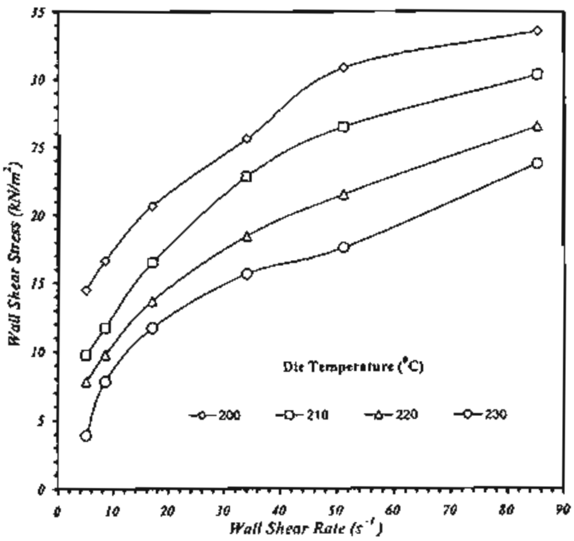

Figure 4. Wall shear stress and wall shear rate of PS melt for different die temperatures.

melt temperature during the flow along/in the barrel and die as a result of shear heating effect. The shear heating effect in the screw extruder is expected to be significant as a result of shearing action of the screw rotating in the barrel. It should also be noted that this shear heating effect was negligible in the case of a conventional capillary rheometer, as evidenced by previous work.[19] Another interesting point observed was that the swell ratio did not change so much with extrusion time in this test. Previous work by Sombatsompop and O-Charoen,[20] who measured the extrudate swell ratio of PS and linear low-density polyethylene (LLDPE) melts in a capillary rheometer, indicated that the swell ratio decreased as the extrusion time increased. The difference of these two results could be explained in terms of thermal history of the material; the thermal history of the material in this case was relatively shorter as the extrusion process was continuous. When comparing the overall swell result measured in the single screw extruder with that in the capillary rheometer,[21] it was found that for any given shear rate and die temperature, the overall swell ratio of the melt in the extruder was greater than that in the capillary rheometer. The discrepancies in the results were probably associated with the thermal histories of the melt in these two test equipments.

Figure 4 shows plots of wall shear stress and wall shear rate for different die temperatures, the results being obtained in the same test conditions as in Fig. 3(a)–3(d). For a given wall shear rate, the shear stress decreased with increasing die temperature. It is associated with the swelling reduction of the extrudate with increasing the die temperature, as mentioned earlier.

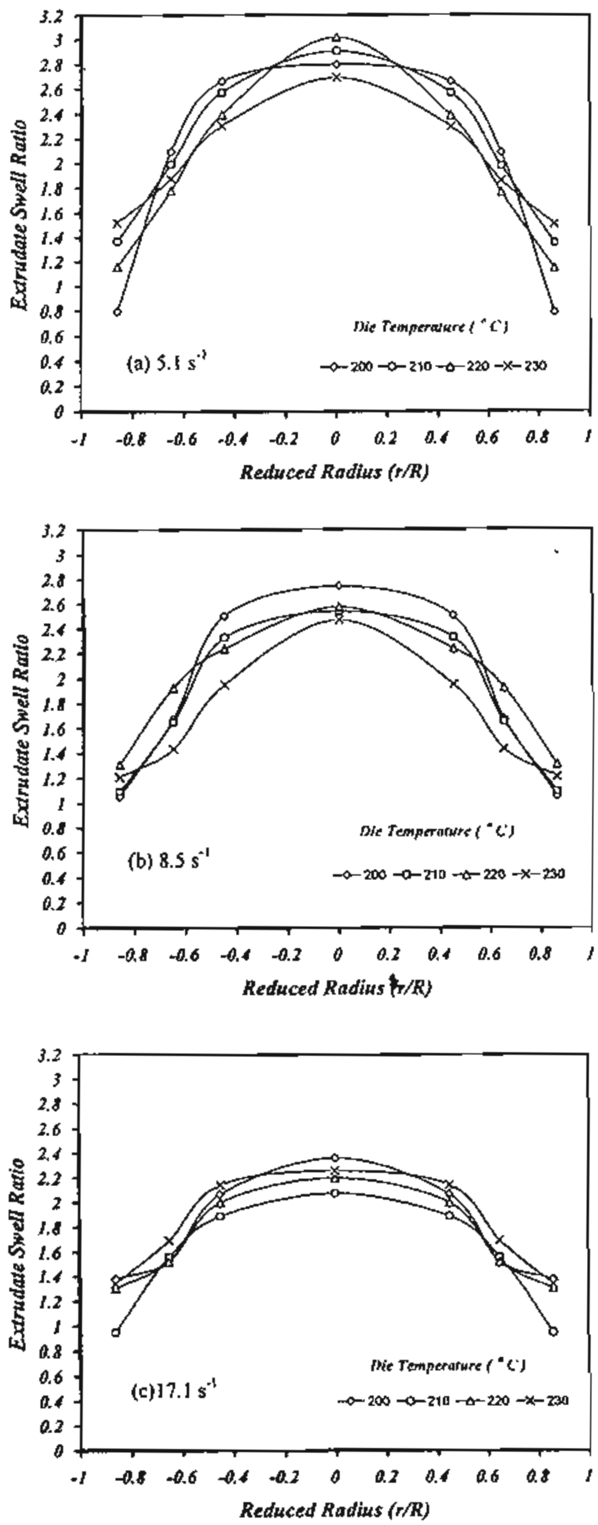

Figure 5. Radial extrudate swell profiles of PS melt at various shear rates and die temperatures: (a) 5.1 sec^{-1}; (b) 8.5 sec^{-1}; (c) 17.1 sec^{-1}.

Development of redial extrudate swell and velocity profiles

Figures 5(a)–5(c) shows the extrudate swell ratio as a function of the r/R position in the die for three wall shear rates. It was observed that, for a given shear rate, the extrudate swell ratio decreased with increasing r/R position. The highest swell ratio occurring at the duct center, was in the range 2.0–3.0 (100–200% extrudate expansion as compared to the die diameter). The value of extrudate swell ratio reached 0.9 around r/R position of 0.85. The swell ratio measured near the die wall implied occurrence of the extrudate contraction. These findings were in line with the work of Mitsoulis[14] and Kiselev and Kanavets.[15] The extrusion at higher shear rates produced flatter shape of the swell profiles. In other words, the differences in the swell ratio of the melt across the die diameter became less as the shear rate increased. When observing the extrudate swell around the centre of the die, it was found that the higher the shear rate, the greater the extrudate swell as expected. In most literatures, the extrudate swell is reported as being related to the stored elastic stress which is released on exit from the die. This stress is predominantly due to the shape of the velocity profile in the die which is believed to be parabolic in nature; the shearing stress (also shear rate) tends to be high at the die wall and low in the center of the flow.[22,23] Therefore, one would expect the extrudate swell across the die to be high at the wall and low at the center. However, it would not be always true, as evidenced by the results in Fig. 5(a)–5(c).

It was observed in the experiment of Fig. 5(a)–5(c) that the velocity profiles became sharper with increasing wall shear rate. The changes of extrudate swell ratio of the melt across the radial position of the die were observed through the velocity profiles of PS melt which were simultaneously measured in the experiments; the results of the melt velocities across radial positions of the die for different wall shear rates are shown in Fig. 6(a)–6(c). A general trend of the radial velocity profiles was very similar to that for the extrudate swell profiles. The highest melt velocity was found at the center of the die, the melt velocity decreasing with increasing r/R value due to the drag effect.[7] The shape of the radial extrudate swell profiles is considered to be associated with that of velocity profiles in the die, and the equalization of the velocity profiles at the die exit. That is, when the melt was flowing in the die it had a relatively high velocity at the center and relatively low near the die wall as can be expected in shear flow. On exiting the die, the velocities of the melt across

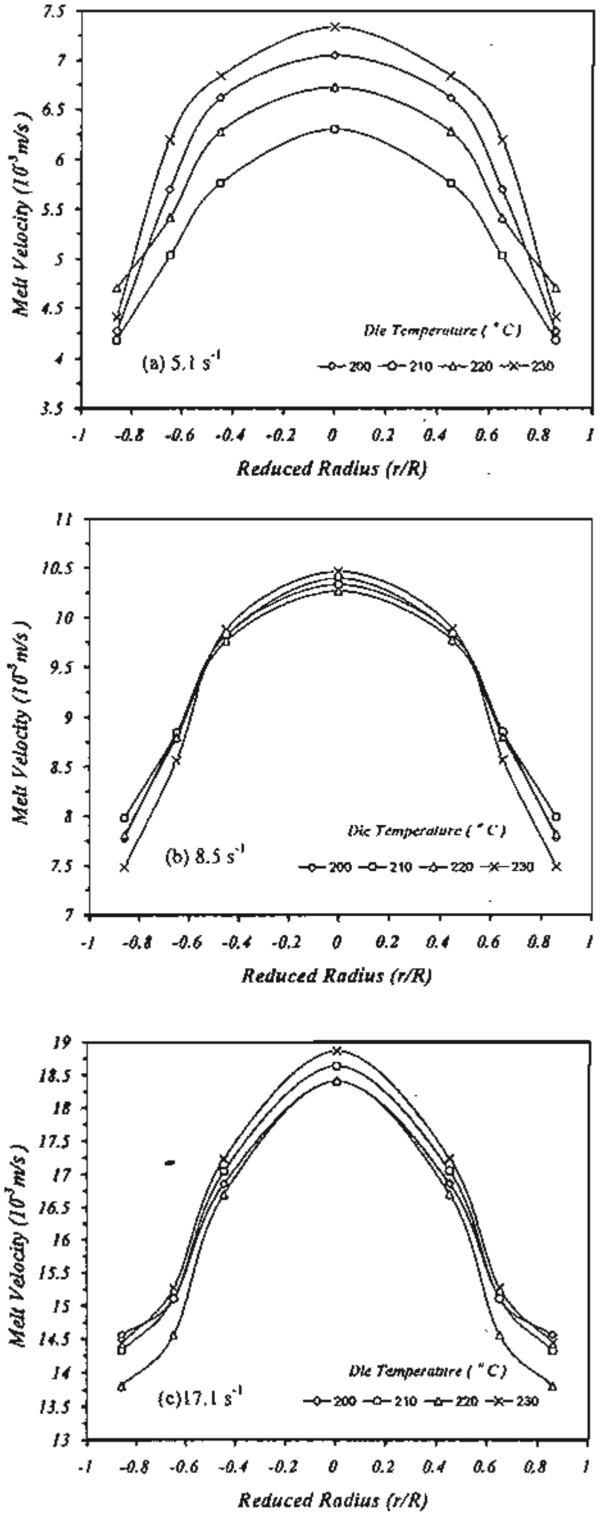

Figure 6. Radial velocity profiles of PS melt at various shear rates and die temperatures: (a) 5.1 sec^{-1}; (b) 8.5 sec^{-1}; (c) 17.1 sec^{-1}.

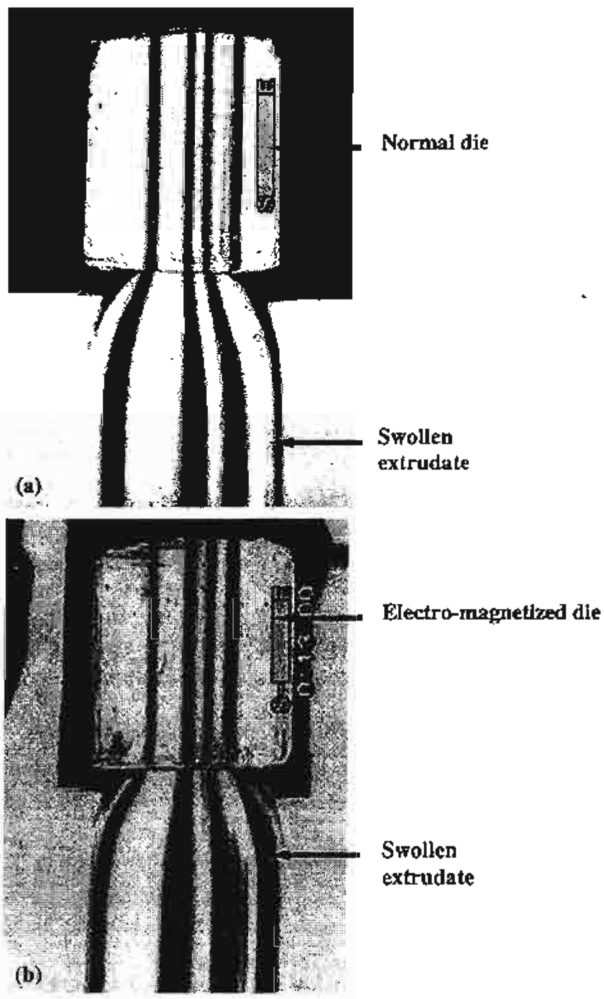

Figure 7. An example of PS extrudates while leaving the die temperature at 200°C: (a) with normal die; (b) with 1.56 Tesla magnetic flux density.

the die diameter had to equalize to form a plug flow. During this flow pattern transition, the melt velocity at the center had to slow down while that near the wall accelerated. A sudden reduction in melt velocity at the center causes the melt to flow radially and led to an expansion of the melt in radial direction. This extrudate expansion suppressed the dimension change of the melt near the die wall—the extrudate swell ratio near the die wall decreased in order to maintain the overall swell ratio of the extrudate for a given shear rate. This explains why the extrudate swell ratio was relatively high at the center of the die. The shape of the velocity profiles can also be used to explain the discrepancies in the radial swell profiles observed in the screw extruder in this work and those in the capillary rheometer previously reported[21]—the melt velocity profile in the extruder was shaper (suggesting higher local shear rate) and the swell ratios for any radial positions of the extrudate from the extruder were relatively greater.

It is interesting to note that die temperatures did not have a considerable effect on the changes in the radial extrudate swell ratio, although it did on the overall swell ratio as discussed earlier. Unlike the radial extrudate swell profiles, the velocity profile gradients appeared to be sharper as the die temperature was increased, this being a result of a slight increase in the melt velocity at the duct centre.

Extrudate swell and velocity profiles using an electro-magnetized die

It is for the first time that the die of a single screw extruder was applied by an electro-magnetic field, and the changes in extrudate swell ratio were monitored not only as a function of the overall swelling but also as a function of radial positions across the die diameter.

