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THE EFFCT OF ASYMMETRIES OF DIE EXIT GEOMETRY ON EXTRUDATE SWELL AND MELT FRACTURE

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Abstract – Experimental investigations were performed to see how the die exit geometry and the extrusion velocity influence on extrudate swell and melt fracture for several polymer melts [low-density polyethylene, styrene-butadiene rubber (SBR) and SBR/HAF (carbon black) compound]. Four different types of die exit geometry were considered; 0° (symmetric, usual capillary die), and 30°, 45° and 60° (asymmetric dies) were chosen for the die exit angle. Extrudate diameters were measured without draw-down under isothermal condition. Polymer melts were extruded into an oil that has the same density and temperature as those of the extrudate. Extrudate swells from dies with different diameters were correlated with volumetric flow rates. It was observed that the extrudate swell increases with increasing volumetric flow rate and exhibits through a minimum value at about 45° die exit angle. As to the fracture phenomena, it was observed that the critical shear for the onset of melt fracture increases with the increasing die exit angle up to 45°. However, for 60° die exit angle, the onset of melt fracture is again similar to that of 0° exit angle.

INTRODUCTION

Dimensional stability and extrudate distortion are two major problems of polymer extrusion. The former is related to the sectional increase of viscoelastic materials at the exit of extrusion dies, the so-called extrudate swell. The latter is the technical limit of extruder output and extrudate roughness will appear above a certain critical rate, which is called as the melt fracture phenomena.

It is well known that if a viscoelastic fluid is extruded from a die into air without substantial drawing, the cross-sectional area of the extrudate will usually exceed the die exit. It is usually called extrudate swell. For the circular die commonly used, diameter ratios (extrudate to die) are often observed in the range of 2 to 3.

Extrudate swell was first discussed by Spencer and Dillon [1] in connection with flow behavior of molten polystyrene. Their analysis was criticized by Mooney [2] who noted that "the type of shearing deformation they assume to exist at exit would only lead to telescopic elastic deformation after exit, not to longitudinal contraction and diameter swell". This difficulty was overcome by Nakajima and Shida [3] to consider that the completely recovered (swollen) extrudate could be pulled lengthwise until its diameter became equal to the diameter of the capillary.

Because of the significance of extrudate swell both in polymer characterization and in polymer processing it is worthwhile to adopt a somewhat simplified approach to the problem. The question of whether steady state has been reached or not in the capillary may not be significant.

Most of the published data on swell were obtained using capillary dies; these observations have been summarized by Han [4]. Recently some attention has been focused on slit dies, too (Huang and White [5, 6]).

Whenever a polymer melt is extruded at high shear rates and stresses, the extrudate becomes distorted. This phenomenon is referred to as melt fracture, or melt instability. There are numerous papers in the literature devoted to find mechanism responsible for this and to determine critical conditions which may depend upon die entry geometry, temperature, and molecular weight. There have been also reported a lot of flow visualization studies aimed at determining the initiation stage for instability with melts. Despite of the huge amount of experimental and theoretical

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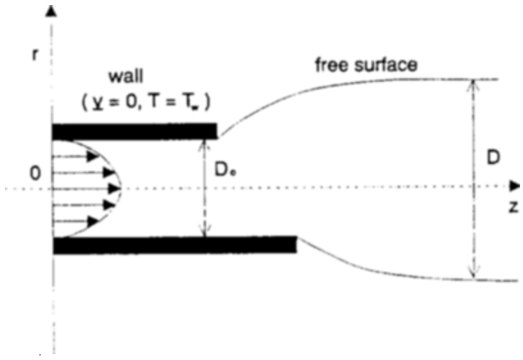

Fig. 1. Definition sketch of our extrusion system.

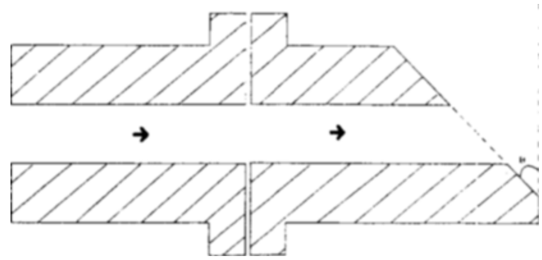

Fig. 2. Cross-sectional view of typical capillary die and adapter.

works, we still do not have a clear understanding of this phenomenon.

