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Melt Rheology of High-Density Polyethylene (HDPE) and Polypropylene (PP) using Single-Screw Extruder

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Melt rheological properties of HDPE and PP were studied using single-screw extruder equipped by capillary rheometer head. The rheological experiments were carried out at 210°C, 220°C, and 230°C. The screw rotational speed varied from 5 to 130 RPM. Power law index (n), consistency index (K), and flow activation energy (E) were determined. The results showed that n values at 210°C, 220°C, and 230°C for HDPE were 0.72, 0.73 and 0.76 respectively, and for PP were 0.40, 0.44, and 0.45. The consistency index (K) values for HDPE and PP decreased with the increased in temperature and the flow activation energy of PP was greater than that of HDPE.

Keywords: Melt rheology, extruder, Power law, HDPE, PP

1 Introduction

Polyolefins, and in particular polyethylene (PE) and polypropylene (PP), represent the most common class of synthetic polymers produced and used nowadays. This is due to their cost-effective of production, excellent chemical resistance and variety of mechanical characteristics. Despite the fact that polyolefins were first produced in the 1930's, significant advances in enhancing the process and performance of these materials are still being achieved, and their use is growing. [1, 2]. In order to produce these materials and polymers in general, extrusion is essential. To melt, mix, and process polymers, nearly all polymer processing operations require an extruder. Therefore, it is necessary to achieve a better understanding of polymer rheology which can lead to improve polymer processing operation or to achive optimal mechanical design of the extruder [3]. Characterization of rheological properties of polymers are important to provide the information for designing of processing equipment, application properties and maintain the quality and consistency of the products. Therefore, understanding of polymer rheology is the key in polymer processing operations and without a significance of understanding of rheology, it would be challenging to develop the materials and processing techniques [3, 4]. The flow behavior of thermoplastic polymer melts is influenced by processing parameters such as temperature and pressure, and the relations of polymers' properties and processing are of paramount importance in all of their industrial applications [3, 4]. Rheological measurements can be performed by using different instruments such as rotational and capillary rheometers, but no attention has been given in the past to rheological characterization of PP and and PE by using an extruder and there is no published information regarding there rheological properties by using extruder which can be used to investigate the rheological behaviour of polymer melts in an industrial scale. By extrusion process, the highest shear rates can be achived, which correspond to operation conditions of industry devices. Hence, the range of shear rates values are extended for application of industry devices design. Singh et al. [5] studied melt rheological properties of high-density polyethylene (HDPE)-based nanocomposites by using a capillary rheometer. Power law exponent and consistency index were evaluated at the shear rate range of 50–1000 s^{-1} at three different temperatures 190°C, 200°C and 210°C. It was observed that n values for HDPE at 190°C, 200°C and 210°C for HDPE were 0.37, 0.40 and 0.45 respectively, which indicates shear thinning behavior of HDPE. The consistency coefficient K ($kPa.s^n$) values for HDPE at 190°C, 200°C and 210°C for HDPE were 1.38, 1.35 and 1.30 respectively. The value of K for HDPE decreased with increase in temperature and this result attributed to the decreased resistance to flow of the melts with increase in temperature. It was also found that the flow activation energy value for HDPE was 2.18 (kJ/mol). McNally et al. [6] prepared polypropylene and a metallocene catalysed ethylene–octene blends using a single-screw extruder and the flow properties of the blends were measured using a capillary rheometer over the shear rate range 300–4600 s^{-1} at 197, 217 and 237 °C. It was found that n values for PP at 197°C, 217°C and 237 °C for PP were 0.27, 0.31 and 0.27 respectively. The consistency coefficient value for PP at 197°C, 217°C and 237 °C were 2.09 $\times 10^4$, 1.35 $\times 10^4$ and 1.69 $\times 10^4$ ($Pa.s^n$) respectively. The value of flow activation energy for PP was 8.9 (kJ/mol). On the other hand, It was also found in the literatures that n and K values for HDPE at 180°C were 0.41 and 2.0 $\times 10^4$ (Pa.s^n) respectively, and for PP at 200°C were 0.38 and 7.5 $\times 10^3$ (Pa.s^n) respectively [7] without mentioned neither the method that used to determine the mentioned values nor the shear rate range of the measurements.