Overall extrudate swell ratio

Figure 7 shows an example of the captured PS extrudate flowing from the die exit at a wall shear rate of 5.1 sec^{-1} and a die temperature of 200°C with and without application of the magnetic field to the die in the single screw extruder. It can be clearly seen that for the same test conditions the swelling ratio of the extrudate (both overall and each colored melt layer) with the magnetic field was much greater than that without the magnetic field. Figure 8(a)–8(d) shows the overall extrudate swell ratio of PS melt as a function of magnetic flux density in the die for three different shear rates at die temperatures of 200, 210, 220 and 230°C, respectively. It was observed that the average swell ratio of the PS melt ranged from 1.23 to 1.72 (23–72% extrudate expansion as compared to the die diameter). The extrudate swell ratio at high shear rate (17.1 sec^{-1}) was lower than that at the shear rates of

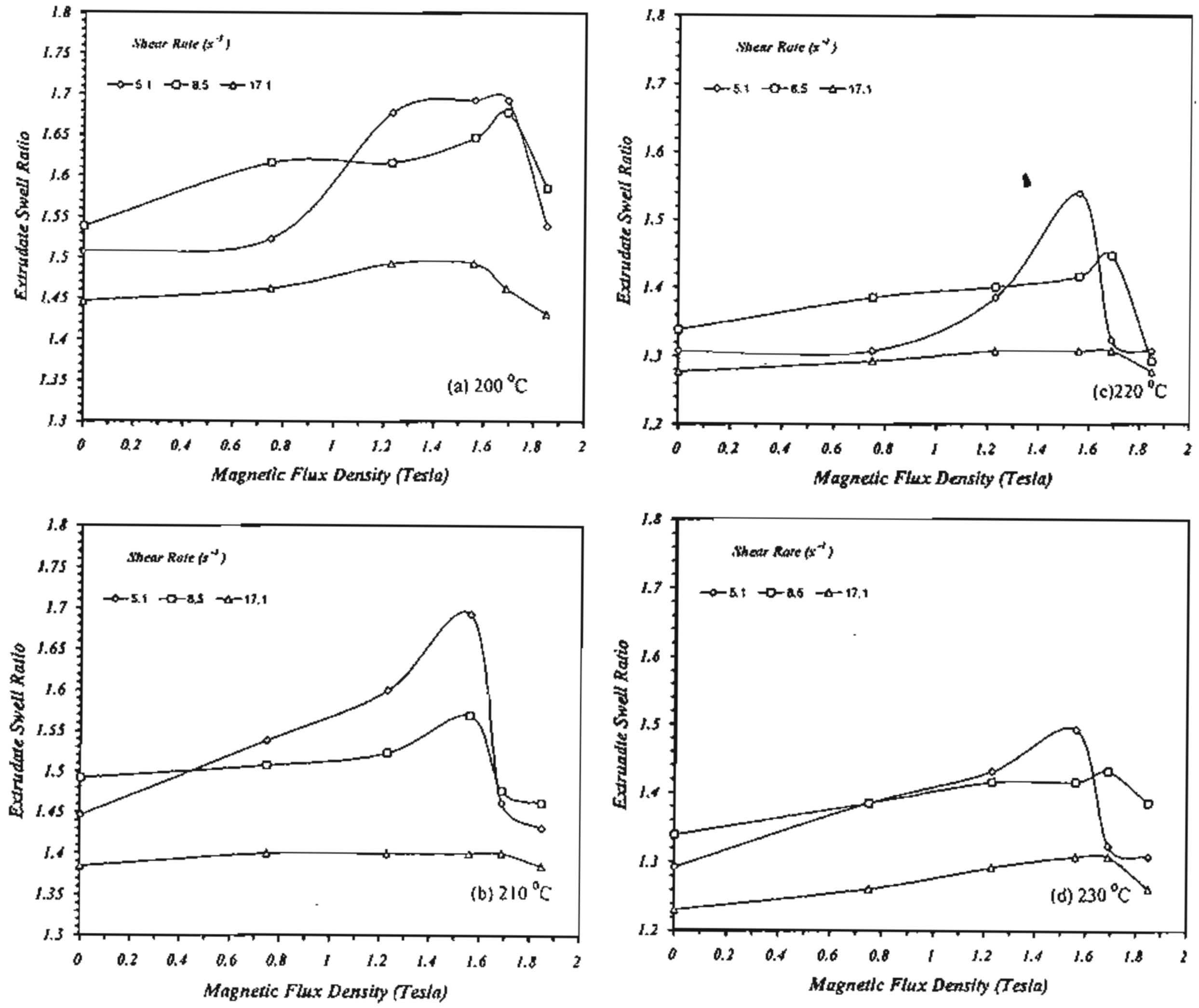

Figure 8. Overall extrudate swell ratio of PS melt as a function of magnetic flux density for different die temperatures: (a) 200°C; (b) 210°C; (c) 220°C; (d) 230°C.

5.1 and 8.5 sec^{-1}. The overall swell for all shear rates increased with increasing magnetic flux density to a maximum value and then started to reduce at higher magnetic flux densities. The magnetic flux density to give the maximum overall extrudate swell varied depending on the shear rate, and this behavior was observed for all die temperatures. The increase in the swell ratio at the magnetic flux density from 0–1.69 Tesla is thought to involve the magnetic torque produced under the electro-magnetic field, and then the change in molecular alignment of the melt during the flow. The polystyrene used in this work is considered to become anisotropic although it is an amorphous material, because the measurement was made during the flow of the PS melt. According to previous work,[24] there are two energy factors affecting the extrudate swell of a melt during extrusion; the first being elastic stored energy due to the shearing ($E_{elastic}$ as in eqn (1)) and the other being anisotropic magnetic energy due to the applied magnetic field (E_{mag} as in eqn (2)).

$$E_{elastic} = (1/2)G'(\lambda - 1)^2 \quad (1)$$

$$E_{mag} = -(2\mu_0)^{-1}\chi_a(B \cdot n)^2 \quad (2)$$

where G' is the elastic modulus, μ_0 is magnetic permeability of vacuum, $\chi_a = \chi_{\|} - \chi_{\perp}$ is the difference between the volumetric diamagnetic susceptibilities in the fiber axis direction ($\chi_{\|}$) and in the direction perpendicular to the fiber axis ($\chi_{\perp}$), B is the applied magnetic flux density (Tesla). The E_{mag} value is dependent on the deformation λ through χ_a.

In this work, magnetic field was applied to the die and the extrudate swell of the melt was studied. Therefore, the resulting swelling of the extrudate was due to a sum of these two energy factors. For a given shear rate, the elastic energy was assumed to be constant and the change in extrudate swell was solely caused by the magnetic energy. This was why the extrudate swell increased with increasing the magnetic flux density. In this respect, the PS melt could exhibit a flow induced anisotropy, and thus be induced by a magnetic field. In other words, the alignment and extensibility of the molecular chains of the PS flowing in the die would allow the melt to be more anisotropic.[25] When the magnetic flux density was increased to higher than 1.69 Tesla, the greater alignment might be expected, and the molecular chains might have experienced relatively high frictions, and this would cause a rise in melt temperature during the flow. This temperature increase in the melt (thus conducted to the die) would then result in reductions of both melt viscosity[26] and magnetic flux density[27]. As a consequence, the extrudate swell can be decreased.

With respect to wall shear rate, the maximum swelling peak of the melt appeared to shift to higher magnetic flux density with increasing wall shear rates, and the value of the maximum swell ratio decreased with increasing wall shear rates. Both effects were linked to the residence time of the melt. If the melt had more time (low extrusion rate) to absorb the magnetic torques produced from the magnetic field in the die, it was likely to swell more. This implies that there is a balance between the shearing and magnetic effects that acted on the molecules of the melt. It was postulated in this work that the magnetic effect was dominant at low shear rates while the shearing force was more pronounced at high extrusion rates. When considering the effect of die temperature, the trend of the swelling curves as a function of magnetic flux density was very similar, but all the values decreased with increasing die temperature. For a given shear rate, the magnetic field had a greater effect on the melt at lower die temperatures. In this case, it could be said that the decrease in the extrudate swell with increasing die temperature is associated with reductions of the melt elasticity and also the magnetic flux density.[27] In summary, the magnetic torque had a considerable effect on the extrudate swell ratio if the melt was extruded at low shear rate and low die temperature..

Radial extrudate swell and velocity profiles

Figure 9(a)–9(c) shows the extrudate swell ratio as a function of the r/R position in the die for different magnetic flux

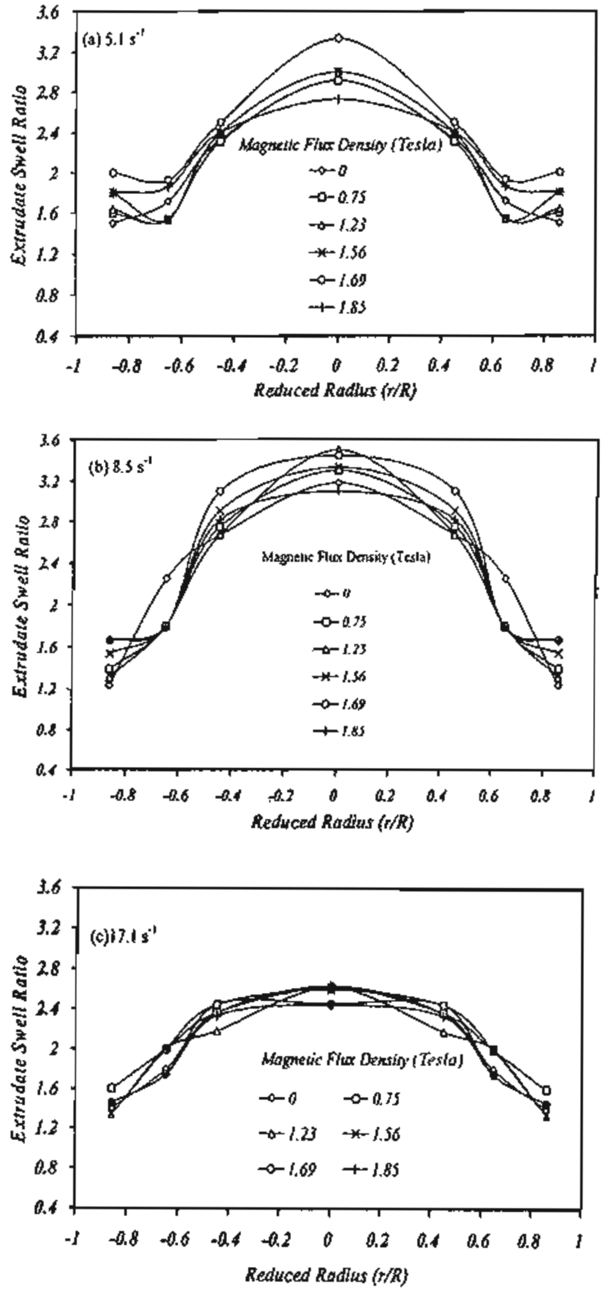

Figure 9. Radial extrudate swell profiles of PS melt at various magnetic flux densities for different wall shear rates at 200°C: (a) 5.1 sec^{-1}; (b) 8.5 sec^{-1}; (c) 17.1 sec^{-1}.

densities at a selected die temperature of 200°C and at wall shear rates of 5.1, 8.5 and 17.1 sec^{-1}, respectively. Similar to the case of non-magnetic die, the radial swell ratio decreased with increasing r/R position for any given shear rate. The highest extrudate swell ratio occurring at the duct center was in the range 2.4–3.3 (~140–230% extrudate expansion as compared to the die diameter). The lowest swell ratio was 1.2 (~20% extrudate expansion) which exhibited around r/R position of 0.85. Unlike the non-magnetic die as discussed earlier, there was no extrudate contraction when using the magnetic die. The change in the extrudate swell profiles across the die diameter had already been discussed to be linked with that in the melt velocities across the die diameter. It was observed that the swell profiles changed continuously with changing magnetic flux density. The extrudate swell changes appeared more pronounced at a low shear rate (5.1 sec^{-1}), especially at the center of the duct. This observation confirmed the earlier statement that the magnetic effect was significant at low shear rate. There were two regions exhibiting obvious changes in swell ratio across the die diameter, one being around the center of the duct and the other being around r/R of 0.65–0.85. At these regions, the extrudate swell ratio appeared to increase with increasing magnetic flux density up to ~1.69 Tesla, and to decrease for higher magnetic flux densities (1.85 Tesla).

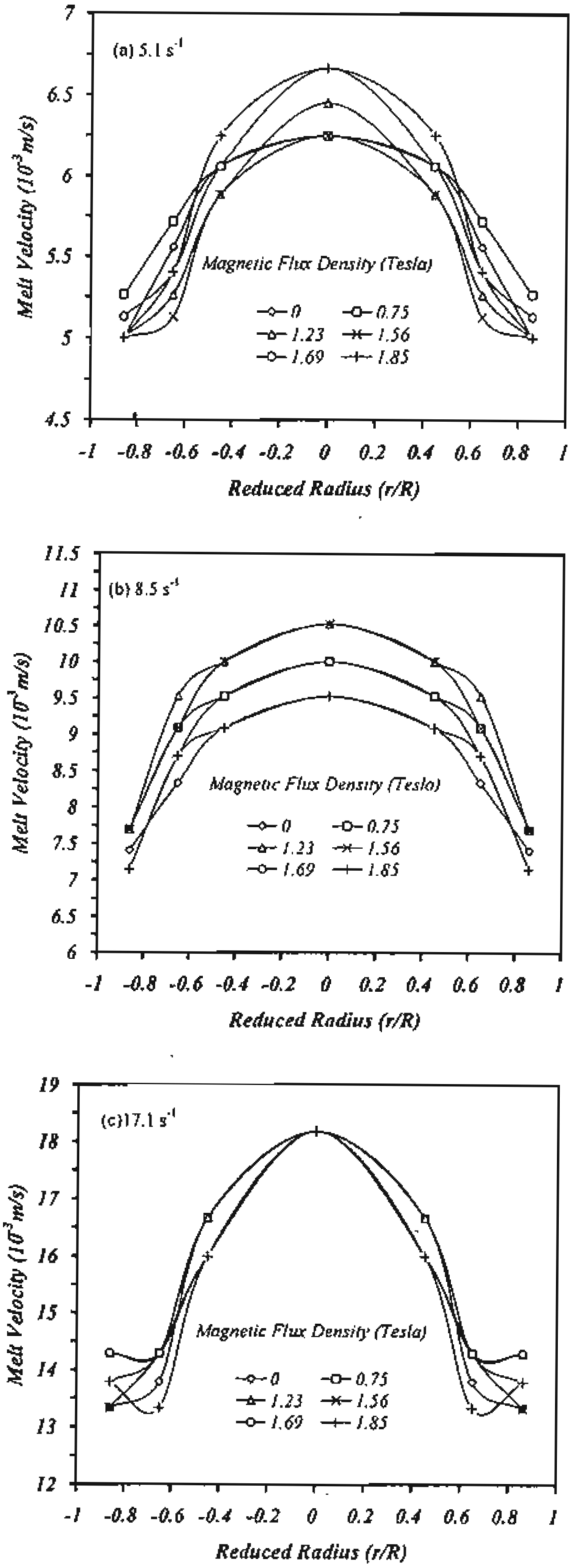

Figure 10. Radial velocity profiles of PS melt at various magnetic flux densities for different wall shear rates at 200°C: (a) 5.1 sec^{-1}; (b) 8.5 sec^{-1}; (c) 17.1 sec^{-1}.