Many attempts have been devoted to develop a proper model to predict the extrudate swell. They can be broadly classified as either simplified approaches based on the theory of rubber elasticity or numerical ones based on a detailed flow analysis. The former method can predict an elastic, instantaneous swell but cannot provide any information on its time dependency. In addition, it will not give a reliable prediction at high shear rate. On the other hand, numerical simulation of extrudate flow can provide a detailed picture of swell, but it has been limited due to the onset of a serious numerical instability at high flow rates. It is fair to say for the present that there is no reliable general model to predict the true extrudate swell, so that a well-designed experiments should be performed to obtain reliable informations on the swell behavior.

Since the swelling is a result of molecular rearrangement from the orientation developed during the die flow, it would be reasonable to assume that geometry of dies has an importance on swelling behavior. In this work, the effect of die exit geometry will be experimentally considered on extrudate swell and melt fracture. Such experiments have not been done as far as we know.

EXPERIMENTS

Fig. 1 shows a sketch of an asymmetric die swell in which final extrudate diameter D_o is considerably larger than die diameter D. It shows an inherently different exit geometry from usual, axisymmetric circular die's. Due to this asymmetry, relaxation pattern of the extrudate will be considerably different from that of a symmetric die. It will be assumed that the flow is fully developed far upstream of the exit region.

Table 1. Dimensions of capillary die adapters

Diameter (mm)	Length (mm)	L/D	Exit angle (deg.)
0.8	16.8	21.00	0°, 30°, 45°, 60°
1.2	17.4	14.50	0°, 30°, 45°, 60°
1.3	19.8	15.23	0°, 30°, 45°, 60°

The extrudate swell ratio is defined as

$$B = D/D_o. \qquad (1)$$

Four different types of die adapters were prepared and attached at the end of Instron capillaries. These were; 0° (typical flat), 30°, 45° and 60° for die exit angle (θ) as shown in Fig. 2. Their dimensions are listed in Table 1.

Materials chosen for the experiments are three, one from typical thermoplastic polymers and the others from rubber and rubber compounds. Low density polyethylene (Han-Yang Chemical Co., LDPE5301), styrene-butadiene rubber (Kumho Synthetic Rubber Co., SBR1500) and SBR1500/HAF (50 phr) are those. LDPE5301 was chosen because it has a low melt index (MI = 0.28), which is expected to have a relatively large extrudate swell. SBR1500 is a synthetic rubber which is used commonly for the truck tread. LDPE 5301 was supplied by the manufacturer in a granular form, and SBR1500 in a bulk form.

Standard HAF carbon black was added to SBR1500 as a filler. Carbon black was sieved in order to obtain reasonably uniform particle size distributions (ASTM No. 30) and dried in an oven at 90°C for 4 hours before compounding. SBR/HAF compound was prepared in a laboratory Banbury, operating at 60 rpm and cooled with running water. The standard mixing cycle consisted of kneading the SBR alone for 1 min prior to addition of carbon black, followed by 3 min of mixing with carbon black.

LDPE, SBR and SBR/HAF were rheologically characterized by measuring their shear viscosities as

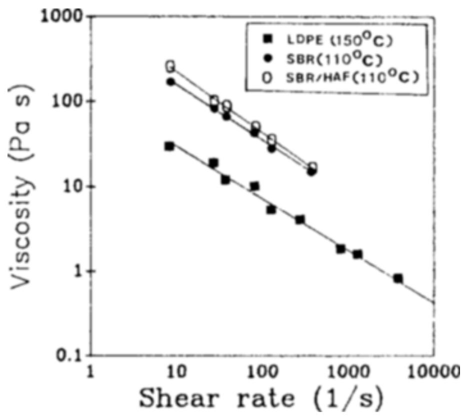

Fig. 3. Log viscosity vs. log shear rate for LDPE5301 at 150°C, SBR and SBR/HAF compounds at 110°C.