The aim of this work is to characterize the rheological properties of HDPE and PP polymer melts using a laboratory scale single screw extruder. Such works are very important to understand and optimize an extrusion process conditions of polymer melts.

2 Materials

The materials used in this study were polypropylene homopolymer (PP); Mosten GB 107 (supplied by ORLEN Unipetrol a. s., Czech Reublic) and polyethylene homopolymer (PE-HD); Liten MB 61 (supplied by ORLEN Unipetrol a. s., Czech Reublic). Table 1 lists PP and HDPE Properties. Density of the melt was measured by MODULAR MELT FLOW 7026 (Instron, USA) with respect to ISO 1133 and ASTM D-1238, for temperature 220 °C [8]. The deviation of the density melts for temperatures 210 and 230 °C is insignificant. The MFI values are identified from the data sheets of polymer producer.

***Tab. 1** Properties of tested polymers*

Polymer/Lot	Melt density (kg/m³)	Melt flow index MFI (g/10 min)
PP/ Mosten GB 107	741.6±5.4	7.0 (230 °C, 2.16 kg)
PE-HD/ Liten MB 61	743.6±2.8	7.5 (230 °C, 2.16 kg)

3 Apparatus and Methodology

Rheological properties of the melts were studied using a laboratory extruder (EB-L30, manufactured by BOCO PARDUBICE machines, Czech Republic). Table 2 lists the extruder properties provided by the supplier.

***Tab. 2** Properties of used extruder*

Extruder performance	2-10 kg/h
Screw diameter	30 mm
Working length of auger	30 L/D
Total power consumption	9 kW
Main engine power	7.5 kW

The extruder has five heating zones equipped with temperature sensors and the last 3 heating zones which located right before the die head of the extruder are equipped with pressure sensors. The extruder is equipped with capillary head. The capillary has dimentsion W =20 mm, h = 1 mm (aspect ratio is 0.05) and the length of the capillary is C = 155 mm. Two pressure transducers MDT 460F (Dynisco, USA) are installed in the capillary. The rheological experiments were carried out for various operating parameters of the extruder. The process temperatures were 210°C, 220°C, and 230°C. The screw rotational speed varied from 5 to 130 RPM. The measurements were performed with increasing and descresing rotational speed of srew. The presented values are average values for same rotational speed. For each rotational speed of the screw during the experiments, two samples were cut to measure the weight after the polymer melt was extruded through the extruder capillary. After all measurements, calculations of the parameters can be done according to the following equations [9]:

$$\dot{Q} = \frac{\dot{m}}{\rho} \ [\mathrm{m^3.s^{-1}}], \qquad (1)$$

Where:
$\dot{Q}$…The volumetric flow rate (m³/s),
$\dot{m}$…The mass flow (kg/s),
ρ…The melt density (kg/m³).

The apparent shear stress (τ_a) is given as following:

$$\tau_a = \frac{\Delta P h}{2L} [\mathrm{Pa}], \qquad (2)$$

Where:
ΔP…The measured pressure drop (Pa),
h…Capillary height (0.001 m),
L…The distance between pressure transducers (0.13 m).

The apparent shear rate (γ_a) is given as following:

$$\dot{\gamma}_a = \frac{6\dot{Q}}{Wh^2} \ [1/\mathrm{s}], \qquad (3)$$

Where:
W…The capillary width (0.02 m).

Equation (3) can be written as Rabinowitsch correction:

$$\dot{\gamma}_a = \frac{2\dot{Q}}{Wh^2}\left(2 + \frac{1}{n}\right) \ [1/\mathrm{s}], \qquad (4)$$

Where:
n…The power-law index,
n=1 for Newtonian fluids.

The apparent shear viscosity (η_a) is given by:

$$\eta_a = \frac{\tau_a}{\dot{\gamma}_a} \text{ [Pa.s]}, \tag{5}$$

The flow activation energy values of HDPE and PP were determined by using Arrhenius equation:

$$\eta_a = Ae^{\frac{E}{RT}} \text{ [Pa.s]}, \tag{6}$$

Where:
A (Pa.s)…Constant related to polymer structure,
R…The gas constant (8.314 J/(mol K)),
E…The flow activation energy (J/mol),
T…Absolute temperature (K).