The changes in the extrudate swell profiles of the melt due to the application of the electro-magnetic field to the die can be explained in terms of the velocity profiles as a function of the r/R positions for different shear rates and magnetic flux densities at a selected die temperature of 200°C. It can be seen from Fig. 10(a)–10(c) that the general trend of the radial velocity profiles was very similar to that for the radial swell profiles. The highest melt velocity was found at the center of the die, the melt velocity decreasing with increasing r/R. The changes in the melt velocity due to changing magnetic flux densities were more pronounced at low shear rate (5.1 sec^{-1}). As stated earlier, the shape of the radial swell profiles was associated with the development of velocity profiles of the melt flowing in the die and the equalization of the velocity profiles at the die exit. A sudden reduction in melt velocity at the center flowing in the die to the die exit caused the melt to flow radially and led to an expansion of the melt in radial direction, this suppressing the dimension change of the melt near the die wall. This implies that the higher the melt velocity, the greater the swelling level of the melt. Similar to the extrudate swell profiles, the melt velocity at the centre increased with increasing magnetic flux density up to a maximum and then decreased at higher magnetic flux densities, whereas that near the die wall progressively decreased with increasing magnetic flux density.

CONCLUSION

An electro-magnetized die was used to monitor the overall and radial extrudate swell ratios of a PS melt in a single screw extruder and the following findings are noted:

- In normal die case, the average overall swell ratio of the PS melt ranged from 1.25 to 1.55 (~25–55% extrudate expansion). The swelling ratio increased with increasing wall shear rate up to 8.5 sec^{-1} and then decreased at 17.1 sec^{-1}. Increasing die temperature caused a reduction of extrudate swell ratio. For radial swell and velocity profiles, the swell ratio decreased with increasing radial position. The highest extrudate swell ratio occurred at the duct center and was in the range 2.0–3.0 (100–200% expansion) whereas the lowest was found near the die wall. An extrudate contraction of the melt layer near the die wall was observed in this case. The explanation for the change in swell ratio at each radial position was linked with the development of melt velocity profiles measured in the die. The die temperature had no effect on the change of the radial swell profiles.

- In electro-magnetized die case, the average overall swell ratio was found to range from 1.23 to 1.72 (~23–72% melt expansion). The overall extrudate swell increased with increasing magnetic flux density to a maximum value and then decreased at higher magnetic flux densities. The magnetic flux density to give the maximum overall swell increased with shear rate. The magnetic energy was thought to have a pronounced effect on the swell ratio when extruding the melt at low shear rate and low die temperature. For radial swell and velocity profiles, the change in the swell ratio as a result of the applied magnetic field was relatively more pronounced at the centre of the duct, especially at low extrusion rate. The highest extrudate swell ratio, which occurred at the duct center, was in the range 2.4–3.3 (140–230% expansion). In the magnetized die case, there was no extrudate contraction of the melt layer near the die wall.

Acknowledgments

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REFERENCES

1. Kar KK, Otaigbe JU. *J. Elast. Plast.* 2001; 33: 297.
2. Huang JC, Leong KS. *J. Appl. Polym. Sci.* 2002; 84: 1269.
3. Guillet J, Seriai M. *Rheol. Acta.* 1991; 30: 540.
4. Sombatsompop N, Dangtagee R. *J. Appl. Polym. Sci.* 2001; 82: 2525.
5. Wilczynski K, Szymaniak Z, Nastaj A. *Polimery* 2003; 48: 204.
6. Yarusso BJ. *J. Non-New. Fluid Mech.* 1991; 40: 103.
7. Christodoulou KJ, Wood AK, Sombatsompop N. *SPE ANTEC* 1998; 44: 915.
8. Munstedt H, Schwetz M, Heindl M, Schmidt M. *Rheol. Acta* 2001; 40: 384.
9. Lee WS, Ho HY. *Polym. Eng. Sci.* 2000; 40: 1085.
10. Sombatsompop N. *J. Appl. Polym. Sci.* 2002; 86: 509.
11. Sombatsompop N, Sergsiri S. *Polym. Adv. Technol.* 2004; 15: 472.
12. Kiriakidis DG, Mitsoulis E. *Adv. Polym. Technol.* 1993; 12: 107.
13. Kajiwara T. *Nihon Reoroji Gakkaishi* 1999; 27: 219.
14. Mitsoulis E. *J. Rheol.* 1986; 30: S23.
15. Kiselev AP, Kanavets IF. *Sov. Plast.* 1967; 34: 10.
16. Intawong NT, Sombatsompop N. *Polym. Eng. Sci.* 2004; 44: 1960.
17. Ulaby FT. *Fundamentals of Applied Electromagnetics*. Prentice-Hall: New Jersey, 2001.
18. Sombatsompop N, Intawong NT, Intawong NS. *Sens. Actuat. A. Phys.* 2002; 102: 76.
19. Sombatsompop N, Patcharaphun S. *Polym. J.* 2001; 33: 491.
20. Sombatsompop N, O-Charoen N. *J. Appl. Polym. Sci.* 2003; 87: 1713.
21. Intawong NT, Sombatsompop N. *Polym. Eng. Sci.* 2004; 44: 2298.
22. Han CD. *Multiphase Flow in Polymer Processing.* Academic Press: New York, 1981.
23. Matsunaga K, Kajiwara T, Funatsu K. *Polym. Eng. Sci.* 1998; 38: 1099.
24. Intawong NT, Kimura T, Tamura M, Xiaojun L, Sombatsompop N. *Polym. J.* in press.
25. Amundson KR. In *Electric and Magnetic Field Effects on Polymeric Systems Exhibiting Long-range Orientational Order*, Wire DL (ed.). Marcel Dekker: New York, 1998.
26. Swan PL, Garcia-Rejon A, Derdouri A, Dealy JM. *SPE ANTEC* 1990; 36: 1607.
27. Callister WD. *Materials Science and Engineering: The Influence of Temperature on Magnetic Behavior.* John Wiley & Sons: Singapore, 1985.

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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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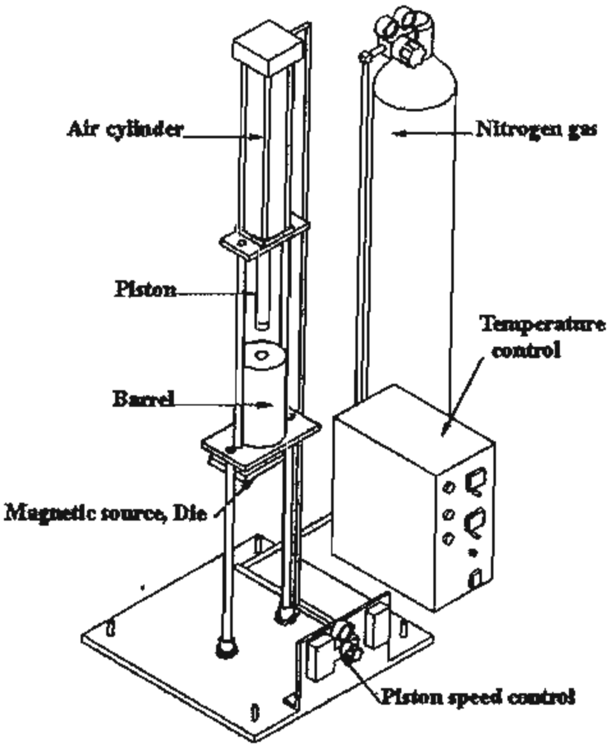

Figure 1. Schematic representation of apparatus used for extrudate swell measurements.

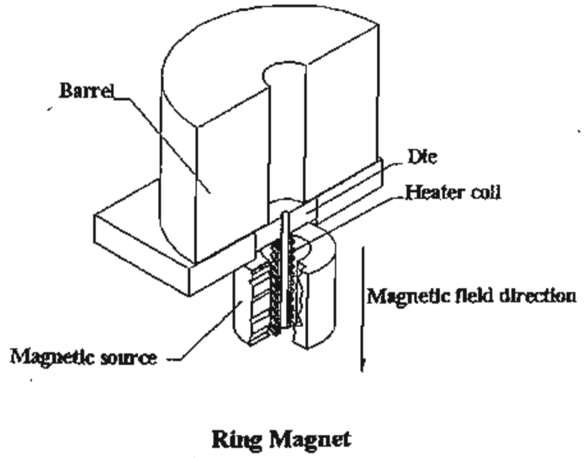

Figure 2. The layout of the permanent ring magnet near the die.

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.

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

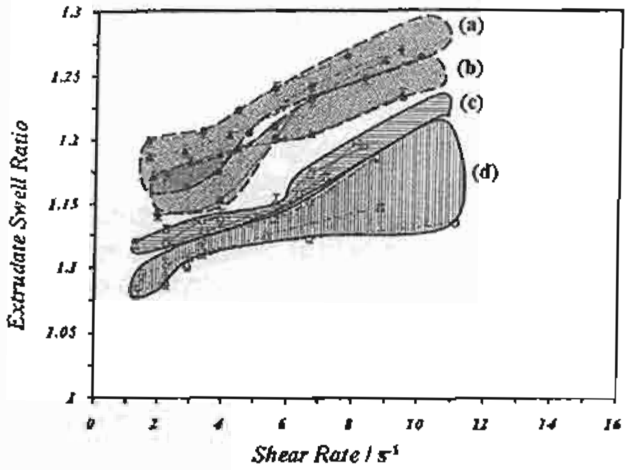

Figure 3. Extrudate swell ratios measured at 190 °C plotted against the shear rate. Groups (a) to (d) enclosing three experimental data indicate the experiments with steel die [(a) with and (b) without magnetic field] and the stainless die [(d) with and (c) without magnetic field].

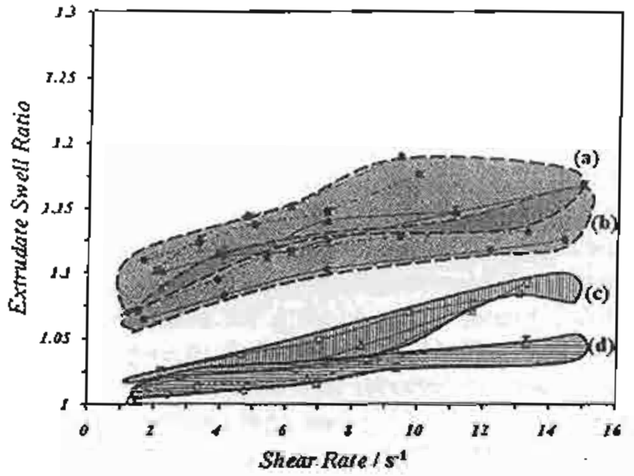

Figure 5. Extrudate swell ratios measured at 230 °C plotted against the shear rate. Groups (a) to (d) enclosing three experimental data indicate the experiments with steel die [(a) with and (b) without magnetic field] and the stainless die [(d) with and (c) without magnetic field].

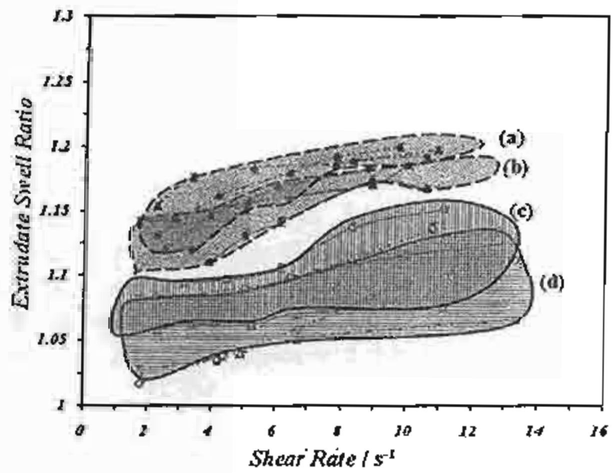

Figure 4. Extrudate swell ratios measured at 210 °C plotted against the shear rate. Groups (a) to (d) enclosing three experimental data indicate the experiments with steel die [(a) with and (b) without magnetic field] and the stainless die [(d) with and (c) without magnetic field].

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.

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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REFERENCES

1. P. L. Swan, A. Garcia, A. Derdouri, and J. M. Dealy, *SPE ANTEC '90*, **36**, 1607 (1990).
2. S. Eggen and E. L. Hinrichsen, *Polym. Eng. Sci.*, **36**, 410 (1996).
3. Y. R. Cao and H. L. Li, *Polym. Eng. Sci.*, **42**, 1534 (2002).
4. N. Sombatsompop, *J. Appl. Polym. Sci.*, **86**, 509 (2002).
5. J. Z. Liang, *Plast. Rubber Compos. Process. Appl.*, **23**, 93 (1995).
6. E. Mitsoulis, *J. Rheol.*, **30**, S23 (1986).
7. B. Yang and L. J. Lee, *Polym. Eng. Sci.*, **7**, 1088 (1987).
8. N. Sombatsompop and S. Sergsiri, *Polym. Adv. Technol.*, **15**, 480 (2004).
9. N. T. Intawang and N. Sombatsompop, *Polym. Eng. Sci.*, **44**, 2298 (2004).
10. N. T. Intawang and N. Sombatsompop, *Polym. Eng. Sci.*, **44**, 1960 (2004).
11. T. Kimura, *Polym. J.*, **35**, 823 (2003).