shown in Fig. 3. The shear viscosity of LDPE5301 was measured at 150℃ since melting temperature of LDPE 5301 is 123℃. 110℃ was chosen for SBR and SBR/HAF. These log viscosity-log shear rate curves exhibit typical power law behavior in the range of shear rate under consideration. These pseudoplasticity may be attributed to disentanglement and orientation of polymer chains at high shear rates. At low shear rates, the thermal movement of the segment of polymer chain predominates over molecular orientation effect. At a fixed shear rate the SBR/HAF compound exhibits high viscosity than that of a raw SBR as expected since fillers, being non-deformable particles, have infinitely large viscosities compared to that of suspending medium. Contribution of carbon black on the viscosity of SBR/HAF compounds decreases as shear rate increases.

In order to obtain reliable die swell data, experiments should be performed under the following conditions: (1) steady, isothermal state; (2) absence of gravitational sagging and swelling due to interfacial tension; (3) the final extrudate should be in a completely recovered state from elastic deformations.

There exist in the literatures two different types of measurements for die swell. In the first, the extrudate is quenched in air or liquid; in the second, the polymer is extruded into a heated chamber and relaxed, and then the dimensions are measured. It is hard to tell whether extrudate is in a completely recovered state from elastic deformations. The simplest way is to measure the diameter of an extrudate 1/4 in. above its free end as employed by a number of workers (Roger [7], Vlachopoulos, Horie and Lindorikis [8], Arai and Aoyama [9]). Nakajima and Shida [3] measured the time variations of diameter after relaxing 1.5 in. long strands of extrudate suspended in an oven. Mendelson et al. [10] relaxed such strands in an oil bath and measured the diameter of a cold sample with a micrometer. It has been demonstated that final diameter of the extrudate does not depend on the temperature of relaxation. Cogswell [11] quenched rod (or disk) shaped extrudates in a cold water first and then these samples were allowed them to relax in a hot oven later. Extrusion into a heated gas chamber was described by Petraglia et al. [12] and Han and co-workers [13, 14]. However, neither of these two methods, quenching in a cold water or extruding into a hot chamber, can satisfy three conditions necessary for reliable measurements of extrudate swell. Quenching introduces an additional thermal history, which in the case of crystallizable polymers introduces additional stresses (Mendelson et al. [10]). Extrusion into a hot gas chamber increases the effects of gravitational sagging and interfacial tension.

Instead, extrusion of polymers into an inert thermostatic fluid with proper density and interfacial tension seems to satisfy these three basic requirements. The experimental apparatus employed here to obtain the swell data was similar to one which was originally designed by Utracki et al. [15]. Extrusion was accomplished by means of an Instron capillary rheometer. A thermostatic barrel having twice the inner diameter of the standard Instron barrel was designed to accomodate the larger dies used. The die adapters and clamping nut were designed in such a way that the tip of the die is visible, enabling one to observe the extrudate from the moment it emerges at the die tip. The extrudates extruded directly into oil having the same temperature and nearly the same density as the extruded melt. Thus it was possible to measure swell in the absence of sagging and solidification. Windows in the transparent oil bath allowed us to measure the extrudate diameter by using camera.

EXPERIMENTAL RESULTS

The effects of die exit geometry on extrudate swell for each resin are shown in Figs. 4-6, where the equilibrium extrudate swell data for three materials are plotted against volumetric flow rate for capillary diameter D = 1.3 mm. In the case of other capillaries with D = 1.2 mm and D = 0.8 mm, similar behaviors were observed. Using the shear rate at the wall as a measure of extrusion rate has been customary in interpreting swell data. However due to the difficulties associated with determining the norminal shear rate at

