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SHORT COMMUNICATIONS

Magnetic Effects on Extrudate Swell of a Polystyrene Melt in Capillary Extrusion Dies

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The extrudate swell occurs in any viscoelastic fluids including polymer melts, and is an important issue in plastic processing. The mechanism of the extrudate swell is usually explained by the release of the elastic energy, at the end of the die, stored due to the deformation in the die. The extrudate swell is one of the main factors that determine the quality and dimension of the polymer product. Understanding of this phenomenon is therefore necessary to design and optimize the polymer processing units such as screws, dies and sizing units. In general, the extrudate swell ratio increases with decrease in the die length and the temperature, and it increases with increase in the shear rate and molecular weight of polymers. In the actual polymer processing, the extrudate swell ratio is controlled by adjusting the extruder screw speed and the barrel and die temperature.

A number of theoretical, computational, and experimental studies[1–10] have been reported on extrudate swell. It was found that the swell was slightly affected by the change in the melt temperature and flow rate but it was strongly affected by the molecular weight distribution.[1] The effect of the die entrance angle and the die length on the extrudate swell in the capillary rheometer was studied.[2] The proposed calculation was based on the analysis of rate and stress corresponding to the elongation and shear flow components. The use of ultrasonic irradiation during extrusion was reported. It was found that appropriate irradiation intensity reduced the extrudate swell ratio, as well as improvement of the quality of the extrudate.[3]

The use of magnetic fields to the polymer processing has been paid attention recently.[4,8,10] The magnetic fields were found to act on feeble magnetic materials including polymers through the magnetic force and the magnetic torque.[11] It is well known that the liquid crystalline polymers undergo magnetic alignment. The alignment is due to the magnetic torque acting on the anisotropic 'domains' in these polymers. In view of anisotropic 'domains', polymer melts under flow have a similarity to liquid crystalline polymers. Both of them possess the oriented regions that could be affected by the magnetic field. In fact, the magnetic effect on extrudate swell of polystyrene (PS) melt has been reported by one of the authors (N.S.).[4,8]

In this paper, we report the magnetic effects on extrudate swell of PS melt studied under a constant shear rate capillary rheometer similar to the system of the previous work by Sombatsompop.[4]

EXPERIMENTAL

Materials

The polymer used in this work was polystyrene with a melt flow index (MFI) of 2.2 and a density of 1.05 g/cm^3 (ISO/JIS113K7210, ISO/JIS1183K7112) supplied by PS Japan Co. Ltd. (Tokyo, Japan).

Experimental Setup

Figure 1 shows the experimental setup, which was composed of a barrel, a piston, a capillary die, the magnetic source (a ring magnet) and the control system. The barrel was made of 304 stainless, 30 mm

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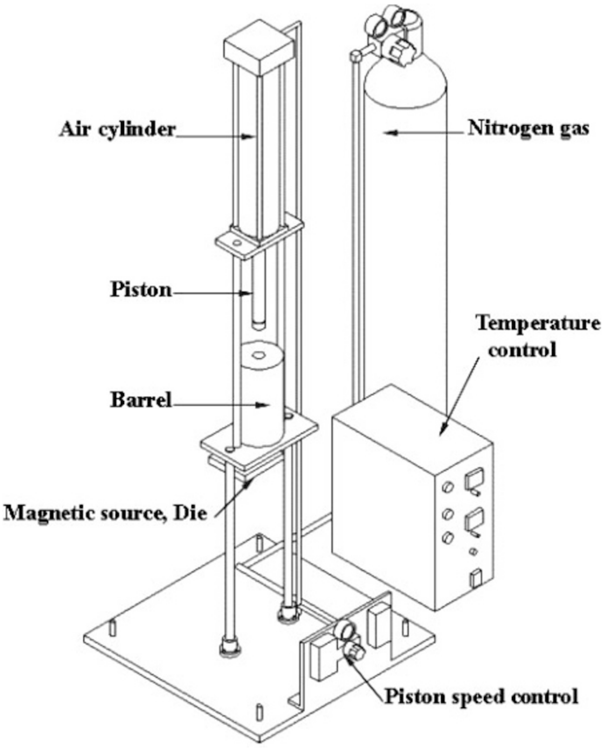