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Test Procedure

A comparative study on extrudate swell ratio of polystyrene in a capillary rheometer and a single screw extruder

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Abstract

This article offers a comparative discussion on the extrudate swell ratios and velocity profiles of polystyrene melt flow using the same die with two different machines which were a constant-rate capillary rheometer and a single screw extruder. The swelling ratio and melt velocity profiles were simultaneously measured at different positions across the die diameter, with and without application of a magnetic field to the die. The results suggested that the overall swell ratio of the PS melt measured in the extruder was greater than that in the rheometer, this being caused by the differences in the flow properties of the melt in these two machines. The discrepancies in the radial extrudate swell profiles from the rheometer and extruder could be explained by the velocity profile development. When an electro-magnetized die was used, the swelling ratio of the melt changed with magnetic flux density and was affected by the size of the machinery used.

Keywords: Extrudate swell; Rheology; Rheometer; Extrusion process; Flow properties

1. Introduction

Under normal processing conditions, polymer melts exhibit viscoelastic behaviour and their rheological characteristics depend on how the polymer melt is being deformed. The extent of deformation and deformation history are strongly affected by the flow geometry, which is determined by the type of deformation [1]. The most common test apparatus used for the determination of the flow properties of polymer melts for any processing technique is the extrusion capillary rheometer [2]. However, it has been evident that the flow property results produced in the capillary Rheometer are dependent on the design of the apparatus, the same die giving different results when used in different apparatus designs [3,4]. In the extrusion process, the swell of he polymer melt is considered one of the main factors to determine quality and dimensions of the final product. Most experimental data are only available for the overall extrudate swell (the ratio of the extrudate size to the die size), and the explanation for the change in extrudate swell is usually associated with the recoverable elastic deformation developed during flow through the die [1]. Scientific evidence [5,6] has clearly suggested that velocity profiles and die swell were closely related, but most work in the literature has been carried out with these two parameters separately and theoretically.

This present article aimed to continue from two recent works [7,8] on measurements of overall and radial extrudate swell and velocity profiles of PS melt flowing in the die of a constant-rate capillary

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rheometer. The first publication [7] proposed a novel experimental technique, so-called Parallel Coextrusion Technique (PCT), to *simultaneously* measure the radial swell and the velocity profiles of the melt and the second publication [8] introduced a novel *electro-magnetized die* for controlling the extrudate swell. In this present article, we attempt to offer comparative discussion on the extrudate swell ratios and velocity profiles of the PS melt flowing using the same die with two different processing machines, these being a constant-rate capillary rheometer (CR) and a single screw extruder (SSE). Such comparison had never been reported in literature, especially the swelling of the extrudate being measured at different positions across the die diameter and under a magnetic field.

2. Experimental

2.1. Raw material

All tests used polystyrene (Styron656D 267) with a melt flow rate of 7 and a density of 1.350 g/cm^3, supplied in granular form by Siam Polystyrene Co., Ltd (BKK, Thailand).

2.2. Experimental technique and apparatus

A circular cross-section die with 64×10^{-3} m in length and 5×10^{-3} m in diameter was used in this work, the design and dimensions of the die used being given in previous works [7,8]. The experimental arrangement was that the circular die system was connected at the bottom of the CR and SSE machines, the size and details of the manufacturers for the rheometer being given in previous published work [7], whereas the single screw extruder (ThermoHaake PolyDrive Extruder) used had a barrel length-to-diameter (L/D) ratio of 200/25 mm/mm. The temperature profile on the extruder length was 170, 180, and 190 °C. The die temperature of both machines used was varied from 200 to 230 °C.

The experimental technique used for the measurements of the radial extrudate swell and velocity profiles was a Parallel Co-extrusion Technique (PCT), the detail of the technique being already published and found elsewhere [7]. The PCT was based on a parallel co-extrusion of colored melt-layers into an uncolored melt-stream from the barrel into and out of the circular cross-section die. The radial extrudate swell ratio values (B_r) were obtained by comparing the thickness of the colored layer of the extrudate outside the die, for a given reduced radius (r/R) position across the die diameter. The r/R range of interest used in this work was from 0.0 to 0.86. The velocity profile, which was also measured by the PCT technique, was based on monitoring a relatively small and light foreign object (corn particles) flowing along the melt streams (colored layers), and the measurements were carried out by recording the times taken for the corn particle loaded into the melt layers to travel for a given distance in the die (10 mm before the die exit). It should be noted that the extrudate swell ratio and velocity profiles at any radial positions across the die were averaged from five determinations, and were obtained in the piston displacement range of 85–92 mm down the barrel. Flow curves of the melt was evaluated in both machines using the same die and test conditions as used in the swell and velocity profile measurements, the experimental procedures being detailed elsewhere [9].

Another novelty offered in this work was the application of an electro-magnetic field to the die for the extrudate swell measurement. The design and manufacture of the electro-magnetized die were detailed in previous work [8]. In this work, we studied the extrudate swell and velocity profile results between the capillary rheometer and the single-screw extruder in the die with and without the magnetic field (varying magnetic flux densities from 0.75 to 1.85 T) for different die temperatures (200–230 °C) and wall shear rates (5.1, 8.5 and 17.1 s^{-1}).

3. Results and discussion

Fig. 1 shows the overall extrudate swell values of the PS as a function of wall shear rate in the capillary

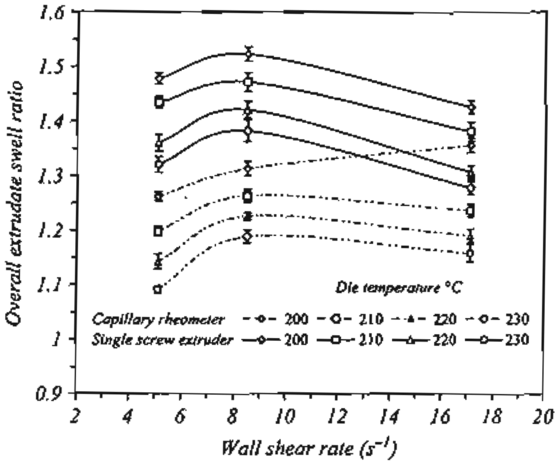

Fig. 1. Overall extrudate swell of PS melt as a function of wall shear rate in the capillary rheometer and extruder for different die temperatures.

rheometer (CR) and the single-screw extruder (SSE) for different die temperatures. Generally, it was found that the overall extrudate swell of the PS from both machines decreased with increase of temperature, and tended to increase with increasing shear rate to the maximum swell of about 8.5 s^{-1}, and then the swelling ratio value decreased with higher shear rate (17.1 s^{-1}). The decrease of the extrudate swell ratio at higher shear rate (17.1 s^{-1}) in all test conditions were probably influenced by more pronounced shear heating effect during the flow of the melt [10,11]. For a given wall shear rate, the extrudate swell ratios from the CR and the SSE machines were different, that of the SSE being approximately 20–25% higher. The discrepancies in the overall extrudate swell can be explained through the flow curve of the melt determined in the two machines, the results being shown in Fig. 2. It was found that the apparent wall shear stress of the PS melt produced from the SSE was higher than that from the CR. This was expected since the extrudate swell was dominated by the shear stress applied to the polymer melt during the flow [1].

Fig. 3 shows the radial extrudate swell profiles of the PS observed in the CR and the SSE for different wall shear rates at a selected die temperature of 210 °C. It was found that the swell ratio of the melt was not uniform across the die diameter, the swelling being greater at the die centre and decreasing with increasing *r/R* positions. As expected, the radial extrudate value in the SSE was higher than that in the CR machine. It was surprising that the extrudate swell decreased with increasing wall shear rate, the effect being more pronounced at the die center. This implied that the flow property results in Fig. 2 could not be used to

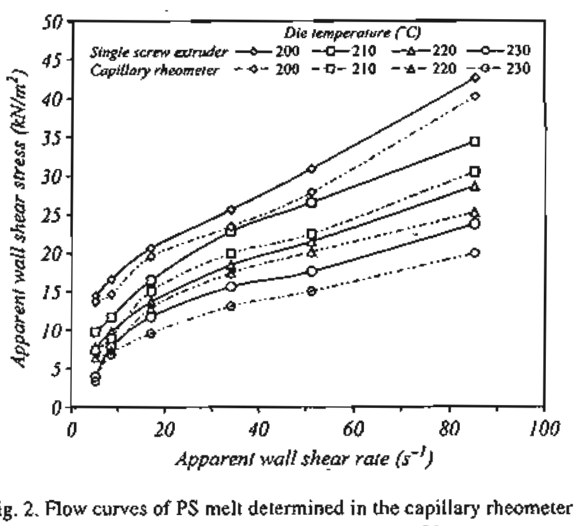

Fig. 2. Flow curves of PS melt determined in the capillary rheometer and screw extruder using a die temperature of 210 °C.

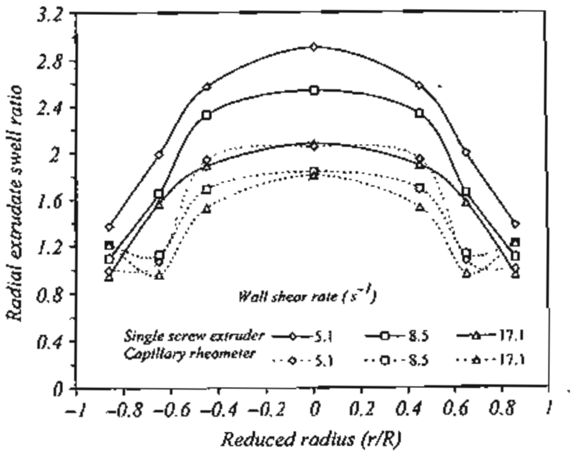

Fig. 3. Radial extrudate swell profiles of PS melt in the capillary rheometer and extruder for various wall shear rates at a selected die temperature of 210 °C.

explain the radial swell of the extrudate. In this article, we believe that the radial extrudate swell profiles were closely linked with the development of the velocity profiles of the melt in the die. Therefore, melt velocity profiles measured across the die diameter from these two machines were required to explain the differences in the radial swell results in Fig. 3. Fig. 4 shows the velocity profiles of PS melt in the CR and SSE machines for various wall shear rates at a selected die temperature of 210 °C. It was observed that the velocity profiles observed in the two machines were very similar, but for given *r/R* radial position, the melt velocity in the SSE was slightly greater than that in the CR. Considering the shape of the velocity profiles, the velocity profiles of the PS melt in the SSE were sharper than those in the CR, implying that the local shear rates across the die diameter of the SSE were greater. As a result, the swell from the single screw extruder was greater. The reason for the sharper velocity profiles in the SSE machine could be expected, probably due to the melt flow from the barrel to the die for swelling in the SSE occurring as a result of a parabolic melt flow by action of screw rotation, while that in the CR occurred due to the movement of a flat piston.

Fig. 5 shows the overall swell of PS extrudate as a function of magnetic flux density using an electromagnetized die in the CR and SSE machines, for various wall shear rates using a die temperature of 210 °C. In general, it was found that the swell ratio increased with increasing magnetic flux density to a maximum (around 1.6 T) and then decreased at higher flux density. The explanation for this behavior has been given in previous works [8,12]. It was very surprising

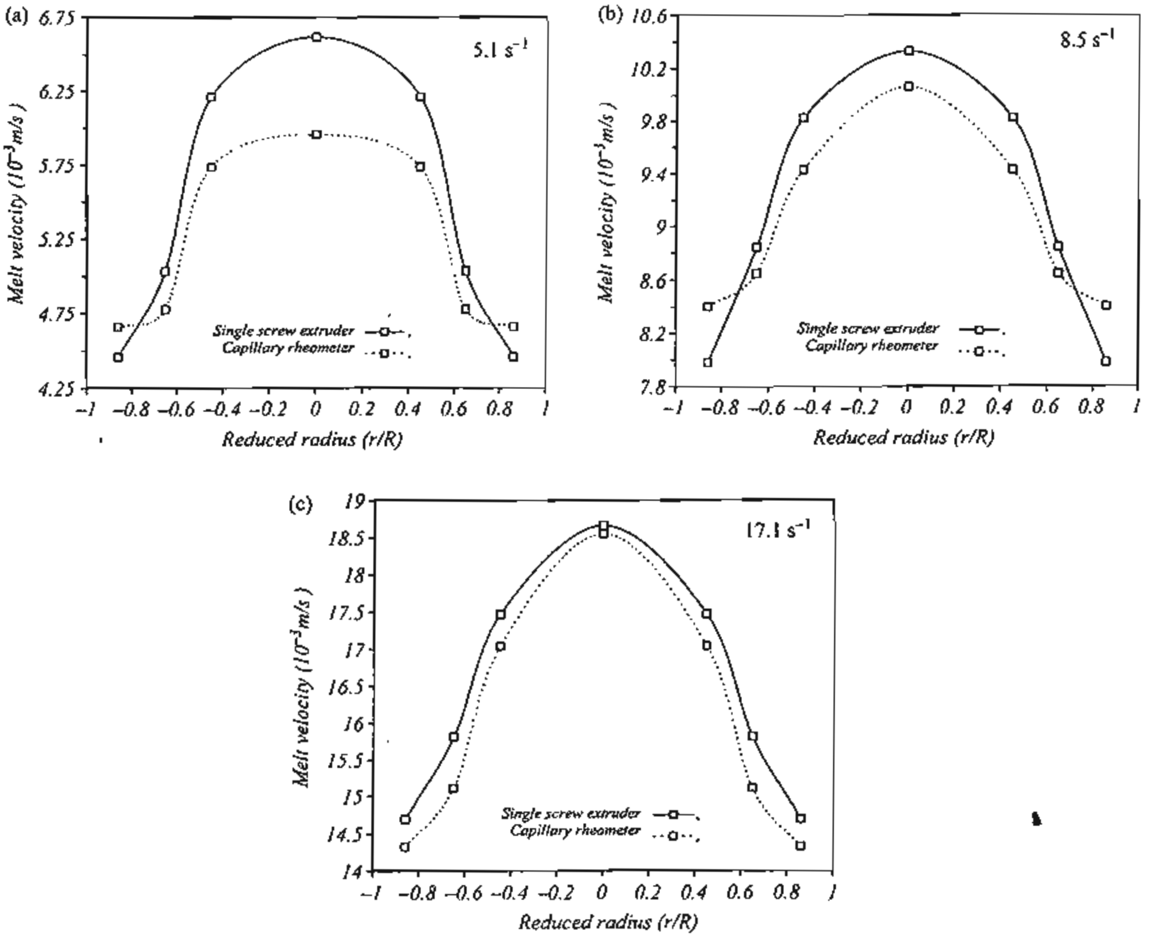

Fig. 4. Velocity profiles of PS melt in the capillary rheometer and extruder for various wall shear rates at a selected die temperature of 210 °C: (a) $5.1\ s^{-1}$, (b) $8.5\ s^{-1}$, (c) $17.1\ s^{-1}$.

when considering the swell results in CR and SSE machines that, for a given magnetic flux density, the extrudate swell ratio in the CR was higher than that in the SSE. This could be explained in relation to previous work by Sombatsompop and Sergsiri [13] who investigated the effect of magnetic field on the extrudate swell ratio in a capillary rheometer using different barrel inner diameters (the outer barrel diameter being the same). They found that the barrel with smaller inner diameter, which had more steel in the cross section, produced greater swelling of the extrudate as compared with that with larger inner diameter. This was because the greater the amount of steel the higher the magnetic flux density for any applied electric load. In relation to this work, the surface areas of the die parts in the CR and SSE systems were calculated, and it was found that the steel surface

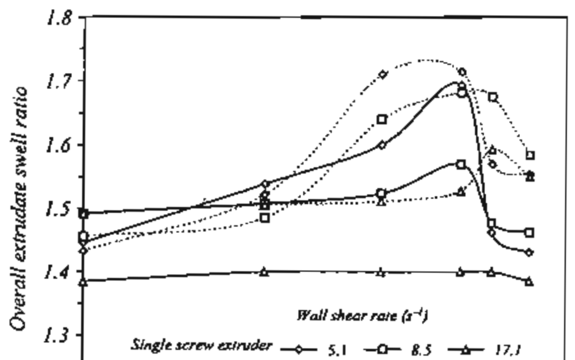

Fig. 5. Overall extrudate swell of PS melt as a function of magnetic flux density in the capillary rheometer and extruder for various wall shear rates using a die temperature of 210 °C.

areas were 1972 and 912 cm^2 for the CR and SSE, respectively. Although this explanation might not be considered totally scientific, it had some practical implication that the machinery size would become an important issue in controlling the sizes and quality of the extrudates or the final products when extruding PS melt using a magnetized die.

