚCINANIA Z ZASTOSOWANIEM NOWEJ REOLOGICZNEJ FORMY WTRYSKOWEJ

Streszczenie — W celu skrócenia czasu opracowywania nowych rozwiązań w dziedzinie przetwórstwa wtryskowego niezbędne jest stosowanie symulacji procesowych na różnych etapach tego opracowywania a także prowadzenia szybkich i dokładnych pomiarów reologicznych dotyczących wielu różnych gatunków tworzyw polimerowych. Do symulacji wytwarzania wyrobów cienkościennych potrzebne są dokładne wartości lepkości mierzone w warunkach szybkości ścinania przekraczającej nawet 800 000 s^{-1}, których nie obejmują komercyjne bazy danych materiałowych. Dlatego w ramach projektu "*Pro4Plast*" finansowanego przez Unię Europejską, Institute of Polymer Processing, Montanuniversitaet Leoben (Austria) opracował łatwy do stosowania system pomiaru danych reologicznych. W projekcie uczestniczyło 28 partnerów z 10 krajów UE, przy czym Polskę reprezentowało Polskie Stowarzyszenie Przetwórców Tworzyw. Forma reologiczna została zaprojektowana jako typowa forma wtryskowa z wymiennymi dyszami (rys. 1). Może ona być montowana w standardowych wtryskarkach (rys. 2) i umożliwia firmom, zwłaszcza małym i średnim przedsiębiorstwom (SMEs) samodzielne pomiary lepkości z wykorzystaniem własnych maszyn w warunkach odpowiednich szybkości ścinania (od 100 s^{-1} do $2 \cdot 10^6$ s^{-1}). Specjalnie opracowana funkcja umożliwia pomiary zależności lepkości od ciśnienia. Przedstawiono wyniki odpowiednich badań procesu wtryskiwania polipropylenu, w których uwzględniano też wzrost temperatury stopionego tworzywa związany ze zjawiskiem rozpraszania ciepła (rys. 3—5).
Słowa kluczowe: reologia, lepkość, zależność od ciśnienia, duża szybkość ścinania, ciepło rozpraszane.

The injection moulding and mould making industry with 1.5 million employees and an annual turnover of EUR 150 billion, mainly concentrated in small and medium-sized enterprises (SMEs), is one of the key industries in Europe now. One of the most important key challenges in a competitive environment is the production of complex and highly functional parts in a fast and cost-ef-

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ficient way. To shorten the development time for injection moulded parts, process simulations in different developing steps as well as quick and accurate measurements of rheological data even for the increasing variety of special compounds are necessary. During the small and thin-walled injection moulding process, shear rates often exceed 10^6 s^{-1} and the injection pressure often exceeds $2 \cdot 10^8$ Pa (2000 bar). Hence for the finite elements method (FEM) simulation of such injection moulded parts, accurate viscosity data measured at shear rates up to $8 \cdot 10^5$ s^{-1} and more — including pressure dependence — are important. More often the pressure dependency of viscosity is neglected and such material data are not available in any commercial material database.

This data cannot be measured using a standard capillary rheometer only. The rheological measurement on the injection moulding machine offers a possibility to measure the viscosity of polymeric materials at practically relevant high shear rates range. W. Knappe *et al.* showed viscosity measurement techniques for four rigid PVC compounds using a slit die rheometer in combination with an injection moulding machine [1]. C. Bader *et al.* showed the possibility to measure the viscosity of a liquid crystal polymer (LCP) material on the injection moulding machine by using a round capillary die which was attached to the machine nozzle [2]. O. Amano *et al.* developed a new measurement adaptor with a round capillary die to measure the flow properties of polymer melts on the injection moulding machine up to a shear rate of 10^6 s^{-1}. He also investigated the effect of pressure on the viscosity; for that purpose a manually adjustable needle valve was used to regulate the exit pressure to the required back pressure value. In his study he concluded that for the four investigated thermoplastic materials the pressure dependent viscosity data measured in the extremely high shear rate region has a significant influence to improve the accuracy of the FEM simulation results [3]. A. L. Kelly *et al.* showed rheological measurements in the shear rate range up to 10^7 s^{-1} by using an instrumented nozzle adaptor with a capillary die [4]. H. Takahashi *et al.* in [5] and A. L. Kelly *et al.* in [4] observed a second Newtonian plateau at shear strain rates above 10^6 s^{-1} [4, 5].