Figure 1. Schematic representation of apparatus used for extrudate swell measurements.

in diameter and 230 mm in height. The pellets of the sample polystyrene were melted in the barrel under temperature control. The piston was divided into two parts: the piston body and the piston tip. The piston body was made of stainless steel (grade 304) while the piston tip was made of copper. The piston was attached to the air cylinder mounted on the top of the apparatus. Capillary dies made of two different materials, steel and stainless (grade 304), were used. The length and the diameter of the capillary were 86 and 6 mm, respectively. The die body was wrapped with the heater coil. The capillary die was connected to the bottom of the barrel. Magnetic field was applied using a ring magnet as shown in Figure 2. The magnetic field direction generated by the ring magnet was parallel to the flow direction. The magnetic field intensity of the magnetic source was *ca.* 0.54 T on the surface. The operation control system was composed of the piston speed controller and the temperature controller (a KT Series 4 Temperature Controller, Matsushita Electric Works Ltd.). The temperature was controlled by two heaters, one located in the barrel and the other located around the die body in order to insure the same temperature at the barrel and the die. The magnet was thermally insulated by placing glass wool between the heater and the magnet. The piston speed was controlled by the pressurized nitrogen gas. The shear rate was determined using the speed of the piston movement.

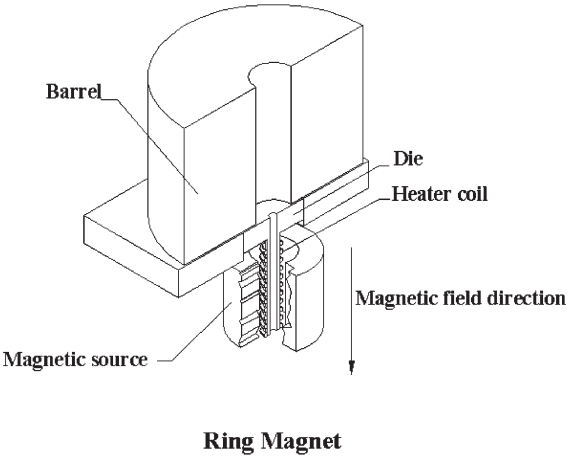

Figure 2. The layout of the permanent ring magnet near the die.

Experimental Procedure

All measurements were performed with and without the application of the magnetic field to the steel and stainless capillary dies. The piston speed was varied from 2 to 24 mm/min, which corresponds to shear rates of 1 to 15 s^{-1}, respectively. The die temperatures were varied from 190 to 230 °C. At a given sample temperature, a series of shear rates were applied consecutively from low to high under a magnetic field generated by a ring magnet. At each shear rate, the extrudate coming out from the die end was chopped off by a pair of scissors to obtain several solidified rods of *ca.* 25 mm length. Then, the same procedure was repeated after the magnet was removed. Similar procedures were carried out at different sample temperatures. The diameters of the chopped and solidified specimen was measured with micrometer at several different positions to determine the extrudate swell ratio defined as diameter of the extrudate/diameter of the die (6 mm). Three experiments were carried out for the same experimental condition (at a given temperature with a given magnetic configuration with or without the field exposure).

RESULTS AND DISCUSSION

Figure 3 shows the extrudate swell ratio as a function of the shear rate measured at 190 °C. Three sets of data obtained under the same experimental condition are enclosed and identified by groups (a), (b), (c), and (d). These four groups correspond to the experiments with steel die [(a) and (b)], where (b) without and (a) with magnetic field, and with stainless die [(c) and (d)], where (c) without and (d) with magnetic field. The data are scattered but the groups for the experiment with the magnetic field [(b) and (c)] are clearly separated from the groups for the experiment