4 Results and discussion

4.1 Flow curves

Figures 1 and 2 show the uncorrected rheomgrams (shear stress versus shear rate) of HDPE and PP melts respectively at 210°C, 220°C, and 230°C. To describe the shear stress as a function of shear rate, power law model was applied:

$$\tau_a = K\dot{\gamma}^n \text{[Pa]}, \tag{7}$$

Where:
n…The power-law index,
K…The consistency index.

The corrected shear rate can be obtained from the apparent shear rate values by using Rabinowitsch correction. Figures 3 and 4 show the corrected flow curves of HDPE and PP melts at 210°C, 220°C, and 230°C.

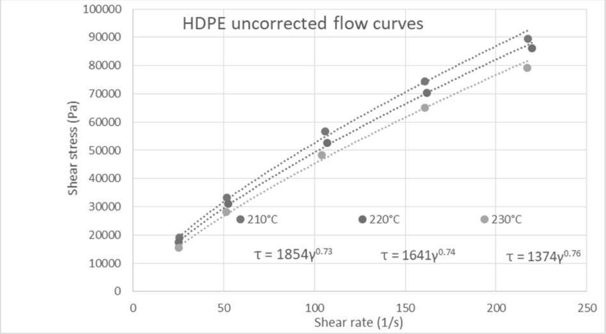

Fig. 1 *Uncorrected flow curves of HDPE melt at 210 °C, 220 °C, and 230 °C*

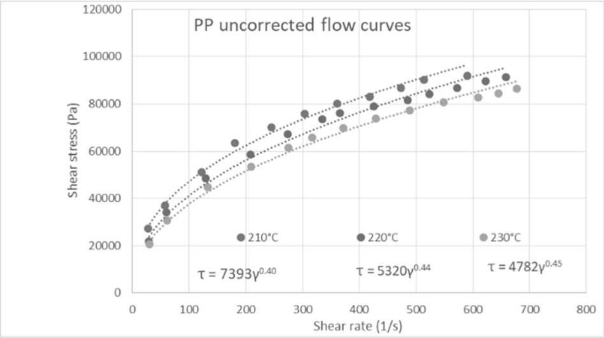

Fig. 2 *Uncorrected flow curves of HDPE melt at 210 °C, 220 °C, and 230 °C*

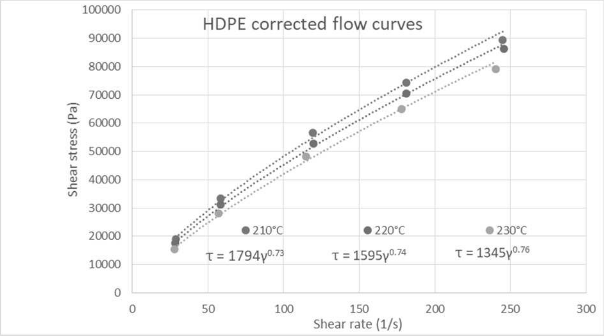

Fig. 3 *Corrected flow curves of HDPE melt at 210 °C, 220 °C, and 230 °C*

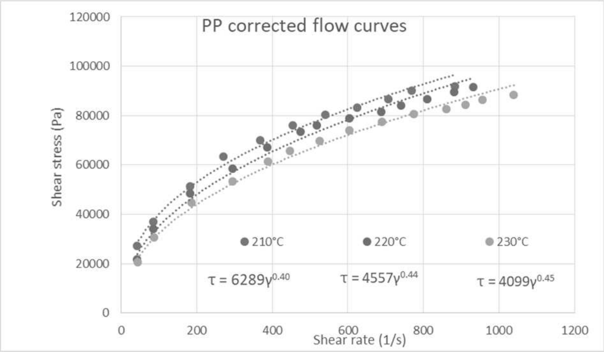

Fig. 4 *Corrected flow curves of PP melt at 210 °C, 220 °C, and 230 °C*

The values of n and K for HDPE and PP are listed in table 3.

