

Instable Melt Extrusion of Polypropylene through Capillary

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Abstract

One polypropylene melt was extruded through capillary rheometer with several die diameters for testing its shear viscosity at different shear rates and temperatures when the extrudate distortion was observed. Under certain conditions, the melt exhibits shear thinning in viscosity with shear rate. The die with greater aspect ratio of length-to-diameter allows the melt to relax more being embodied by decrease shear viscosity. In capillary flow, the applied shear orients, uncoils or disentangles molecular chains to store energy for elasticity. At 180 °C, the extrudate swells and distorts spirally until random fracture. The distortion and the swelling can be weakened by using the die with larger aspect ratio or by increasing temperature due to chain relaxation.

Keywords: Instable melt extrusion; Capillary rheometer; Polypropylene melt; Distortion; Swell; Viscosity

Introduction

Polypropylene (PP) is a kind of general synthetic thermoplastic resin with the characteristics of non-toxic, tasteless, low density, good rigidity, impact resistance, flexure resistance and chemical corrosion resistance. It is widely used in civil and industrial and engineering's such as packaging, coatings and building and parts of electronic devices [1]. There are two types of factors that affect the viscosity of polypropylene. The internal ones include molecular structure, molecular weight and its distribution as well as branching characteristics. The external ones involve such processing parameters as shear rate and temperature as well as geometry of flow field [2,3]. PP melt is non-Newtonian fluid [4]. Its flow ability can be tested by capillary rheometer, from which the plug pressure versus its velocity is converted into shear stress versus strain rate, further viscosity is calculated using different constitutive models reflecting the influences of shear rate and temperature as well as die geometry of diameter and length [5]. The ratio of diameter to length is also called aspect ratio. Besides, the extrudate surface and sizes are also observed reflecting the extrusion characteristics. The surface gradually appears smooth, rough, sharkskin, spiral, oscillated and fractured in extrusion instabilities with the increase of shear rate [5-7]. The ratio of extrudate diameter to die inner diameter is usually larger than one, which is termed as swell ratio. The swelling degree and the surface roughness are embodied of melt elasticity [6]. Die swell as one characteristic elasticity is the functions of material parameters, flow field characteristics, the reservoir-capillary entry effect and the outside swell environment.

When molten polymer flows through a capillary, molecular chains become oriented, uncoiled or disentangled due to the applied shear. The entanglements will, to some extent, prevent the molecules from slipping past one another, thus preventing total relaxation of the molecules. As melt leaves the die, molecular chains tend to recoil in the flow direction and grow in the normal direction, leading to extrudate swell. The mechanism and degree of swelling are usually explained in terms of elastic recovery or effect of residence time on the applied stresses. The phenomenal [6] or physical models [7] have been developed for showing the rules in addition to texture checks [8]. Such analyses are important to study the relationship between rheological properties of polymer melt and processing conditions for the determination of process parameters and the improvement of plastic forming quality. Here, the capillary is used to analyze the effects of temperature and die geometry on viscosity and extrudate surface.

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Test Materials and Methods

Materials

Polypropylene (PP 4220) with melt mass flow rate of 0.92-0.94g/10min and density 0.963g/cm³ was from Yanshan Petrochemical Co., Ltd.

Test method

One capillary rheometer was used at constant pressure. The die length is of 5,10 or 20mm while diameter is 1, 1.5 or 2mm. Process temperature was 180, 190, 200, 210 or 220 °C. The shear rate was set as 10, 50, 100, 300 or 500s⁻¹.

Results and Analysis

Effect of length-diameter ratio of die on shear viscosity of polypropylene

(Figure 1) presents the shear viscosity versus shear rate curve of PP melt through dies of different length-diameter ratio at 180 °C for capillary barrel under constant pressure. The shear viscosity decreases with the increase of shear rate, showing PP melt being pseudoplastic fluid. Under shear against barrel wall, the macromolecular chains are continuously oriented and disentangled along the flow direction. Besides, the shear viscosity decreases with the increase of die aspect ratio due to the greater content of disentanglement and relaxation. The extrudates from three die ratios at 180 °C are shown in Figure 2. For each die ratio, four specimens are obtained at 10, 50, 100, 300 and 500s⁻¹ respectively. Only one smooth specimen is obtained for 5:2 die at 10s⁻¹. All of other specimens are spiral. The helix pitch turns greater when the melt were extruded at higher shear due to a greater elasticity. The similar tendency in pitch occurs in longer die of the same diameter of 2mm when the roughness becomes smaller due to a greater relaxation. The distortion results from the stored elasticity at entrance and shear orientation. However, the depth of spiral groove is not monotone function of length-diameter ratio in Figure 2. It is reduced when the inner diameter or when the length of die is increased as listed in Table 1. Table 2 shows the swell ratio changes with die sizes. It is reduced when the length of die is increased

while it is increased when the inner diameter of die is increased as listed in Table 1. The former case is due to the longer residence time of melt in the die for elastic recovery, i.e. relaxation. The latter case is due to easier elastic recovery outside the die of larger diameter. This tendency becomes weaker when the die length is longer because a greater content elastic deformation at entrance can be relaxed in longer die.