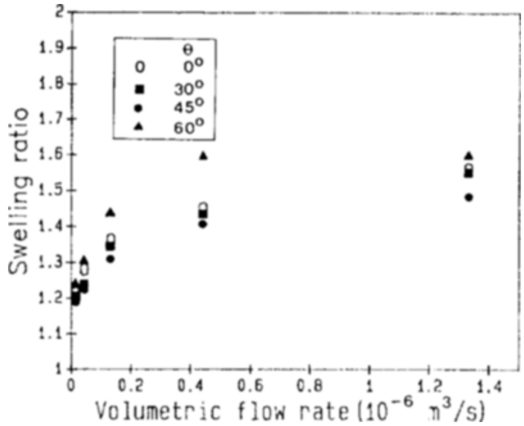

Fig. 4. Exrudate swell ratio vs. volumetric flow rate for LDPE5301 extruded in various die adapters (D= 1.3 mm) at 150°C.

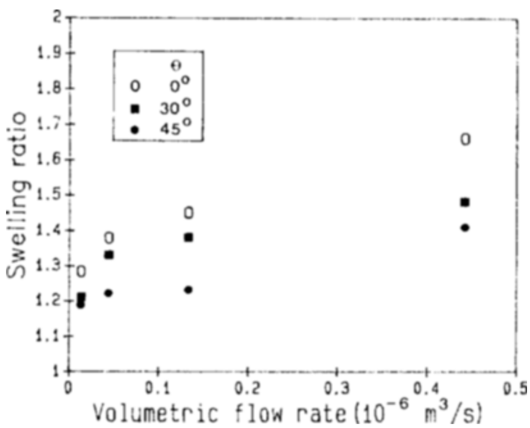

Fig. 5. Extrudate swell ratio vs. volumetric flow rate for SBR1500 extruded in various die adapters (D= 1.3 mm) at 110°C.

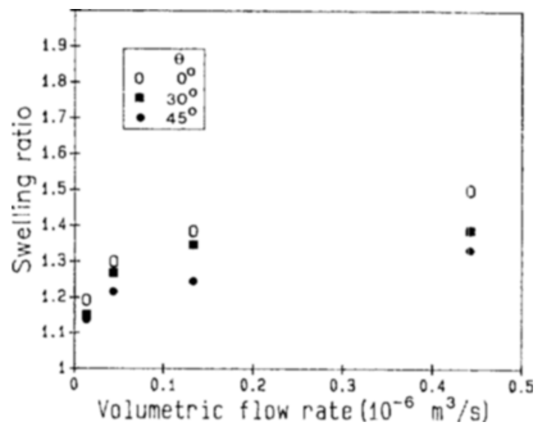

Fig. 6. Extrudate swell ratio vs. volumetric flow rate for SBR/HAF compound extruded in various die adapters (D= 1.3 mm) at 110°C.

the wall in complex die geometries used in this study, volumetric flow rate was chosen as the independent flow variable here.

It can be easily understood that die flow causes partial orientation in the axial direction due to the shearing deformation of a fluid element. Orientation in the axial direction would be expected to produce a swelling behavior which was isotropic in a plane normal to the flow axis. Then, because of asymmetry this isotropic reaction is not accepted at the die exit. So the complex swelling mechanism is involved in extrudate. As shown in Figs. 4-6, extrudate swelling increased with volumetric flow rate, since it is the manifestation of the recovery by the material of elastic strains acquired during extrusion. Since increasing extrusion rate results in shorter capillary residence times, the emerging material more clearly remembers its state prior to capillary entry and consequently results in greater extrudate die swell. Indeed, these have been observed for both thermoplastics (LDPE 5301) and rubbers (uncompounded SBR and compounded SBR) which generally exhibit similar behaviors.