According to the published research results, rheological measurements up to shear rates of 10^6 s^{-1} were carried out without taking into account the temperature rise due to viscous heating. Hence, for a number of thermoplastics the measured data are presented in the form of apparent viscosity curves only. Therefore, within the EU project "*Pro4Plast*"*) an easy to handle and on-site-applicable rheological measurement system developed by the Institute of Polymer Processing, Montanuniversitaet Leoben, Austria (IKV-MUL). A specialized software was developed for the rheological evaluation of measured values and the approximation of true viscosity taking into account the temperature rise due to shear heating and compression heating by temperature correcting the measured pressure values with respect to actual temperatures.

*) In the project involved were 28 partners from 10 EU countries inluding Poland represented by Polish Plastics Converters Association.

INJECTION MOULDING MACHINE RHEOMETER

Rheological injection mould construction

In order to measure the flow behaviour within a wide shear rate range, a special injection mould (Fig. 1) with interchangeable conical slit-die inserts with varying slit heights has been developed. The rheological mould is very similar to a standard injection mould, but without sprue, runner system and cooling circuit. It can be easily

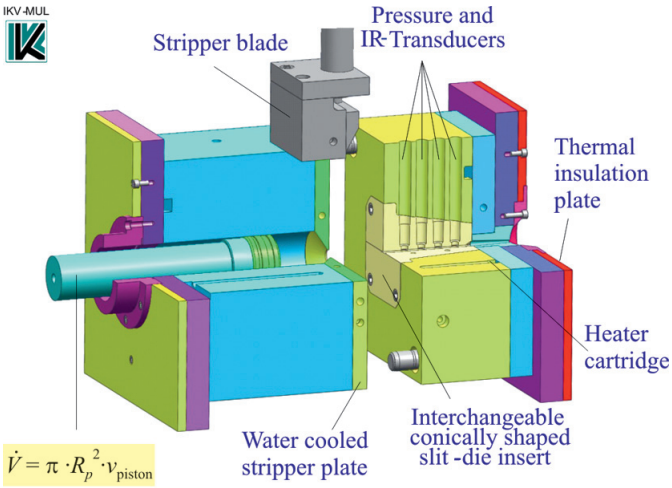

Fig. 1. Rheological injection mould

mounted onto an injection moulding machine and enables operators to measure viscosity on their own machines. In order to achieve a wide range of practically relevant shear rates three dies with different slit heights (1 mm, 0.5 mm and 0.35 mm, entrance angle of 60°) are used. The width of the flow channel is 10 mm and the total length is 105.5 mm, so to achieve a fully developed flow and to avoid entrance effects.

Along the flow path on the top and bottom side pressures and temperatures are measured by four sensors located 20 mm apart from each other in order to calculate shear stress and to identify the wall temperature increases along the flow length due to shear heating.

The pressure sensors are flush-type sensors, which have direct contact with the melt. Melt temperature is directly measured by infrared sensors. The wall temperatures are measured using a special kind of heat flux sensors, with three thermocouples positioned in such a way

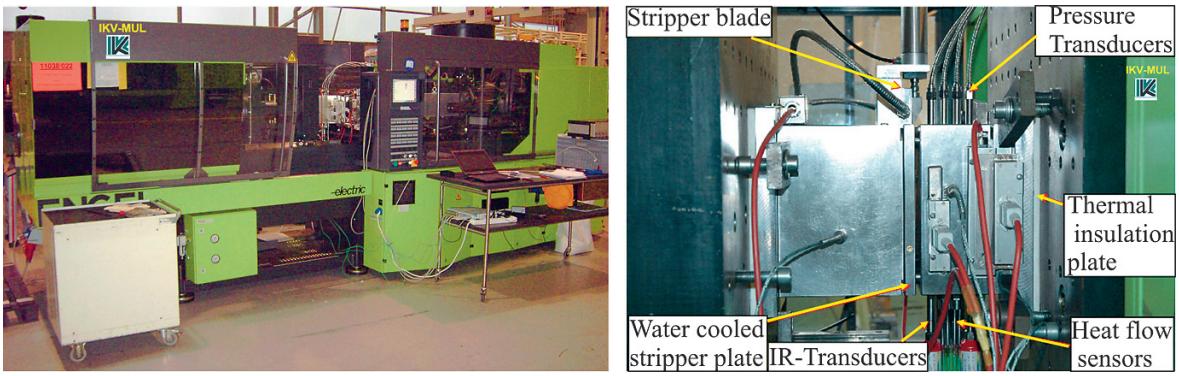

Fig. 2. Injection moulding machine with rheological injection mould

that each thermocouple measures the temperature at a distance of 1 mm, 1.5 mm and 3 mm away from the flow channel wall. The wall temperatures are calculated by linear extrapolation and measured near the entrance and the exit of the flow channel.