4. Conclusion

This article has suggested that the overall swell ratio of polystyrene melt measured in a single screw extruder was greater than that in a capillary rheometer, this resulting from the differences in the melt flow properties in these two machines. The discrepancies in the radial extrudate swell profiles from the rheometer and screw extruder machines could be explained through the simultaneously measured velocity profiles. When a magnetized die was used, the swelling behaviour of the PS melt varied with magnetic flux density and was dependent on the size of the machinery used.

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References

[1] F.N. Cogswell, Polymer Melt Rheology, George Godwin Ltd, London, 1981.
[2] N. Sombatsompop, M.C. Tan, A.K. Wood, Polym. Eng. Sci. 37 (1997) 270.
[3] A.K. Wood, A.G.G. Read, J.G.A. Lovegrove, Plast. Rubb Proc. Appl. 12 (1998) 15.
[4] N. Sombatsompop, A.K. Wood, I.S. Godfrey, SPE ANTEC 43 (1997) 392.
[5] H. Munstedt, M. Schwetz, M. Heindl, M. Schmidt, Rheol. Acta 40 (2001) 384.
[6] K. Matsunaga, T. Kajiwara, K. Funatsu, Polym. Eng. Sci. 38 (1998) 1099.
[7] N.T. Intawong, N. Sombatsompop, Polym. Eng. Sci. 44 (2004) 1960.
[8] N.T. Intawong, N. Sombatsompop, Polym. Eng. Sci. 44 (2004) 2298.
[9] N. Sombatsompop, N. O-Chareon, J. Appl. Polym. Sci. 87 (2003) 1713.
[10] N. Sombatsompop, W. Chaiwattanapipat, Adv. Polym. Technol. 19 (2000) 79.
[11] A.K. Wood, Y.H. Judeh, M. Yue, UK Patent Application GB291197, 1996.
[12] N.T. Intawong, T. Kimura, M. Tamura, L. Xiaojun, N. Sombatsompop, Polym. J. 37 (2005) 541.
[13] N. Sombatsompop, S. Sergsiri, Polym. Adv. Tech. 15 (2004) 472.

Effect of Molecular Structure on Extrudate Swell Behavior for Different Thermoplastic Melts in an Electro-Magnetized Die

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The extrudate swell ratio of five different thermoplastic melts flowing in a constant shear rate rheometer having a capillary die with and without application of magnetic field was studied. The effects of the magnetic flux direction and density, die temperature, and wall shear rate on the extrudate swell and flow properties were investigated. The experimental results suggested that an increasing wall shear rate increased the swelling ratio for the polystyrene (PS), LLDPE, and PVC melts, but the opposite effect was observed for the ABS and PC melts. The extrudate swell ratio for the PS, ABS, PC, and LLDPE melts decreased with increasing die temperature, the effect being reversed for the PVC melt. Thermoplastic melts having high benzene content in the side-chain and exhibiting anisotropic character were apparently affected by the magnetic field, the extrudate swell ratio increasing with magnetic flux density. The effect of the magnetic field on the extrudate swell ratio decreased in the order of PS → ABS → PC. The extrudate swell ratio for the co-parallel magnetic field system was slightly higher than that for the counter-parallel magnetic field system at a high magnetic flux density. POLYM. ENG. SCI., 47:270–280, 2007. © 2007 Society of Plastics Engineers

INTRODUCTION

In polymer extrusion, extrudate swell is widely known as one of the main factors to determine the quality and dimensions of the polymer product and can be explained either by the recoverable elastic deformation developed during flow [1–3], or by the re-organization of melt velocity profiles developed in the die and the die exit [4–8]. Most published papers [9–18] have studied the dependency of extrudate swell and velocity profiles of polymer melts on the die size, die shapes (simple to complex die profiles), number of flow channels in a die, shear rate, molecular weight and structure, melt and die temperatures, and fillers. Apart from these papers, there have been some recent publications dealing with relatively new die design related parameters such as ultrasonic irradiation and magnetic field application during extrusion [19–21]. Cao and Li [19] applied ultrasonic irradiation during extrusion to, and monitored the swelling ratio of, a polypropylene melt and observed that the extrudate swell ratio was reduced and the apparent quality of the extrudate was improved by the use of appropriate irradiation intensity. Kimura [20] suggested that the effect of a magnetic field on nonmagnetic polymers was relatively small, but occurred in anisotropic polymers. In the case of amorphous polymers, which lack anisotropic structures, the alignment of the molecules under the magnetic field became very difficult and the direction of the magnetic alignment of these polymers is so far still unknown.

Recently, Sombatsompop and coworkers [21–26] published a number of papers on the effect of a magnetic field on the extrudate swell ratio of polystyrene (PS) melt in a capillary die of a capillary rheometer and a single screw extruder. Sombatsompop [21] originally designed and manufactured a magnetic circular die in a capillary rheometer and studied the effect of the magnetic flux density on the extrudate swell ratio of PS melt. The results showed that application of a magnetic field to the die gave a significant increase in the swelling ratio of the PS melt of up to 15–25%. The author proposed that the increased swelling of the PS could be caused by the changes in the molecular orientation or alignment due to magnetic torque. Later, Sombatsompop and Sergsiri [22] also studied the effect of the barrel diameter and magnetic flux density on the overall extrudate swell for PS melt in a capillary rheometer with the use of an electro-magnetized die, and found that a barrel diameter of 30 mm was the critical value at which the extrudate swell ratio and flow properties of the PS melt were significantly affected by the magnetic flux density. Works on the magnetic field effect were extended by Intawong and Sombatsompop [23–25] to examine the extrudate swell at different positions across the die diameter in both a capillary rheometer and a single screw extruder. They concluded that the changes in radial extrudate swell profiles could not be

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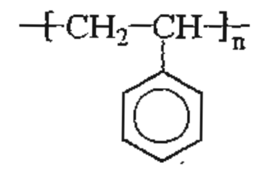
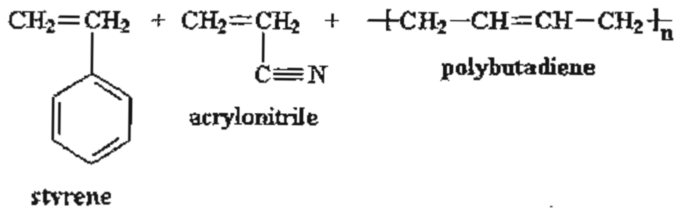
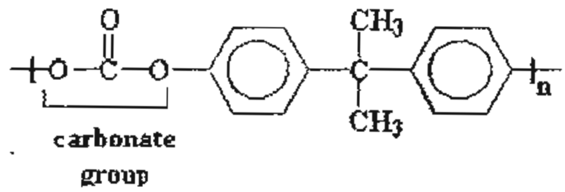
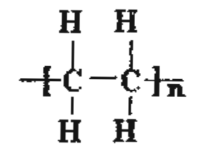
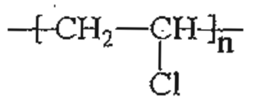
TABLE 1. Chemical structures and physical and thermal properties of thermoplastics used.

No.	Polymer	Grade/supplier	Chemical structure	MFI	T_g	Density
1	Polystyrene (PS)	Styron 656D267/ Siam Polystyrene Co., Ltd. (Thailand)		7.0	+105	1.35
2	Acrylonitrile butadiene styrene copolymers (ABS)	PA-747SS/Global Connections Co., Ltd. (Thailand)		0.7	+105	1.03
3	Polycarbonate (PC)	E2000R/Mitsubishi Engineering-Plastics Co., Ltd. (Thailand)		5.0	+150	1.20
4	Linear low-density polyethylene (LLDPE)	L2009F/Thai Polyethylene Co., Ltd. (Thailand)		0.9	−100	0.92
5	Poly(vinyl chloride) (PVC)	Y5911BLA/Thai Plastics & Chemicals Co., Ltd. (Thailand)		2.5	+80	1.54

reasoned by the shear rate change, but were closely linked with the development of the velocity profiles of the melt in the die. The extrudate swell ratio was high at the center and low near the die wall. The magnetic field effect was more pronounced if a steel die was used, when compared to a stainless steel die [26].

The above works by Sombatsompop and coworkers [21–26] logically stated that using an electro-magnetized die has become a new and unique parameter to altering the extrudate swell level in polymer extrusion applications. However, most published works of Sombatsompop and coworkers [22, 24–26] have been carried out on PS melt only. This article was aimed to extend an understanding of the magnetic field effect on other types of polymers. Therefore, a relationship between the molecular structure of polymer melts and their swelling behavior under a magnetic field during extrusion was of our interest. This was achieved by extruding different thermoplastic melts through a capillary die with adjustable magnetic flux densities. The effects of magnetic flux direction to the melt flow, wall shear rate, and die temperature on the extrudate swell were also examined.

EXPERIMENTAL

Raw Materials

Different thermoplastic melts were used to investigate the effect of the molecular structure of the polymer on the extrudate swell and the flow properties. The chemical structures and physical and thermal properties of the thermoplastics used are listed in Table 1; all the polymer being supplied in granular form.

Capillary Rheometer and an Electro-Magnetized Die

A constant-shear rate rheometer was employed in this work as used in previous works [18, 22–24]. A barrel, having a 30 mm in diameter and 150 mm in length, was used. The capillary die used was made of steel, 6 mm in diameter and 60 mm in length. To make an electro-magnetized die, the die body was wrapped with copper wires in order to apply and generate an electromagnetic field to the die. To prevent an electrical short-circuit, Teflon film was used between copper coil layers, the experimental apparatus arrangement being

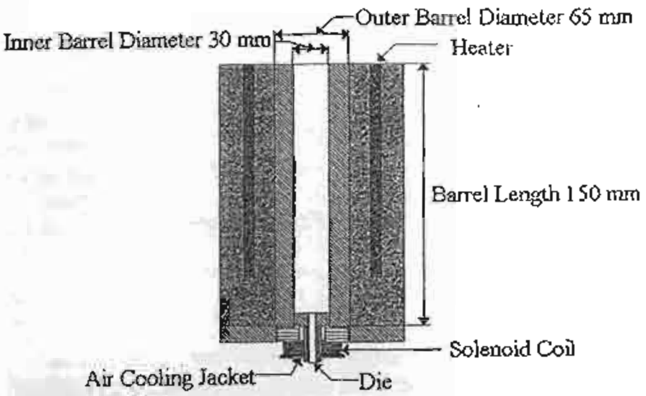


FIG. 1. A shear rate constant capillary rheometer for extrudate swell measurement and eletro-magnetized die system.

shown in Fig. 1. An electro-magnetic field was generated to the die by applying electricity to the copper solenoid coil, the electricity varying from 0 to 16 A. The dimensions and arrangement of the electro-magnetized die are shown in Fig. 2, which illustrates the magnetic field in the direction along the flow of polymer melts. It was of some concern that the application of the electro-magnetic field would result in additional heat because of the induction effect across the solenoid coil. This additional heat would then affect the die and melt temperatures during the tests. Therefore, an air-cooling jacket unit was constructed and placed around to the electro-magnetized die (see Fig. 1), the cooling unit being located between the solenoid coil and the die body. The flow-rate of the cooling air was adjusted and calibrated to reduce the additional heat so that the required temperature was achieved—some experimental results in the discussion section will confirm that no additional heat occurred during the test. A small pressure hole was located between the two die locations to detect the die entrance pressure drop by using a pressure transducer (Dynisco, Model PT460E-2CB-6, Franklin, MA). The temperature was controlled using a Digicon, Model DD6 RTD Pt-100 temperature controller.

Extrudate Swell Ratio and Flow Properties

Extrudate Swell Ratio. The swelling ratio (B) of the polymer extrudate was determined by the ratio of the diameter of the extrudate to the die, the extrudate diameter being based on the size of the extrudate diameter in the fully swollen state, approximately 1–2 in. away from the die lip. A video-camera (SSC-DC398P with 752H × 582V pixels and 480TV lines) and a high resolution macro zoom lens (3.3× magnification) were used to visualize the extrudate leaving the die exit whose results were recorded and displayed in real time using a personal computer. The size of the extrudate was carefully measured by replaying the recorded flow on the computer. All the extrudate swell results reported in this work were averaged from five determinations. With this procedure, any errors of the extrudate swell measurement in relation to the cooling and gravity effects could be minimized, the experimental errors being estimated to be ±0.5%. The scattering of the data reported in this paper was analyzed and indicated by error bars obtained from the Microsoft Office 2003 Excel program.