For the 30° and 45° die adapters, the extrudate swell was somewhat reduced compared with that for the flat (0°) die adapters. However the 60° die adapter, with sharp slope of exit angle, produced much higher extrudate swell than any others. It should be noted that swell passes through a minium value at about 45° exit angle. The inlet geometry was kept constant for all measurements. The characteristic times for swelling processes are much longer than those associated with relaxation processes in simpler shear flow. On the other hand, the time required to reach a steady state stress at the onset of steady simple extension is much longer. It may be relevant to swell dynamics, as it is now recognized that extrudate swell involves considerable stretching and compression along streamlines. It seems that the effect of die exit angle on swelling behavior is similar to these swelling mechanisms in terms of stretching of the melt in the die exit. The effect of die exit angle on swelling behavior can be understood in terms of the time gap of relaxation, stretching to axial direction and flow pattern change in inner capillary of the melt. In the 30° and 45° dies, a strong stretching works axial orientation to cause a substantial reduction in extrudate swell but because of high slope of exit angle at 60° die, this

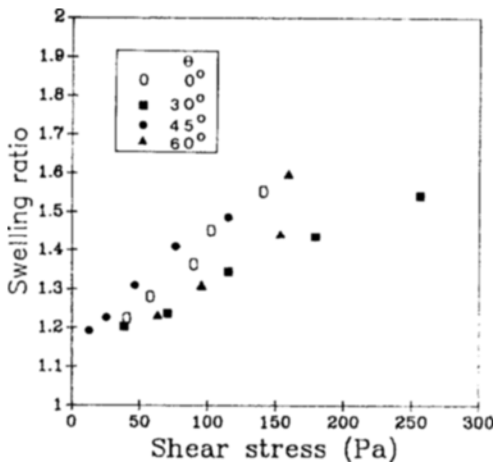

Fig. 7. Swell ratio as a function of shear stress for various exit angle at 150°C (D=1.3 mm).

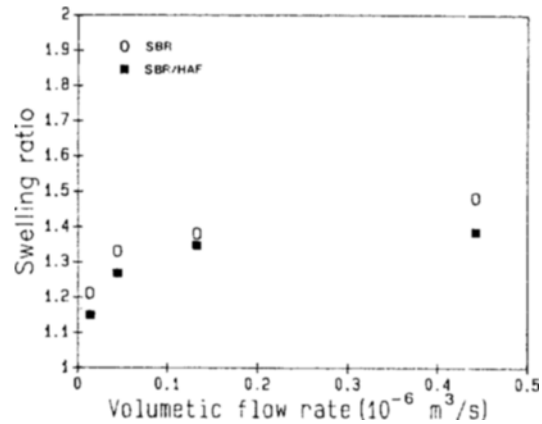

Fig. 9. Effect of carbon black on swell ratio of SBR and SBR/HAF compound for θ=30° at 110°C (D=1.3 mm).

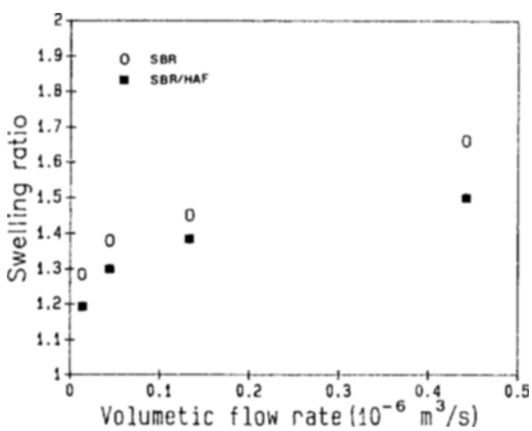

Fig. 8. Effect of carbon black on swell ratio of SBR and SBR/HAF compound for θ=0° at 110°C (D=1.3 mm).

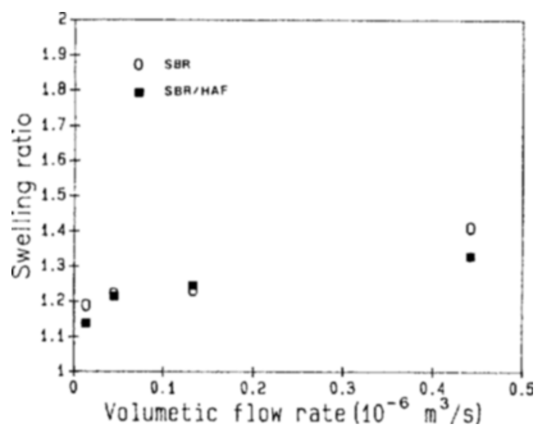

Fig. 10. Effect of carbon black on swell ratio of SBR and SBR/HAF compound for θ=45° at 110°C (D=1.3 mm).

interpretation is not applicable.