Tab. 3 *Flow behavior index (n) and consistency index (K) of HDPE and PP melts*

Polymer	n			K (Pa.s^n)		
	210°C	220°C	230°C	210°C	220°C	230°C
HDPE	0.73±0.02	0.74±0.01	0.76±0.02	1794±1	1595±1	1345±1
PP	0.40±0.01	0.44±0.01	0.45±0.01	6289±1	4557±1	4099±1

As shown in table 3, the flow behavior index (n) values are found to be less than 1 which indicating that HDPE and PP are pseudo plastic fluids over the range of the studied shear rates. n values for HDPE are higher than that for PP which means that HDPE has less viscosity–sensitivity to shear rate where the value of n represents the deviation from the Newtonian fluids. The consistency index (K) values decreased with the increased in temperature. The consistency index value reflects the viscosity of the polymer at unit rate of shear [10]. With increasing temperature, the viscosity of the polymer decreases which leads to decrease K-value. Similar behavior has been reported in literature [5].

4.2 Viscosity curves

The plots of corrected viscosity versus shear rate of HDPE and PP at different temperatures are shown in figures 5 and 6.

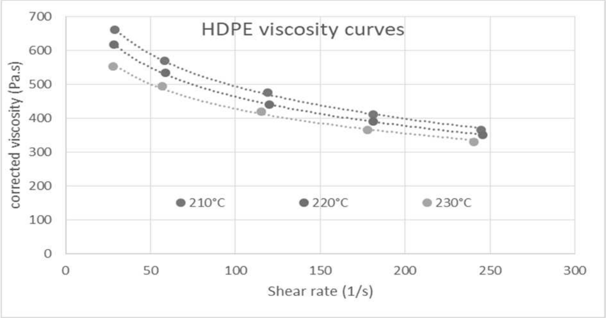

Fig. 5 *Corrected viscosity versus shear rate of HDPE melt at 210 °C, 220 °C, and 230 °C*

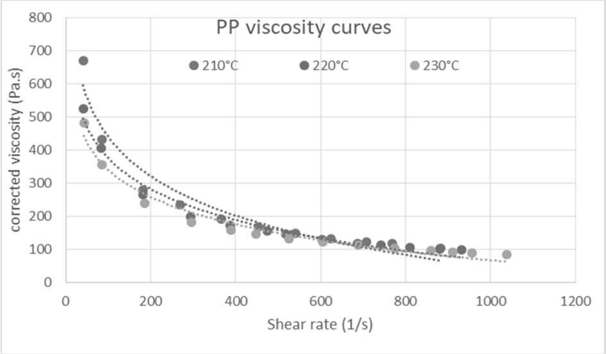

Fig. 6 *Corrected viscosity versus shear rate of PP melt at 210 °C, 220 °C, and 230 °C*

The viscosity of HDPE and PP decreased with increasing shear rate, showing a shear thinning behaviour. It also appeared that with increasing shear rate, the influence of shear rate on the viscosity of HDPE and PP reduced and the results showed that the shear rate has no significant influence on PP viscosity at the shear rate range of 600–1000 s^{-1}.

4.3 Flow activation energy

The flow activation energy and constant A values of HDPE and PP were calculated from the slope of log corrected viscosity versus 1/T plots. The plots of log corrected viscosity versus 1/T for HDPE and PP are shown in in figures 7 and 8 respectively.