The movable mould part consists of a cylinder and a piston system to measure the volumetric flow rate and to calculate the corresponding apparent shear rate. Thus measurement errors due to leakage flow in the back flow valve of the plasticizing unit can be avoided. The displacement rate of the piston is measured using a stroke sensor. The forward and backward motion of the piston is attained by the movement of the machine ejection system. The volumetric flow measurement cylinder has a maximum storage capacity of 424.115 cm^3.

A pneumatically operated stripper blade is attached on the top of the mould to wipe the melt flowing out of the cylinder during ejection from the stripper plate.

Heating of the mould is done using four cartridge heaters as well as four heater plates covering the moveable mould part. They are controlled directly by the injection moulding machine.

Experimental equipment and procedure

After closing the mould the measurement starts with melt injection at the pre-specified constant injection velocity. The main parameters to be measured for calculating the viscosity are: pressure loss along the flow length, temperature of the polymer melt, wall temperature rise along the flow length and volumetric flow rate. The melt exits the flow channel and enters into the cylinder making the piston move in the backward direction.

A special feature allows measuring the pressure dependency of viscosity using closed loop counter pressure control. Data acquisition is done by commercial software, while the rheological data evaluation is done by the software "Rheosoft V.1.0" developed by the Institute of Polymer Processing, Montanuniversitaet Leoben, Austria. The measurements were performed using a special hybrid injection moulding machine, ENGEL VC 940/130, supplied by ENGEL Austria GmbH, Schwertberg, which is equipped with an electrical plasticizing unit.

To test and to optimize the new measurement system, the viscosity values were measured for several polymers and counter pressure up to $4 \cdot 10^7$ Pa (400 bar). The injection moulding machine with the complete measurement system is shown in Fig. 2.

RHEOLOGICAL EVALUATION TECHNIQUE

Calculation procedure

The main parameters to be measured for calculating the viscosity at a particular shear rate are pressure loss along the flow length, temperature of the polymer melt and volumetric flow rate.

The first step is to calculate shear rate and shear stress. The apparent shear rate ($\dot{\gamma}_{app}$) was calculated using the shape factor (F_p) for pressure flows [equation (1)]. If the shape factor assumes values smaller than 1, the two-dimensional geometry and its influence on shear rate is taken into account increasingly.

$$\dot{\gamma}_{app} = \frac{6 \cdot Q}{B \cdot H^2} \cdot \frac{1}{F_P} \tag{1}$$

where: Q — *volumetric flow rate*, B — *width of the slit*, H — *height of the slit*.

The actual volumetric flow rate is calculated from the rate of piston displacement, which is measured by a stroke sensor. By plotting the stroke length *vs*. time, we calculate the velocity of the piston.

The wall shear stress (τ) was determined using equation (2):

$$\tau = \frac{\Delta p}{2 \cdot \Delta L}\left(\frac{B \cdot H}{(B+H)}\right) \tag{2}$$

where: Δp — *pressure drop along flow length*, ΔL — *flow length*.

The true shear rate ($\dot{\gamma}$) was obtained from the $\dot{\gamma}_{app}$ value by applying the correction for the non-Newtonian behaviour of the melt according to Weissenberg-Rabinowitsch [equation (3)]:

$$\dot{\gamma} = \frac{\dot{\gamma}_{app}}{3}\left(2 + \frac{d \lg \dot{\gamma}_{app}}{d \lg \tau}\right) \tag{3}$$

After this correction, the true viscosity (η) of the polymer melt can be calculated by dividing the wall shear stress by true shear rate [equation (4)]:

$$\eta = \frac{\tau}{\dot{\gamma}} \tag{4}$$

The Cross viscosity model as in the equation (5) was used for the approximation of the shear dependency of the measured viscosity curves.