Flow Properties. The flow properties of polymer melts can be expressed in terms of the relationship of the wall shear stress to the wall shear rate, which were simultaneously determined under the conditions at which the extrudate swell was measured, whose calculations can be obtained elsewhere [12]. It should be noted that Bagley's and Rabinowitch corrections were not applied to the shear stress and shear rate data generated in this work because of two reasons. First, the shear stress and shear rate data were used solely for comparative reasons to illustrate the magnitude of the changes observed in the flow characteristics of the materials as a function of the electro-magnetic intensities applied. Second, the die dimensions used were constant throughout this work. There have been recent evidences [27, 28] that the Bagley's and Rabinowitch corrections are necessary if one wishes to obtain materials information that is independent of the die dimensions used.

Experimental Variables

All measurements were performed with and without the application of an electro-magnetic field to the die, the test variables studied being listed later.

Magnetic flux density and flux direction: In this work, the magnetic flux lines generated in the die had two different flux directions as follows.

1. Co-parallel (COP) direction to melt flow: The magnetic field occurred in the same direction and parallel to the flow direction of the melts. The magnetic flux density was altered by varying the amount of the electric current from 0, 10, 12, 14 to 16 A, these values corresponding to the magnetic flux densities of 0, 30, 37, 44, and 51 mT, respectively. These values were measured using a Teslameter at the centre position of the copper coil length, which appeared to give the maximum value for any given electric current. It was accepted that the generated magnetic flux density varied from location to location on the experimental rig (barrel and die bodies). Therefore,

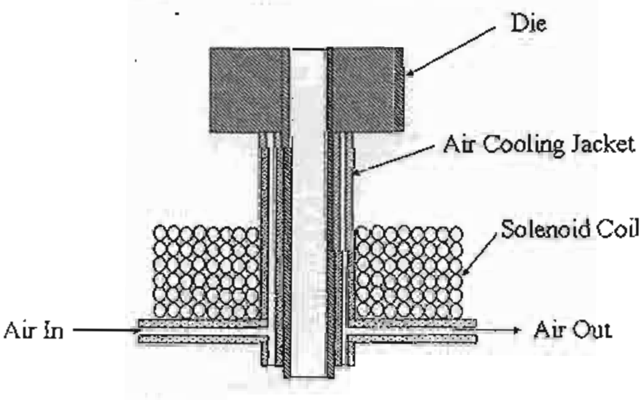


FIG. 2. The design and configuration of the electro-magnetized die.

TABLE 2. Die temperatures used for different thermoplastic melts.

Polymer	Die temperature (°C)
Polystyrene (PS)	175 and 215
Acrylonitrile butadiene styrene copolymers (ABS)	175 and 215
Polycarbonate (PC)	230 and 250
Linear low-density polyethylene (LLDPE)	160 and 190
Polyvinyl chloride (PVC)	160 and 180

the extrudate swell results obtained in this work were reported as a function of the maximum magnetic flux density, which occurred at the centre position of the copper coil in the capillary die.

2. Counter-parallel (CUP) direction to melt flow: The magnetic field occurred in the counter direction and parallel to the flow direction of the melts. The varying of the magnetic flux density was the same as explained in the COP direction.

Die temperature: To evaluate the effects of temperature on the extrudate swell and flow properties with and without the application of an electro-magnetic field to the die, two die temperatures were used, the selection of die temperatures being based on normal processing temperatures of the polymers used, which were varied depending on the polymer types. The die temperatures for all thermoplastics used are listed in Table 2.

Wall shear rate: By trial and error experiments, the shear rates used were below the critical shear stresses for the onset of melt distortions (sharkskin and melt fracture) under the test temperatures, this allowing the swelling measurements to be more accurate. It should be noted that the share rates used in this work for the PVC were very much dependent on the die temperature used, when compared to other types of melt. Lower die temperatures tended to achieve lower critical shear stress values for the onset of melt distortion. The wall shear rates used were 2.8, 5.6, 11.1, 16.7, and 27.8 s^{-1}, which were achieved by varying piston speeds during extrusion.

RESULTS AND DISCUSSION

Extrudate Swell Under a Magnetic Field With COP Direction to Flow

Figures 3–7 show the extrudate swell ratio (B) for various thermoplastic melts; namely, PS, ABS, PC, LLDPE, and PVC, respectively, as a function of the wall shear rate and magnetic flux densities (ranging from 0 to 51 mT) using two different die temperatures.

Effects of Wall Shear Rate and Die Temperature. Considering the extrudate swelling ratio without magnetic field (-◇-) under the effect of the wall shear rate, it was found that the extrudate swell ratio for PS, LLDPE, and PVC melts increased with increasing wall shear rates, but this was not the case for ABS and PC melts. The differences can be explained through the melt property sensitivities of these polymers to the temperature and deformation rate changes. Shah [29] stated that viscosities of polymers with complex structures such as PC, PMMA, Nylon, PS, and ABS were more sensitive to temperature change when compared to those of polymers with simple molecular structures such as PE and PP. The viscosity changes due to temperature for different polymers can also be considered in connection with Williams, Landel and Ferry (the so-called WLF equation) theory. The WLF equation is based on the concept of free volume, the relative changes in free volume, and vis-

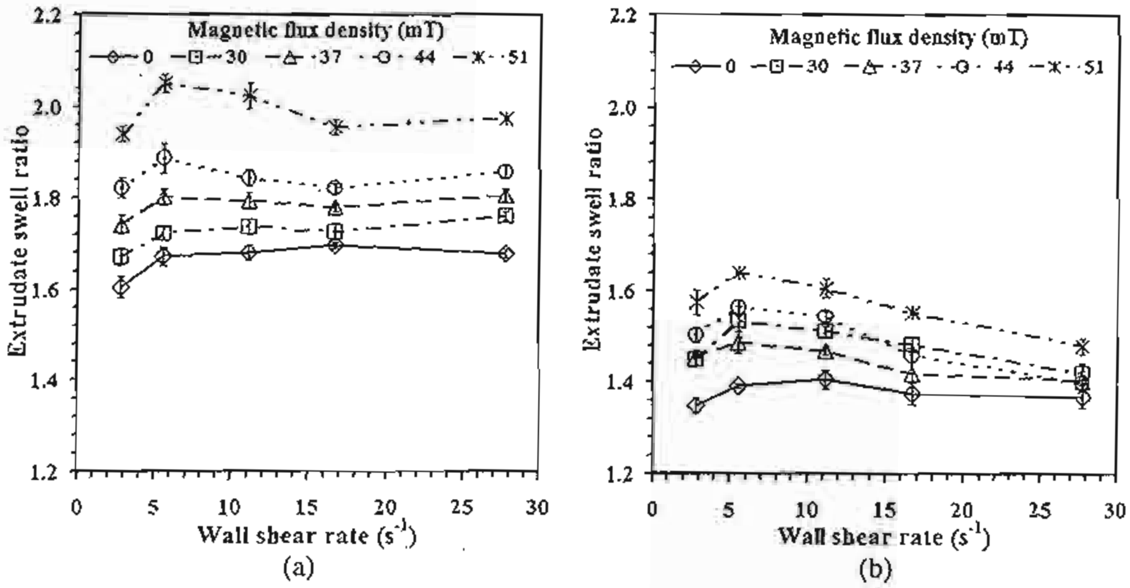


FIG. 3. Extrudate swell ratio of PS melt as a function of wall shear rate at various magnetic flux densities for two different die temperatures: (a) 175°C and (b) 215°C.

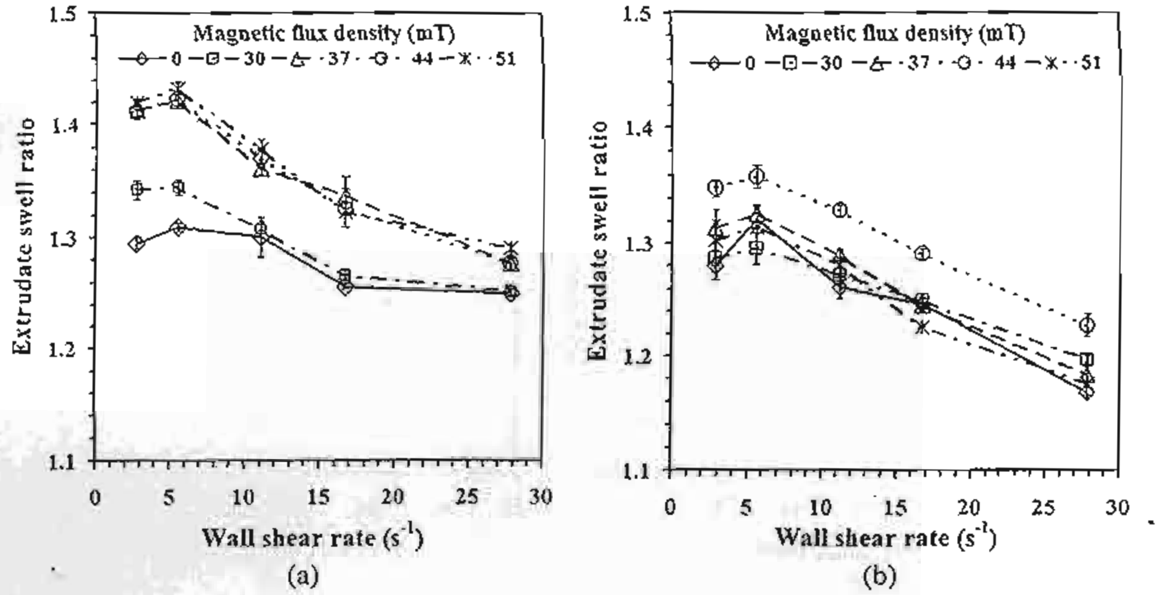


FIG. 4. Extrudate swell ratio of ABS melt as a function of wall shear rate at various magnetic flux densities for two different die temperatures: (a) 175°C and (b) 215°C.

cosity of a polymer being associated with the glass-transition temperature of the polymer [30]. These statements can be confirmed by flow curve results (wall shear stress vs. wall shear rate) for all polymers that are shown in Figs. 8–12. The flow curve results suggest that all polymer melts exhibited pseudo-plastic non-Newtonian character and their wall shear stresses for given shear rates decreased with increasing die temperature. Relating the work by Shah [29] and the WLF theory [30] to this work, it is believed that increasing the wall shear rate would result in a rise in melt temperature, as a result of the shear heating effect during the flow in the capillary die [31–33], and it is widely accepted that the temperature and shear rate have an opposite effect on the melt viscosity and the extrudate swell, the extrudate swell increasing with the shear rate and decreasing with the temperature. In this particular case, we proposed, by considering the complexity of the molecular structures and the glass-transition temperatures of the polymers as listed in Table 1, that the effect of temperature rise on the change in the extrudate swell of the PC and ABS melts was more predominant than that of the increasing shear rate, thus the decreasing extrudate swell of PC and ABS with increasing wall shear rate. For the effect of the die temperature, it was observed that the extrudate swell of all polymer melts, except for PVC melt, decreased with increasing die temperature as expected, because of increases in the viscous component and molecular relaxation

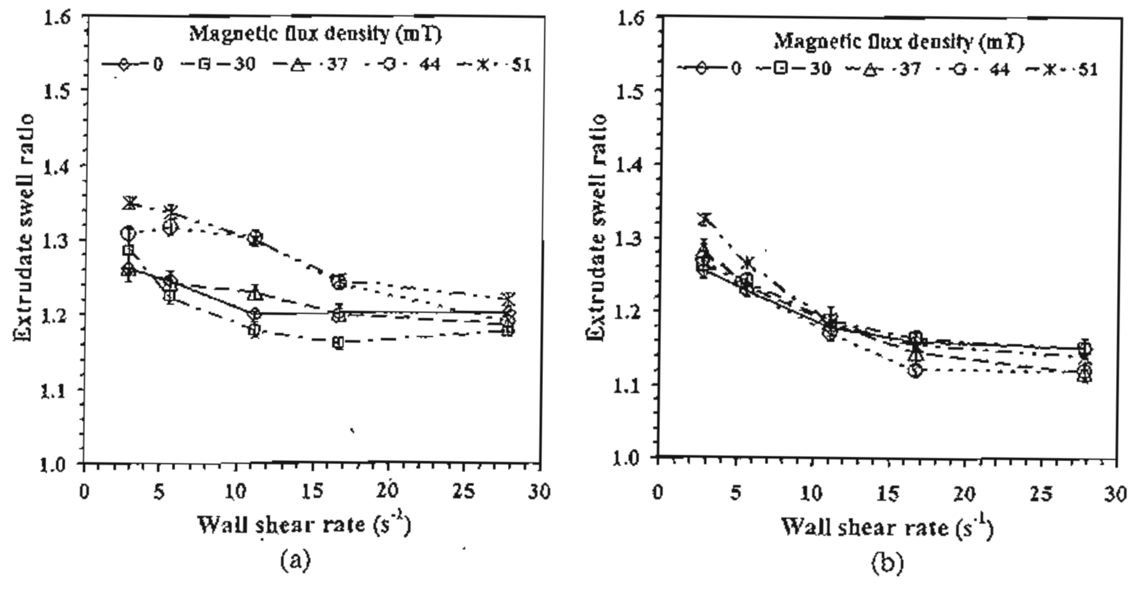


FIG. 5. Extrudate swell ratio of PC melt as a function of wall shear rate at various magnetic flux densities for two different die temperatures: (a) 230°C and (b) 250°C.