Also one of the interesting topics is the relationship between shear stress and extrudate swell. Fig. 7 shows equilibrium value of extrudate swell as function of wall shear stress, measured from the Instron capillary rheometer. As shown here the swell is a linear function of shear stress for various die exit angles. Whereas all modified dies exhibited very similar extrudate swelling behavior. There is a jump in the extrudate curve of LDPE5301 for θ=60° at around 170 Pa shear stress. This phenomenon is commonly associated with melt fracture and is sometimes referred to as slip flow. This discontinuity does not occur for branched polymers. Experiments with different dies lead to the suggestion that slip at the die wall may result in melt fracture.

Extrudate swell was reduced by adding carbon black in SBR and its reduction was promoted as volumetric flow rate is increased as shown in Fig. 8. The reason is that carbon black decreases the elasticity of the rubber and consequently reduce the extrudate swell. For a 30° die adapters, the presence of carbon black effect is still there (Fig. 9) but situation is changed for 45° die adapter as shown in Fig. 10. Contribution due to the adapter is too large to show additional reduction of swell due to the carbon black.

The effect of die exit geometry on extrusion defect has been also specifically studied. The die exit angle

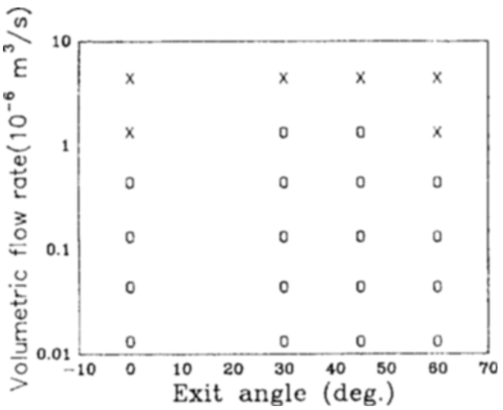

Fig. 11. Effect of die exit angle on melt fracture region of LDPE5301 for D=1.3 mm at 150°C (○: no melt fracture, ×: melt fracture).

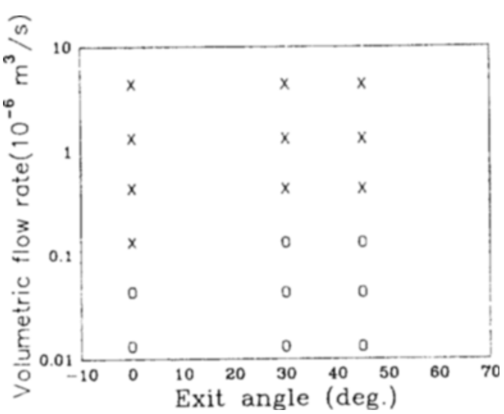

Fig. 12. Effect of die exit angle on melt fracture region of SBR for D=1.3 mm at 110°C (○: no melt fracture, ×: melt fracture).

is a main factor affecting the melt fracture as shown in Figs. 11-13. The critical shears increase with increasing die exit angle (θ) from 0° to 45°. It means that the melt fracture at this range does not occur until some high extrusion rate or shear rate and the polymer processing is possible at high flow rate. When the shear exceeds a certain critical value, the extrusion output of polymeric materials increases extremely and extrudate distortion occurs. The rod surface and section become uneven and the distortion increases severely with shear. It has been known that the flow in the die entrance zone is responsible for extrusion instabilities. When the material converges towards the entrance, flow lines are disturbed. As the shear stress increases, the imposed deformation rates exceed the relaxation of the polymer, and then the stress can exceed the material resistance to result in a rupture. It is customary to feed the die in the middle of the barrel or in the stagnation zones in order to reduce the flow disturbances.