Table 1: The depth (mm) of spiral groove on the extrudate with different dies.

| Diameter D/Length L | 1mm | 1.5mm | 2mm |
|---------------------|---------|----------|----------|
| 5mm | 0.4-0.7 | 0.8-1.59 | 1.05-2.0 |
| 10mm | | 0.45-1.4 | 0.9-1.66 |
| 20mm | 0.2-0.5 | 0.3-0.8 | 0.65-1.1 |

Table 2: The extrudate swell ratio for dies with different sizes at the same shear rate.

| Diameter D/Length L | 1mm | 1.5mm | 2mm |
|---------------------|------|-------|-------|
| 5mm | 1.84 | 1.87 | 1.925 |
| 10mm | | 1.77 | 1.85 |
| 20mm | 1.6 | 1.66 | 1.7 |

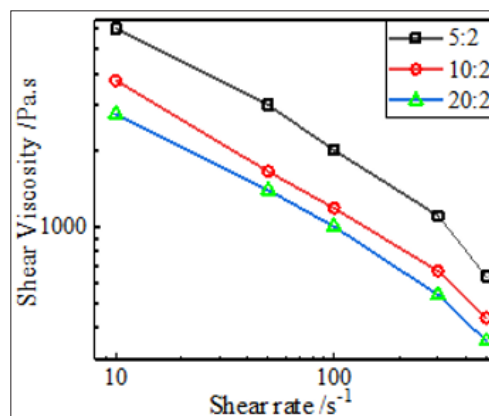


Figure 1: Shear viscosity versus strain rate for dies with different length-diameter ratios.

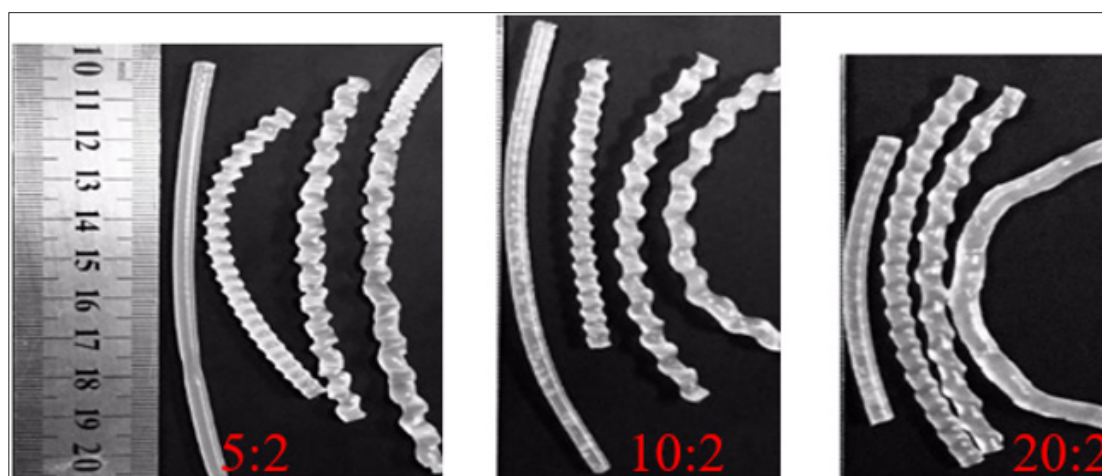


Figure 2: The extrudates under four shear rates with three die length-diameter ratios.

The effects of temperature on shear viscosity of polypropylene

Figure 3a shows the shear viscosity versus shear rate of PP melt at different temperatures under constant pressure and die length diameter ratio of 5:2. At the same temperature, the shear thinning is preliminary. Under the same shear rate, the viscosity gradually decreases with the increase of temperature. Higher temperature allows molecular chains to deform and relax faster. Figure 3 shows the influence of temperature and die length-diameter ratio on the swell ratio. For a certain die, the higher the temperature is, the smaller the extrusion swell ratio is. The movement ability of PP molecular chain increases at higher temperature corresponding to faster relaxation and smaller orientation. At the same temperature,

the swell ratio drops with length-diameter ratio showing more relaxation, resulting in the lower resident elasticity and higher viscous dissipation of deformation energy stored in the melt. The extrudates exhibit differently as illustrated in Figure 4 when the melt is extruded from die of 5mm in length and of 2mm in diameter. All specimens extruded at different are smooth at low shear rate 10s^{-1} . The extrudates become smoother when increasing barrel temperature at the same shear rate. The distortion is strengthened in stronger shear at the same temperature, the shape turns from spiral state to sharkskin state. More importantly, the smooth surface is obtained for four shear rates when temperature is not below $200\text{ }^{\circ}\text{C}$. With the increase of temperature, the extrusion distortion is reduced. When the temperature is higher than $210\text{ }^{\circ}\text{C}$, the distortion phenomenon has disappeared completely.