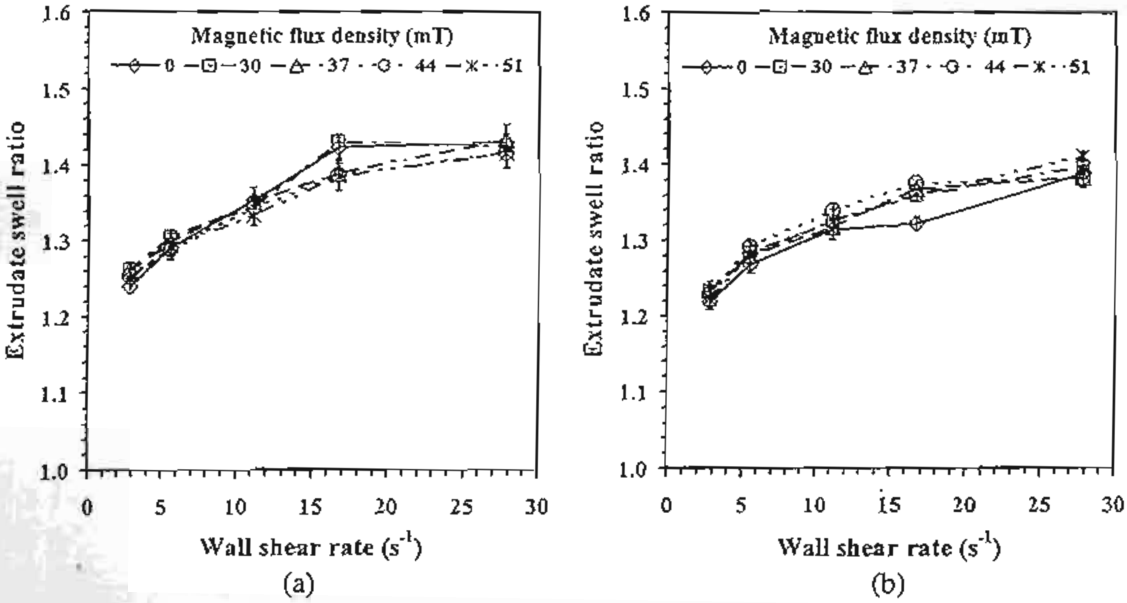


FIG. 6. Extrudate swell ratio of LLDPE melt as a function of wall shear rate at various magnetic flux densities for two different die temperatures: (a) 160°C and (b) 190°C.

of the polymer molecules. The increase in the extrudate swell ratio as a result of increasing die temperature for the PVC melt was related to a gelation effect related to irreversible entanglements of the molecules during processing, this behavior also being evidenced by a number of researchers [34, 35]. It was again found that the effect of the die temperature on the change in the extrudate swell for PC, PS, and ABS melts was more pronounced than that for the LLDPE melt.

Effect of Magnetic Flux Density. In this work, we proposed by observations from the results in Figs. 3–7 and from previous works [22, 24] that in order for a polymer melt to be induced by a magnetic field, the melts have to possess two concurrent characteristics, one exhibiting an anisotropic character and the other containing benzene groups in the molecules. A review by Kimura [20] stated that the alignment of a polymer under a magnetic field could occur only if the polymer exhibits an anisotropic characteristic, which was found for solid crystallizable polymers. However, this was not the case for the results reported in this work. We proposed that all the polymers in the molten state did not have an anisotropic structure to be induced by a magnetic field, but as they were flowing (simultaneously with the extrudate swell measurement) in the die, the molecular orientation inevitably took place, this creating an anisotropic character of the melts, and thus was

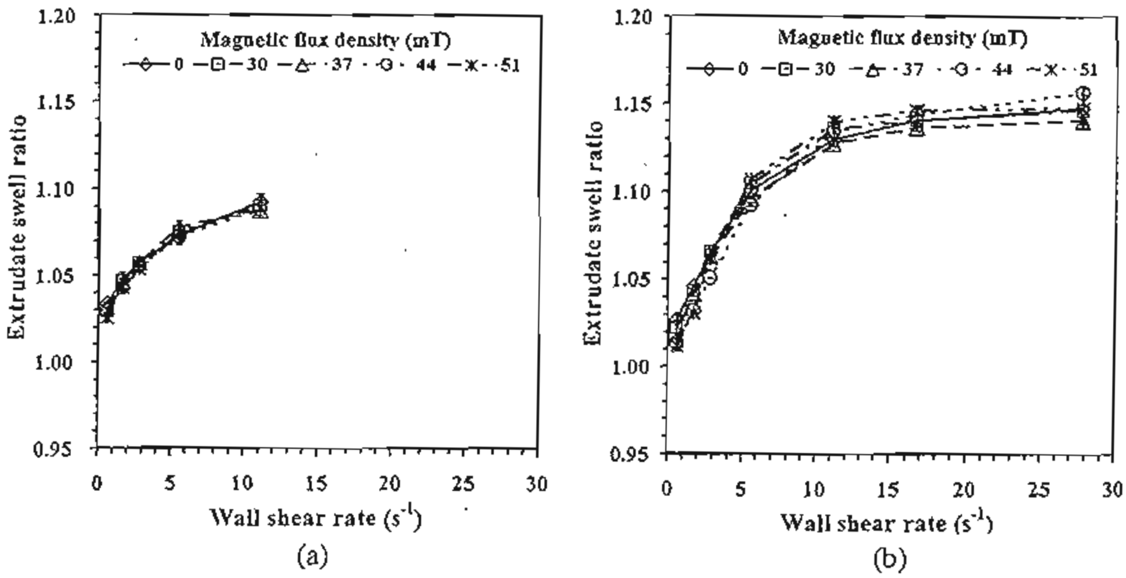


FIG. 7. Extrudate swell ratio of PVC melt as a function of wall shear rate at various magnetic flux densities for two different die temperatures: (a) 160°C and (b) 180°C.

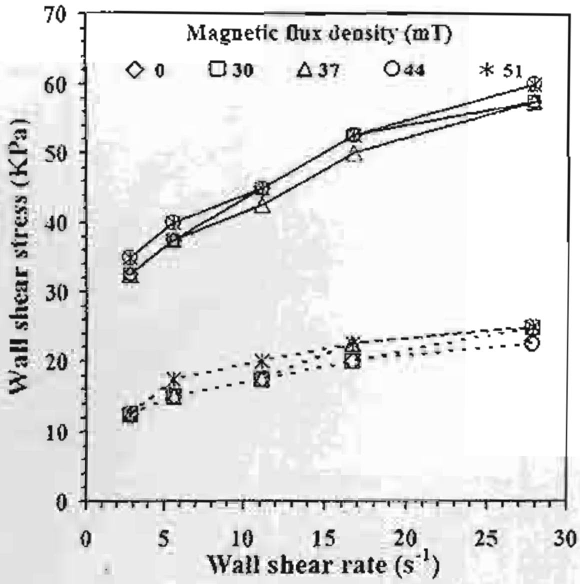


FIG. 8. Flow curves of PS melt at various magnetic flux densities for two different die temperatures: (solid) 175°C and (dashed) 215°C.

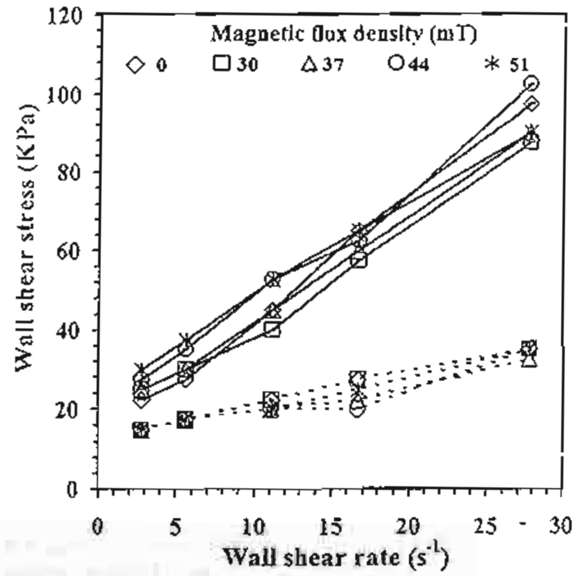


FIG. 10. Flow curves of PC melt at various magnetic flux densities for two different die temperatures: (solid) 230°C and (dashed) 250°C.

induced by the magnetic field. However, since the polymers used in this work had different molecular structures, they would react differently with the magnetic field. Figures 3–7 show that the magnetic field had an effect on the extrudate swell of PS, ABS, and PC, and the extrudate swell changes of LLDPE and PVC were independent of the magnetic field. The differences between the PS/ABS/PC and the PVC/LLDPE were associated with the benzene groups in the structures. For the PS, ABS, and PC melts, the extrudate swell tended to increase with increasing magnetic flux density. It is interesting to note that although the magnetic field had quite a pronounced effect on the extrudate swell, it seemed to have no effect on the flow curves as shown in Figs. 8–12. The mechanism for the magnetic field effect on the extrudate swell ratio of benzene-containing polymers was associated with a large diamagnetic anisotropy of the benzene molecules, which results in the magnetic torque acting on the polymer molecules [36]. The magnetic torque, occurring as a result of the motion of the existing electrons and distortions of the electron clouds within the benzene rings, can be sufficiently large to induce appreciable orientation and alignment of the molecules [24, 36, 37]. If so, the molecular configuration and alignment changes of the melts during the flow in the die would, to some extent, cause an increase in the swell ratio of the extrudate on exiting the die. This view can be substantiated by Kimura [20] and

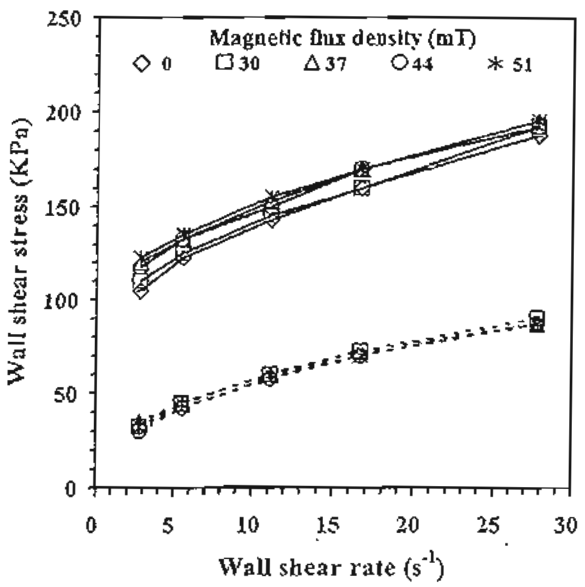


FIG. 9. Flow curves of ABS melt at various magnetic flux densities for two different die temperatures: (solid) 175°C and (dashed) 215°C.

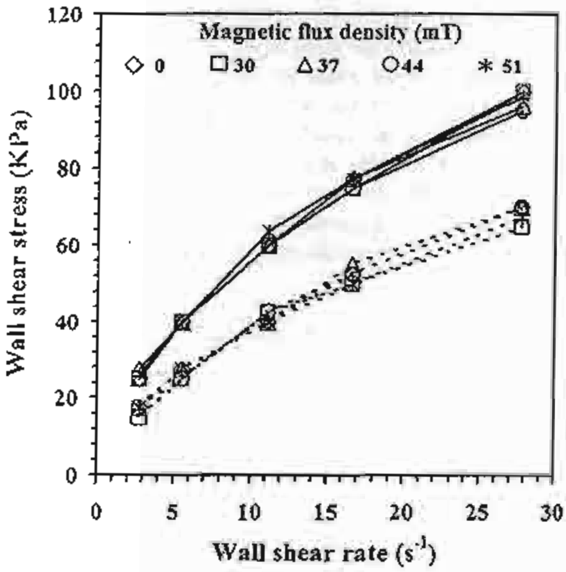


FIG. 11. Flow curves of LLDPE melt at various magnetic flux densities for two different die temperatures: (solid) 160°C and (dashed) 190°C.

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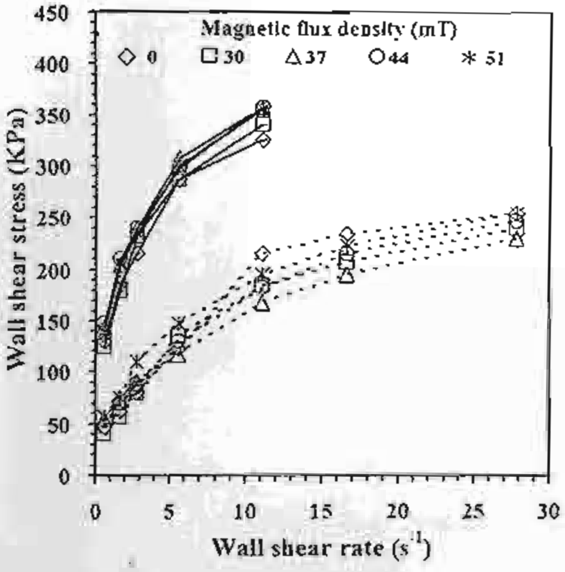

FIG. 12. Flow curves of PVC melt at various magnetic flux densities for two different die temperatures: (solid) 160°C and (dashed) 180°C.

Chandrasekhar [38] who stated that the magnetic torque acting on an anisotropic material led to a molecular rotation, and aromatic rings possessing a large diamagnetic anisotropy tend to align with the ring plane parallel to the applied field. It should also be noted that the increase in the extrudate swell ratio when using the electro-magnetized die confirmed that no additional heat had occurred from the induction effect from the solenoid coil. If heat had occurred, one would expect to observe a decrease in the extrudate swell ratio [9]. It was also observed that the magnetic effect on the extrudate swell for the PS, ABS, and PC melts was more pronounced at low shear rates and low temperatures. The changes in the extrudate swell under magnetic fields with associations to shear rate and temperature have already been discussed in our previous works [22, 24, 25].

To quantify the magnitude changes in the swelling ratio due to the magnetic field, the percentage differences (ΔB) in the extrudate swell for all thermoplastic melts for various magnetic flux densities were re-plotted in comparison with their individual extrudate swell ratio without the magnetic field, as expressed in *Eq. 1*.

$$(\Delta B) = \frac{B_m - B_n}{B_n} \times 100 \qquad (1)$$

where B_n is the extrudate swell ratio *without* magnetic field application and B_m is the extrudate swell ratio *with* magnetic field application

Figure 13 shows the percentage differences (ΔB) in the extrudate swell ratio against wall shear rate for all melts using their low die temperatures. It should be noted that the exact values of ΔB cannot be compared directly among these five polymer melts because the die temperature and shear rate ranges were different. The results were only considered in terms of the change in ΔB value as a consequence of changing magnetic flux densities. As mentioned earlier that the magnetic field affected the extrudate swell only for the PS, ABS, and PC melts, the results in Fig. 13 clearly suggest that the effect of the magnetic field on the extrudate swell decreased in the order of PS → ABS → PC. The results of the ΔB differences for these five polymer melts were now used to draw the following observations regarding the effect of molecular structures on the extrudate swell behavior (using PS melt as the comparing polymer).