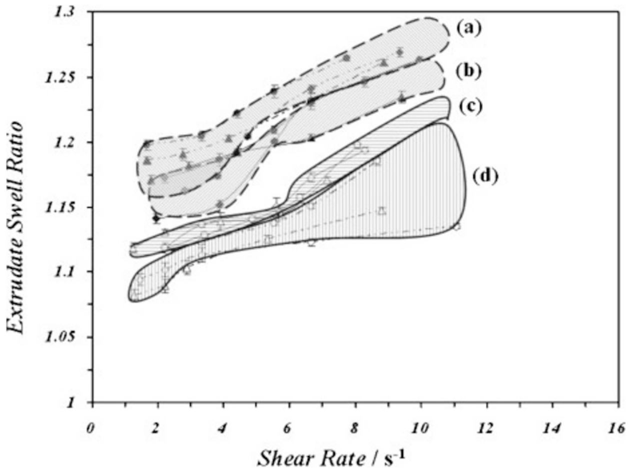

Figure 3. Extrudate swell ratios measured at 190 °C plotted against the shear rate. Groups (a) to (d) enclosing three experimental data indicate the experiments with steel die [(a) with and (b) without magnetic field] and the stainless die [(d) with and (c) without magnetic field].

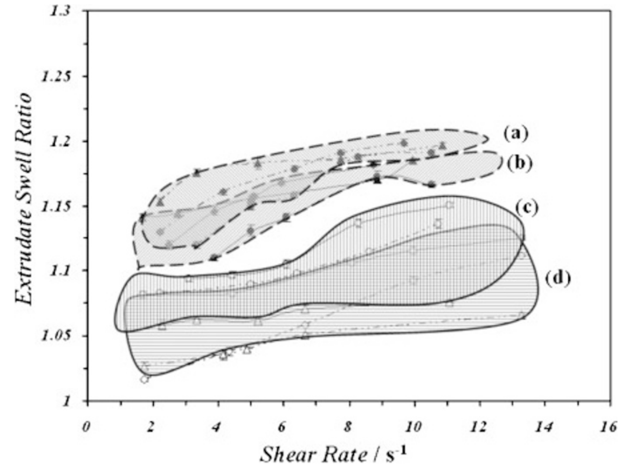

Figure 4. Extrudate swell ratios measured at 210 °C plotted against the shear rate. Groups (a) to (d) enclosing three experimental data indicate the experiments with steel die [(a) with and (b) without magnetic field] and the stainless die [(d) with and (c) without magnetic field].

without the magnetic field [(a) and (d)], respectively. There are overlaps between the groups, but the effect of the magnetic field is clearly observed. Figures 4 and 5 show the results for the measurements at 210 and 230 °C, respectively. Here again, the effect of the magnetic field is evident.

The extrudate swell ratio increased with increase in the shear rate because the shear rate was a direct function of the shear stress generated during the flow, and the extrudate swell is generally associated with the storage elastic stress (shear stress), which is released on exit from the die. The decrease in the extrudate swell with increase in die temperature was associated with reductions of melt elasticity and melt viscosity.

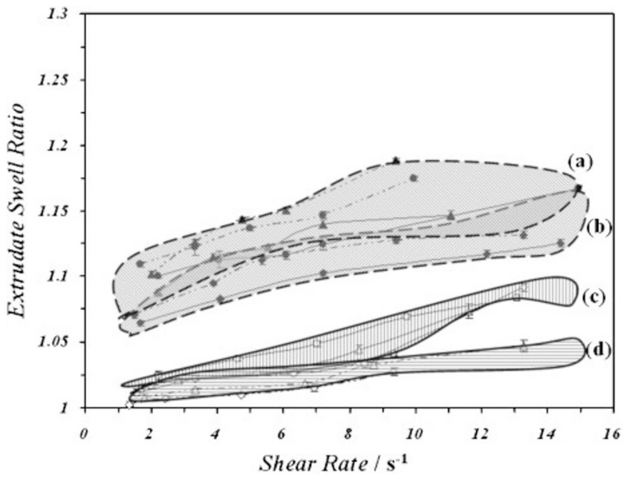

Figure 5. Extrudate swell ratios measured at 230 °C plotted against the shear rate. Groups (a) to (d) enclosing three experimental data indicate the experiments with steel die [(a) with and (b) without magnetic field] and the stainless die [(d) with and (c) without magnetic field].