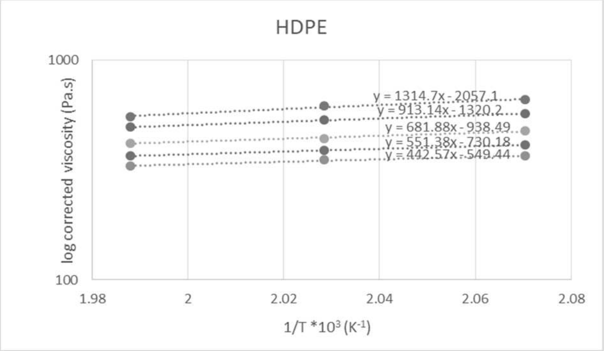

Fig. 7 *Plots of log corrected viscosity versus 1/T for HDPE melt*

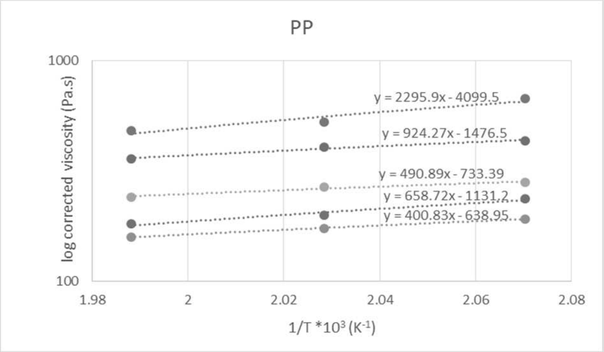

Fig. 8 *Plots of log corrected viscosity versus 1/T for HDPE melt*

The values of flow activation energy of HDPE and PP melts at different viscosity ranges are listed in table 4.

Tab. 4 *Fow activation energy (E) and constant A of of HDPE and PP melts*

Polymer	Viscosity range (Pa.s)	E (kJ/mol)	Constant A (Pa.s)
HDPE	553-662	10.93	-2057
	495-570	7.59	-1320
	420-476	5.67	-938
	366-411	4.58	-730
	329-366	3.68	-549
PP	482-670	19.09	-4100
	357-433	7.68	-1477
	240-281	4.08	-733
	181-235	5.48	-1131
	158-191	3.33	-638

Trends of activation energy indicate that the flow behavior of PP is more temperature –sensitivity compared with HDPE. Comparing with the literature, the difference in the obtained n, K, and E values could be attributed to the difference in the measurement methods and conditions such as pressure, temperature and variety of used shear rates range.

The relationship between activation energy and shear rate for HDPE and PP are shown in figures 9 and 10 respectively.

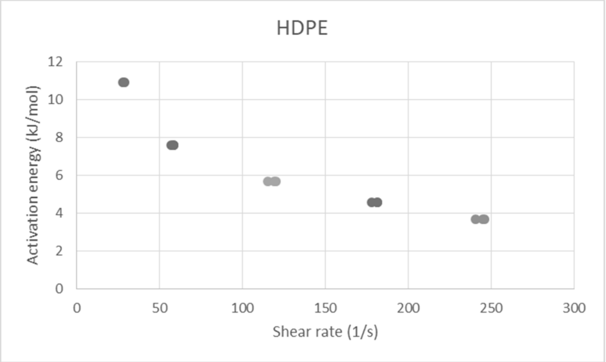

Fig. 9 *Plots of activation energy versus shear rate for HDPE melt*

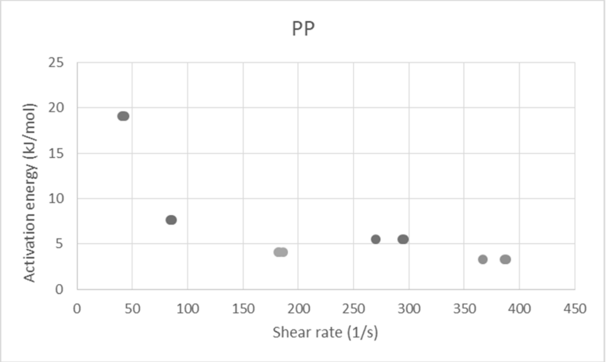

Fig. 10 *Plots of activation energy versus shear rate for PP melt*

It could be noted that the activation energy for HDPE and PP decreases with increasing shear rate. On the other hand, at a given shear rate (270-295 s^{-1}), the activation energy of PP slightly increased, which can be explained due to error measurement. Similar behaviour of decreasing activation energy with increasing shear rate was also observed in literatures [11, 12].

5 Conclusion

In this paper, melt rheological behavior of HDPE and PP were investigated using a single-screw extruder equipped with capillary head. Non-newtonian pure viscous Power law model was used to describe the relationship between shear stress and shear rate. Flow index (n) values for HDPE and PP are found to be less than 1 and the consistency index (K) values decreased with increasing temperature. Both polymer melts exhibit shear thinning behaviour, where PP has stronger Non-newtonian effect. The flow activation energy of PP found to be greater than that of HDPE. However literature data contain enormous rheological data for HDPE and homopolymer PP, the polymers are different by its composion and distribution of macromolecules. Therefore it is highly important to asses the rheological properties of specific polymer to get the proper data for process optimization of polymer treatment.

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