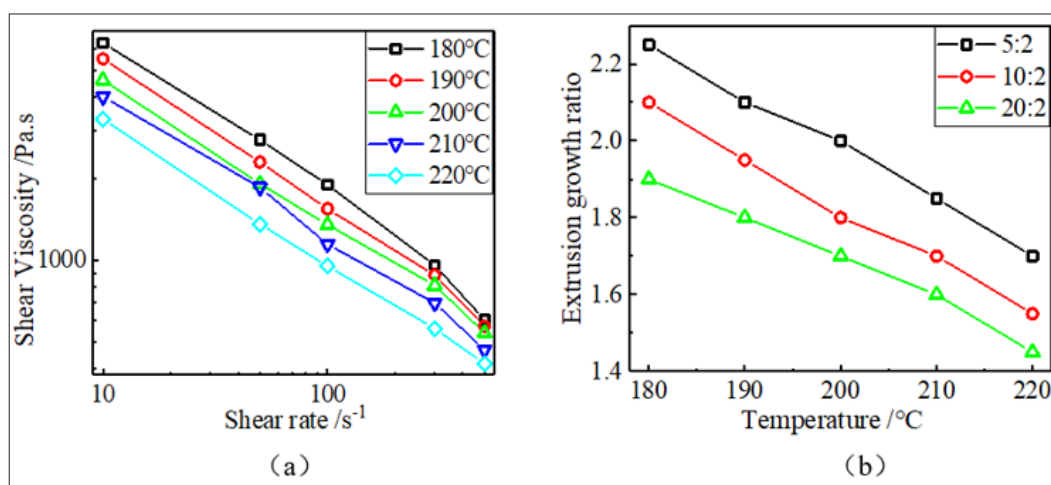


Figure 3: The viscosity curves versus shear rate in increasing temperature (a) and swell ratio versus temperature for dies with different aspect ratios. (b) shows the influence of temperature and die length-diameter ratio on the swell ratio.

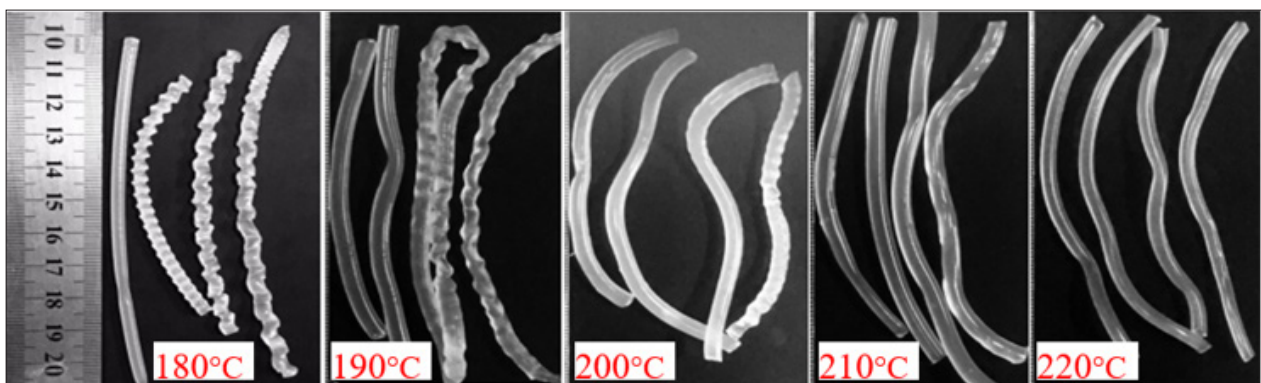


Figure 4: The extrudates from die of 5:2 for barrels of different temperatures at four shear rates.

Conclusion

- The melt shear viscosity decreases increasing shear rate or die aspect ratio under the same other conditions.
- The viscosity decreases with the increase of temperature under the same other conditions.
- At $180\text{ }^{\circ}\text{C}$, the extrudate became rougher in spiral from smooth surface until random fracture after being distorted. The distortion and die swell ratio decreases with the increase of die aspect ratio.
- The swell ratio and extrudate distortion drop when increasing temperature.

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