$$\eta = \frac{\eta_0}{\left[1 + \left(\frac{\eta_0}{\tau^*} \cdot \dot{\gamma}\right)^{(1-n)}\right]} \tag{5}$$

where: η_0 — *zero shear viscosity,* τ^* — *critical shear stress roughly characterizing transition shear stress from the Newtonian range to the pseudo-plastic region,* n — *shear rate sensitivity (0 < n < 1, where 1 – n roughly characterizes the slope of the line over the pseudo-plastic region in the logarithmic plot [6]).*

The above calculated viscosity does not taken into account the temperature increase due to shear dissipation at high shear rates. In general, at very high shear rates, the wall temperature rises by up to 90 °C and the melt temperature also increases considerably due to shear dissipation, which can highly influence the viscosity of the melt. So the melt temperature rise due to dissipative heating and compression heat was taken into account by applying a special temperature correction factor and the temperature corrected viscosity was obtained [7]. For this purpose a thermo-dynamic dimensionless number, the so-called Cameron number, is used. Utilizing a modified Agassant method [8] the average melt temperature inside the flow channel is calculated and applied to adjust the viscosity.

The shear viscosities obtained under varying temperatures can be transformed into a single master curve by applying the time-temperature superposition principle.

Effect of pressure on viscosity

The pressure coefficient of the shear viscosity at constant shear rate ($\beta_{\dot{\gamma}}$) which accounts for the pressure dependency of measured viscosity, may be evaluated for different pressure levels by means of the Eyring-Hole theory for polymeric fluids from equation (6):

$$\beta_{\dot{\gamma}} = \frac{(\ln \eta_{0,2} - \ln \eta_{0,1})}{(p_2 - p_1)} \tag{6}$$

where: $\eta_{0,2}$ — *viscosity with pressure effect,* $\eta_{0,1}$ — *viscosity under reference pressure,* $(p_2 - p_1)$ — *pressure difference.*

The pressure coefficient of viscosity at constant shear stress (β_τ) can be calculated by shifting the measured viscosity curves at different counter pressures into the reference viscosity curve. The pressure shift factor (a_p) is calculated by equation (7):

$$a_p = \exp[\beta_\tau \cdot (p - p_0)] \tag{7}$$

where: $(p - p_0)$ — *pressure difference.*

The relationship between the pressure coefficient of viscosity at constant shear rate and pressure coefficient of viscosity at constant shear stress is shown in equation (8):

$$\beta_{\dot{\gamma}} = \beta_\tau \cdot n \tag{8}$$

where: n — *the flow index of power law.*

EXPERIMENTAL RESULTS

The experiments had been carried out with the injection moulding grade polypropylene PP HG313MO, which is a homopolymer manufactured by Borealis A/S.

Figure 3 shows the comparison of the viscosity values measured at 210 °C on the rotational rheometer (Rheometrics, RMS800), the high pressure capillary rheometer (HPCR) and the injection moulding machine with the rheological injection mould (slit die H = 1 mm). The viscosity values are approximated using the Cross viscosity model. Dissipative heating and compression heat were taken into account, so in Fig. 3 the temperature corrected viscosity is presented.

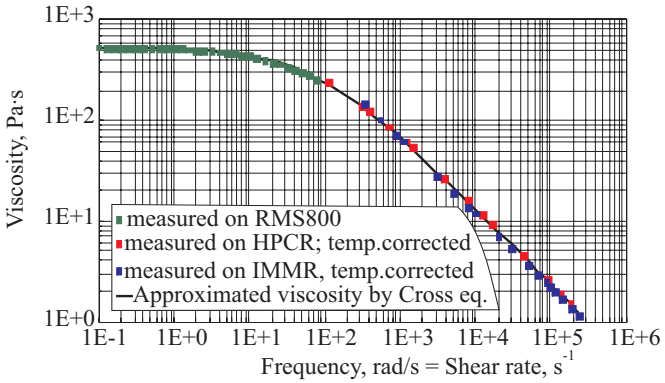

Fig. 3. Viscosity curve of polypropylene PP HG313MO at a temperature of 210 °C, measured using standard rheometers (RMS800 or HPCR) and the injection moulding machine rheometer (IMMR), cross approximation [9]

The viscosity values measured using three different rheometers remain quite close. Within the shear rate range from 3.000 s^{-1} to 200.000 s^{-1} the viscosity values measured with the rheological injection mould are 10 % lower than the viscosity values measured by the HPCR due to preheating of the melt in the plasticizing unit.