1. Benzene content effect: This was explained by comparing either PS with LLDPE melts, or PS with ABS melts. It was found that the magnetic field had no effect on the extrudate swell of the melt with no benzene groups, referred to as LLDPE melt. The PS melt having higher benzene content in the molecule also showed a greater ΔB change than the ABS melt.
2. Benzene location effect: This can be discussed by comparing PS with PC melts. It was found that the magnetic field had a relatively more pronounced effect on the extrudate swell for the PS melt, which has benzene groups at the side-group location. The differences in the swell results for PS and PC melts may be associated with the molecular flexibility and the benzene ring location as they flowed in the electro-magnetic die. The benzene groups on the backbone of the PC probably made the molecular chain more rigid and difficult to align under the magnetic field and thus, a lower extrudate swell ratio.
3. Polarity and electron density effects: This was explained by comparing either PS with PVC melts, or PS with PC melts, or PS with ABS. It can be seen that the polarity did not concern the change in extrudate swell by a magnetic field. Although PVC has a higher electron density than the PS, the extrudate swell level for the PS was more induced by the magnetic field. Thus, the effect of electron density was not significant in the extrudate swell change under the magnetic field.
4. Combined effects of polarity and benzene content: This was explained by comparing either PS with ABS melts, or PS and PC melts. It can be observed that the magnetic field had less effect on the extrudate swell for the ABS melt because of lower benzene content when compared to that for the PS melt. The changes in extrudate swell ratios for both PC and ABS melts effected by the magnetic field were lower than those for PS melt, and resulted from the presence of benzene groups in the molecules rather than their polarity effects.

Effect of the Magnetic Field Direction on Extrudate Swell Ratio

The extrudate swell results for PS melt were selected for studying the effect of the magnetic field direction because the changes in the swelling ratio were comparatively more apparent when applying the magnetic field. Figure 14 shows the extrudate swell ratio for the PS melt as a function of wall shear rate at a die temperature of 175°C using two magnetic field directions, one being in a COP and the other being in a CUP directions to the melt flow direction. It can

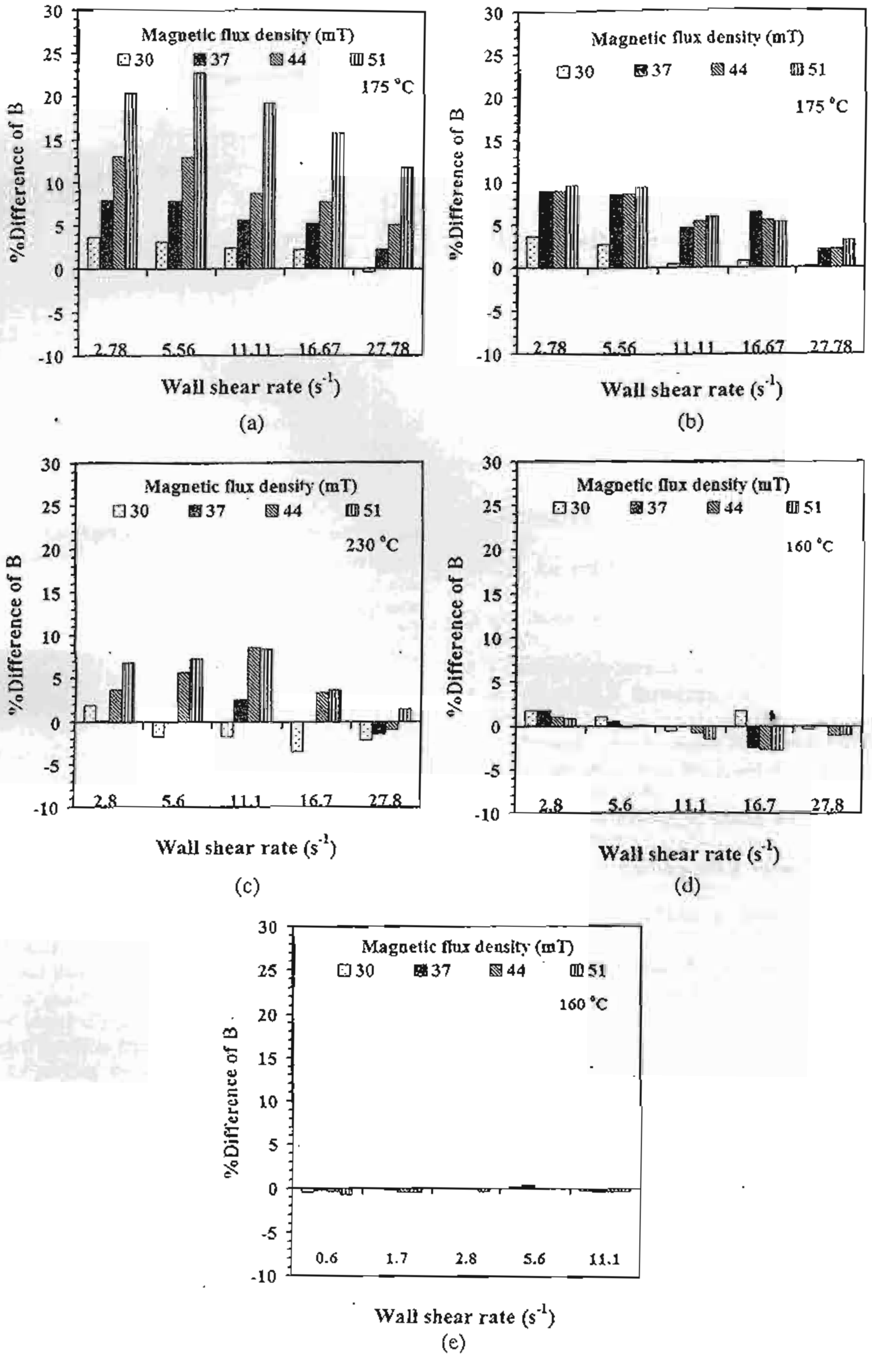

FIG. 13. Percentage differences (ΔB) in extrudate swell ratio VS shear rates at various magnetic flux densities and die temperatures: (a) PS 175°C, (b) ABS 175°C, (c) PC 230°C, (d) LLDPE 160°C, and (e) PVC 160°C.

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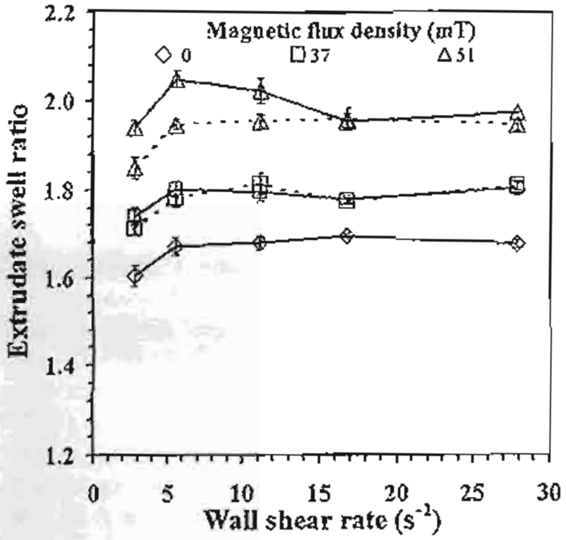

FIG. 14. Extrudate swell ratio for PS melt and wall shear rate at various magnetic flux densities using a die temperature of 175°C; COP direction (solid line) and CUP direction (dashed line).

be seen that the effect of the magnetic flux density on the extrudate swell ratio change for the CUP magnetic field system was the same as that for the COP magnetic field system, the swelling ratio increasing with increasing magnetic flux density. However, at a high magnetic flux density (51 mT), the swelling ratio for the COP magnetic field system seemed to be slightly higher than that for the CUP magnetic field system. This can be explained by a diamagnetic levitation effect [20] since the direction of the melt flow was opposite to that of the magnetic flux lines in the CUP system. For the diamagnetic levitation effect, the melt reacting with the magnetic field in the opposite direction may result in difficulties in the molecular alignment and disentanglement during the flow in the die, and thus the swelling ratio on exiting the die decreased.

In summary, the application of magnetic field to an extrusion die and the knowledge of its effect on the swelling ratio changes of polymer melts would be of great advantage in quality and dimension controls of polymer products in the extrusion process. The findings obtained in this work clearly had practical implications in that the dimensions of the extruded products from individual polymers could be adjusted by varying the magnetic flux densities without having to change any normal processing parameters (note that in practice, adjusting one processing parameter will automatically change other parameters, and this makes the process more difficult to control).

CONCLUSIONS

In this work, a relationship between the molecular structure and extrudate swell ratio of five different thermoplastic melts under a magnetic field during extrusion in a rate-constant capillary rheometer was examined. The results suggested that the extrudate swell ratio for PS, LLDPE, and PVC melts increased with increasing wall shear rates, but the opposite effect was found for ABS and PC melts. The extrudate swell ratio for the PS, ABS, PC, and LLDPE melts decreased with increasing die temperature while that for PVC melt increased with die temperature because of a gelation effect. Thermoplastic melts having high benzene content in the side-chain and exhibiting an anisotropic character appeared to be affected strongly by the magnetic field, causing an increase in the extrudate swell ratio. The magnetic field had a significant effect on the extrudate swell ratio of the PS, ABS, and PC melts, the extrudate swell ratio of the LLDPE and PVC melts being independent of the magnetic field. It was observed that the effect of the magnetic field on the swelling ratio decreased in the order of PS → ABS → PC. The magnetic field direction had no pronounced effect on the changing trend of the extrudate swell ratio for all melts at low magnetic flux densities. At a high magnetic flux density (51 mT), the extrudate swell ratio for the COP magnetic field system was slightly higher than that for the CUP magnetic field system.

REFERENCES

1. K.K. Kar and J.U. Otaigbe, *J. Elastomers Plast.*, **33**, 297 (2001).
2. J.C. Huang and K.S. Leong, *J. Appl. Polym. Sci.*, **84**, 1269 (2002).
3. J. Guillet and M. Seriai, *Rheol. Acta*, **30**, 540 (1991).
4. K. Wilczynski, Z. Szymaniak, and A. Nastaj, *Polimery*, **48**, 204 (2003).
5. B.J. Yarusso, *J. Non-Newtonian Fluid Mech.*, **40**, 103 (1991).
6. K.J. Christodoulou, A.K. Wood, and N. Sombatsompop, *SPE ANTEC*, **44**, 915 (1998).
7. H. Munstedt, M. Schwetz, M. Heindl, and M. Schmidt, *Rheol. Acta*, **40**, 384 (2001).
8. K. Matsunaga, T. Kajiwara, and K. Funatsu, *Polym. Eng. Sci.*, **38**, 1099 (1998).
9. P.L. Swan, A. Garcia-Rejon, A. Derdouri, J.M. Dealy, *SPE ANTEC*, **36**, 1607 (1990).
10. S. Eggen, E.L. Hinrichsen, *Polym. Eng. Sci.*, **36**, 410 (1996).
11. N. Sombatsompop, R. Dangtungee, *J. Appl. Polym. Sci.*, **82**, 2525 (2001).
12. N. Orbey and J.M. Dealy, *Polym. Eng. Sci.*, **24**, 511 (1984).
13. C.Y. Ma, J.L. White, F.C. Weissert, A.I. Isayev, N. Nakajima, and K. Min, *Rubber Chem. Technol.*, **58**, 815 (1985).
14. K.C. Shin, J.L. White, and N. Nakajima, *J. Non-Newtonian Fluid Mech.*, **37**, 95 (1990).
15. D.G. Kiriakidis and E. Mitsoulis, *Adv. Polym. Technol.*, **12**, 107 (1993).
16. S. Tanoue and Y. Iemoto, *Polym. Eng. Sci.*, **39**, 2172 (1999).
17. W.S. Lee and H.Y. Ho, *Polym. Eng. Sci.*, **40**, 1085 (2000).
18. N. Sombatsompop, N. O-Charoen, *Polym. Adv. Technol.*, **14**, 699 (2003).
19. Y.R. Cao and H.L. Li, *Polym. Eng. Sci.*, **42**, 1534 (2002).
20. T. Kimura, *Polym. J.*, **35**, 823 (2003).
21. N. Sombatsompop, *J. Appl. Polym. Sci.*, **86**, 509 (2002).

22. N. Sombatsompop, S. Sergsiri, *Polym. Adv. Technol.*, **15**, 472 (2004).
23. N.-T. Intawong and N. Sombatsompop, *Polym. Eng. Sci.*, **44**, 1960 (2004).
24. N.-T. Intawong and N. Sombatsompop, *Polym. Eng. Sci.*, **44**, 2298 (2004).
25. N. Sombatsompop and N.-T. Intawong, *Polym. Adv. Technol.*, **16**, 505 (2005).
26. N-T. Intawong, T. Kimura, M. Tamura, L. Xiaojun, and N. Sombatsompop, *Polym. J.*, **37**, 541 (2005).
27. R.D. Chien, N.R. Jong, and S.C. Chen, *J. Micromech. Microeng.*, **15**, 1389 (2005).
28. N. Sombatsompop and N.-T. Intawong, *Polym. Test.*, **20**, 97 (2001).
29. P.L. Shah, "Rheology" in *Handbook of Plastic Materials and Technology*, I. Rubin, Ed., Wiley, New York (1990).
30. R.B. Bird, R.C. Armstrong, and O. Hassager, *Dynamics of Polymeric Liquids*, Wiley, New York (1987).
31. N. Sombatsompop and S. Patcharaphun, *Polym. J.*, **33**, 491 (2001).
32. L.P. Tan, S.C. Joshi, C.Y. Yue, Y.C. Lam, X. Hu, and K.C. Tam, *Acta Mater.*, **51**, 6269 (2003).
33. P. Wapperom and O. Hassager, *Polym. Eng. Sci.*, **39**, 2007 (1999).
34. N. Sombatsompop and S. Thongsang, *J. Appl. Polym. Sci.*, **82**, 2478 (2001).
35. M.A. Huneault, P.G. Lafleur, and P.J. Carreau, *Plastics Engineering*, **39**, September (1989).
36. P.C.M. Christianen, I.O. Shklyarevskiy, M.I. Boamfa, and J.C. Maan, *Phys B: Condens Matter*, **346/347**, 255 (2004).
37. K.R. Amundson, "Electric and Magnetic Field Effects on Polymeric Systems Exhibiting Long-Range Orientational Order," in *Electrical and Optical Polymer Systems*, D.L. Wis, Ed., Marcel Dekker, New York (1998), Chapter 32, pp 1079–1139.
38. C. Chandrasekhar, *Liquid Crystals*, Cambridge University Press, London (1992).

DOI 10.1002/pen