Regarding the measurements without the magnetic field, we find that the steel die exhibits larger extrudate swell than the stainless die by *ca.* 8% at any temperatures. This difference could occur because the drag friction between the melt layer and the inner die surface is different depending on the material of the die though the detailed surface analysis was not carried out for the dies used in the present study. The steel die surface might cause more friction leading to higher shear rates and resultant higher extrudate swell ratios than the stainless one.

In Figure 3, we find that the use of the steel die causes the increase in the extrudate swell upon application of the magnetic field, while the use of the stainless die causes the decrease. The average direction of the magnetic field produced by the ring magnet is parallel to the flow direction irrespective of the dies used. However, the detailed field profile within the die may differ depending on the materials of the dies. In the case of the steel die, the magnetic flux is condensed in the die body to generate a field gradient over the inner surface of the capillary and the resultant force acting on the flow from the inner surface. This gradient is especially enhanced on the edge of the die, creating additional forces acting on the flow. No such effect occurs in the stainless die because no flux condensation occurs. These forces occurring when the steel die is used, however, may not be strong enough to bring about the swell behavior opposite to the stainless die because the field intensity used in the present study is not very high. As discussed previously, the flow profile could be different between the steel and stainless dies. This might cause the difference in the resultant extrudate swell when the magnetic field is applied because the flow profile is an important factor that determines the anisotropic magnetic energy. All these are possible factors that could explain the different extrudate swell behavior between the steel and stainless dies, but we cannot draw the definite conclusion at present time.

Because the magnetic field used here is not strong enough to deform a random coil, the magnetic effect observed here should be mainly attributed to the magnetic anisotropy caused by the deformation of polymer chains due to the flow. Similar to the flow-induced birefringence, this magnetic anisotropy is proportional to the deformation γ, if the deformation is small. On the other hand, the elastic energy stored by the deformation is proportional to γ^2. Then, the energy stored in a unit volume of the polymer melt under the deformation γ in the presence of the magnetic field B is expressed as:

$$E = G\gamma^2/2 - (2\mu_0)^{-1}\chi_a^{\infty}\gamma B^2\cos^2\theta \qquad (1)$$

where G and μ_0 are the elastic modulus and the magnetic permeability of vacuum, respectively, χ_a^{∞} is the anisotropic diamagnetic susceptibility of the polymer chain at its full elongation, and θ is the angle between the elongation direction and the magnetic field. In the case of deformation of rubber, the deformation of the individual polymer chains between cross linking points is proportional to the applied macroscopic deformation, but in the case of the flow of the polymer melt at its flow regime, as is the case of the present study, the actual deformation of the chain is far smaller compared to the applied deformation because the relaxation occurs concomitantly.

Because the extrudate swell is due to the release of the deformation energy stored during the flow in the die, eq 1 is useful to understand and interpret the effect of the magnetic field on the extrudate swell. The actual elongation of the chain in the flow regime is small, then, the magnetic term in eq 1 could be comparable to the elastic term, affecting the flow and the resultant extrudate swell. The elongation direction in the flow differs from place to place inside the die depending on the flow profile. Therefore, the total energy, which is the summation of the local energy expressed in eq 1 over the whole flow in the die, could depend on the flow profile as well as the field profile.

CONCLUSIONS

Magnetic effects on the extrudate swell ratio of a polystyrene melt were studied. The swell ratios changed upon exposure to the magnetic field. The magnitude of the change was just slightly larger than the scattering of the data, but we believed that the difference was significant. The extrudate swell increased for a steel die and decreased for a stainless die (about 2–5%) under exposure to the magnetic field. The observed change was smaller than that reported previously by two of the authors (N.S. and N.I.). The reason was not clear at the present time. A preliminary interpretation of the observed phenomena was made in terms of the elastic energy and anisotropic magnetic energy stored by the deformation of the chains under flow. Further study is under way to clarify the effect of the magnetic field on the flow of the polymer melt using viscoelastic measurement under the magnetic field.

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