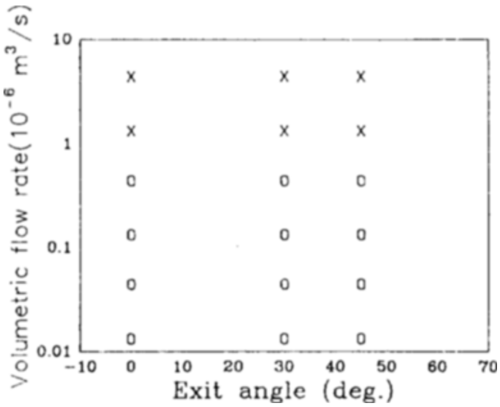

Fig. 13. Effect of die exit angle on melt fracture region of SBR/HAF for D=1.3 mm at 110°C(○: no melt fracture, ×: melt fracture).

It seems that exit geometry has considerable effect in our system and the distortion severity also decreases with the increase of the exit angle. Our experimental data are, however, still insufficient to establish the above observation as a general rule. The critical shear for the onset of extrudate defect (melt fracture) increases with both adding carbon black and increasing die exit angle to 45°. However in the case of die exit angle 60°, the onset of extrudate defect is similar to that of the case of 0° exit angle. From above experimental data, it may be suggested that presence of carbon black are able to interact or aggregate in such a way to produce gel-like structure or apparent gel-like structure which can resist the high stress levels before flow disturbance begins. The gel can be easily reformed when stresses are removed. It also explains the reduction in extrudate swell in the highly filled system.

CONCLUSIONS

The main factors affecting post-extrusion swell and the critical conditions for the onset of extrudate distortion (melt fracture) are experimentally studied for asymmetric exit geometries of dies. The effect of die exit angle on extrudate swell behavior can be understood in terms of the time gap of stretching of the melt in the die exit. Swell passes through a minimum value at about 45° exit angle when the same volumetric flow rate is maintained. It seems that the stretching of the melt in the die exit is dominant for 45° exit angle. Extrudate swells are increased with volumetric flow rate. Die exit angle was found to be one of the main factors affecting the melt fracture. Onset of melt fracture increases with the increase of the die exit angle up to 45°. The critical shear level depends not only upon the exit geometry but also upon the amount of carbon black added. However, the available data is still insufficient to establish the observation as a general rule.

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REFERENCES

1. Spencer, R. S. and Dillon, R. E.: *J. Colloid. Sci.,* **3**, 163 (1948).
2. Mooney, M.: "Rheology", Vol. 2, Chap. 5, edited by Eirich, F. R., Academic Press, New York (1967).
3. Nakajima, N. and Shida, M.: *Trans. Soc. Rheol.,* **10**, 299 (1966).
4. Han, C. D.: "Rheology in Polymer Processing", Academic Press, New York (1976).
5. Huang, D. C. and White, J. L.: *Polym. Eng. Sci.,* **19**, 609 (1979).
6. Huang, D. C. and White, J. L.: *Polym. Eng. Sci.,* **20**, 182 (1980).
7. Rogers, M. G.: *J. Appl. Polym. Sci.,* **14**, 1679 (1970).
8. Vlachopoulos, J., Horie, M. and Lindorikis, S.: *Trans. Soc. Rheol.,* **16**, 669 (1972).
9. Arai, T. and Aoyama, H.: *Trans. Soc. Rheol.,* **7**, 333 (1963).
10. Mendelson, R. A., Finger, F. L. and Bagley, E. B.: *J. Polym. Sci.,* **c35**, 177 (1971).
11. Cogswell, F. N.: *Plastics and Polymers,* 391 (1970).
12. Petraglia, G. and Coen, A.: *Polym. Eng. Sci.,* **10**, 79 (1970).
13. Han, C. D. and Charles, M.: *Trans. Soc. Rheol.,* **14**, 213 (1970).
14. Han, C. D. and Yu, T. C.: *AIChE J.,* **17**, 1512 (1971).
15. Utracki, L. A., Bakerdjian, Z. and Kamal, M. R.: *J. Appl. Polym. Sci.,* **19**, 48 (1975).