The effect of pressure on the viscosity of PP HG313MO at a temperature of 210 °C and 5 different counter pressures is presented in Fig. 4. A significant rise in measured viscosity values with increasing counter pressure is seen, which indicates that the pressure dependency of viscosity is not negligible.

Figure 5 shows the pressure invariant master curve of PP HG313MO at a temperature of 210 °C. The least-squares method was used to find the best fit for the approximation coefficients. The calculated pressure coef-

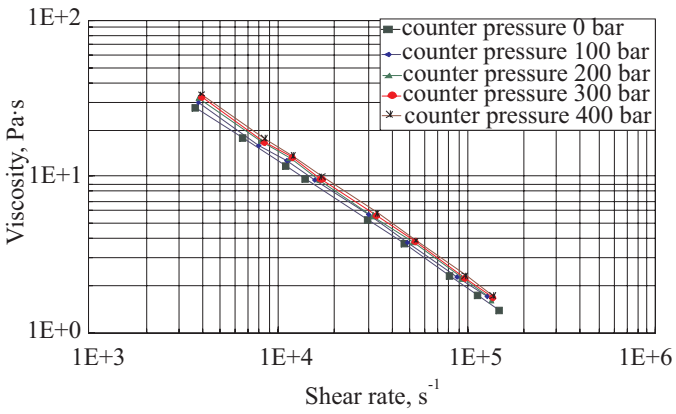

Fig. 4. Pressure dependency of polypropylene PP HG313MO, measured at a temperature of 210 °C and 5 counter pressure levels

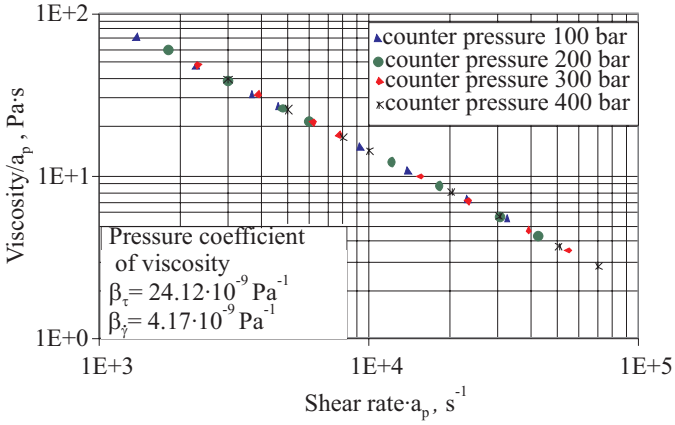

Fig. 5. Pressure invariant master curve of polypropylene PP HG313MO, shifted to the reference pressure of 2 · 10⁷ Pa (200 bar) at a temperature of 210 °C

ficient of viscosity at constant shear stress is $\beta_\tau = 24.12 \cdot 10^{-9}$ Pa^{-1}. The calculated pressure coefficient of viscosity at constant shear rate, which describes the value of the pressure coefficient of viscosity in the high shear rate range, is $\beta_\tau = 4.17 \cdot 10^{-9}$ Pa^{-1} and the flow index of power law is $n = 0.17$.

CONCLUSIONS

Within the EU project "*Pro4Plast*" a new measurement system for rheological measurements on the injection moulding machine was developed and successfully tested with several thermoplastic materials. Besides measuring viscosity at high shear rates the system allows to measure pressure dependency of viscosity within a wide range of shear rates.

For data evaluation purposes a special rheological software application was developed to calculate true viscosity values. This software takes into account the temperature rise due to shear heating at shear rates higher than 5.000 s^{-1} and the compression heat by adjusting the measured pressure values for temperature.

The influence of pressure on the viscosity of polypropylene grade PP HG313MO was studied at a temperature of 210 °C and counter pressure up to $4 \cdot 10^7$ Pa (400 bar). The calculated value of the pressure coefficient at constant shear stress for this type of PP is $24.12 \cdot 10^{-9}$ Pa^{-1}, which fits well with the results of previous studies on PP [9].

Hence, for the accurate calculation of viscosity data, the combined effects of pressure on both polymer melt flowability and polymer melt temperature as well as shear heating effects must be taken into account.

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For further information on the Pro4Plast results or support please contact the Polish Plastics Converters Association (www.pspts.com.pl) being Pro4Plast national contact point in Poland.

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