Flow Visualization and Extrudate Swell of Natural Rubber in a Capillary Rheometer: Effect of Die/Barrel System

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ABSTRACT: An investigation was carried out to examine the effect of die/barrel system on the flow patterns and extrudate swell of natural rubber in the barrel of a capillary rheometer, using a colored tracer as the visualization technique. The capillary rheometer used in this work had two dies located along the barrel, which is novel in rheometer design. The flow of the rubber in the upper barrel was dependent on the piston/barrel action and changed with piston displacement, whereas the complexity of the flow in the lower barrel was dependent not only on the piston displacement, but also on the geometry of the upper die design. The flow patterns that developed in the whole barrel were independent of the die located at the bottom of the barrel. In addition, the change in extrudate swell was associated with the flow occurring in the barrel, residence time, elastic characteristic, and the temperature rise during the flow. It was concluded that the general style of the flow patterns of natural rubber was greatly dependent on the die geometry that the material had previously moved past. © 2001 John Wiley & Sons, Inc. J Appl Polym Sci 82: 2525–2533, 2001

Key words: flow visualization; die design; capillary rheometer; die swell; natural rubber; rheology; rubber; swelling; processing; extrusion

INTRODUCTION

In polymer extrusion, it is widely accepted that to develop or optimize existing machine technology and dies it is necessary to have precise knowledge of the flow properties and flow patterns of polymer melts in the process. The properties of the end-product are very dependent not only on the materials used, but also on the design of the processing equipment. Unstable flows occurring during processing can result in low-quality products. Complex flows are due to the design of the equipment, the material characteristics, the pressure, and the temperature.[1] The most common technique used for the determination of the flow properties of polymer melts is that of the capillary rheometer. The flow property results produced in the capillary rheometer depend on the design of the apparatus, with the same die giving different results when used in different apparatus designs.[2] The differences in the results are associated with the flow patterns.[2] As a consequence, studies on flow patterns of polymer melts in the capillary rheometer have been widely carried out.[1–4]

Ma et al.[5] studied the flow patterns of various elastomers in the entrance region of a circular die with a wide range of geometries (converging and diverging entrance dies, and 180° entrance angle dies with off-center hole and with double hole). The results indicated that the elastomers exhib-

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ited streamline flow into the entrance in all cases except degraded rubber, which gave evidence of vortices in the die corners. Song et al.[6] conducted flow marker experiments of various rubber compounds, including natural rubber (NR), styrene butadiene rubber (SBR), and ethylene propylene-diene monomer (EPDM), in the barrel of a capillary rheometer using a wide range of die designs. They found that the radial flow simply moved inward to the capillary die as the ram moved down the barrel, with no evidence of secondary flows. Sombatsompop et al.[1,4] investigated the flow patterns of NR compounds developed inside the barrel and the capillary of a rheometer with a pigmented rubber compound and found that the flow patterns in the barrel of the capillary rheometer were complex and a function of piston displacement, with the general style of the flow patterns being independent of die design (size and die entry angle). The complex flows resulted from material adjacent to the piston that was used to extrude the polymer, with the surface flowing across the face of the piston and down the center of the barrel. Although the die design did not affect the general style of the flow patterns in the barrel, it influenced the flow patterns in the die; that is, the flow patterns in the die were linked with the occurrence of flow irregularities along the die and melt fracture at the die exit.[4] This phenomenon was related to the pressure drop, which was in turn affected by die design developed at the die entrance. Sombatsompop and Intawong[7] proposed a novel design of a constant shear rate rheometer, which features the possibilities of moving either the piston or the barrel, and studied the flow properties and flow patterns of a LDPE melt with these two modes of rheometer operation. They found that the mode of rheometer operation had an effect on the flow properties and flow patterns being monitored.

In polymer extrusion, unstable flow of polymer melts through capillaries can cause various phenomena in the extrudate, such as extrudate swelling and fracture.[8] Die swell or extrudate swell is an important parameter determining the size and the quality of the extruded polymer products, and the extrudate swell is also used for assessing the polymer elasticity during extrusion. Extrudate swell has been widely studied, primarily with capillary rheometers. The mechanism and degree of swelling of the extrudate are usually explained in terms of elastic recovery or effect of residence time on the applied stresses, the extrudate swell of a polymer melt being varied by shear rate, temperature, fillers, and die land length.[5,6,8]

In this article, attempts were made to visualize the flow of a natural rubber compound in the barrel of a capillary rheometer with respect to the effect of the cross-sectional geometry of the dies used. Extrudate swell at the die exit was studied in accordance with the experimental conditions under which the flows were visualized. This is the first time that the relationship between both the flow patterns and extrudate swell in a capillary rheometer and the die design was established, with the results being then explained using independent experimental results. The findings in this paper offer some new ways of explaining such a relationship. Unlike other work, the rheometer used in this work had two dies, one located at the bottom of the barrel and the other inserted a certain distance above the first die for the flow studies.

EXPERIMENTAL

Materials and Compounding

The flow patterns and extrudate swell in the rheometer were investigated using natural rubber (SMR-CV60) supplied by the Malaysian Rubber Producers' Research Association (MRPRA). The formulation of the rubber compound, in parts-by-weight, was natural rubber 100, zinc oxide (ZnO) 3, stearic acid 2.5, CBS (accelerator) 1, sulfur 1, and pigment masterbatch (TiO_2) 1. The materials were compounded in accordance with the experimental procedure of Sombatsompop et al.[1] The compound was divided into two separate parts, one of which was pigmented with TiO_2 to give a white compound. Previous work[1,4] indicated that the pigmentation by TiO_2 did not affect the rheological properties of the rubber compound. After compounding, the rubber compound was pressed between layers of polyester film for 60 min at room temperature using a hydraulic press to produce a 5-mm thick sheet. A 25-mm diameter cork punch was used to cut the resulting rubber sheet into discs while the discs were kept between the polyester film to prevent elastic contraction of the rubber prior to further use.

Experimental Apparatus

The experimental arrangement of a constant-shear-rate rheometer (including barrel/die design

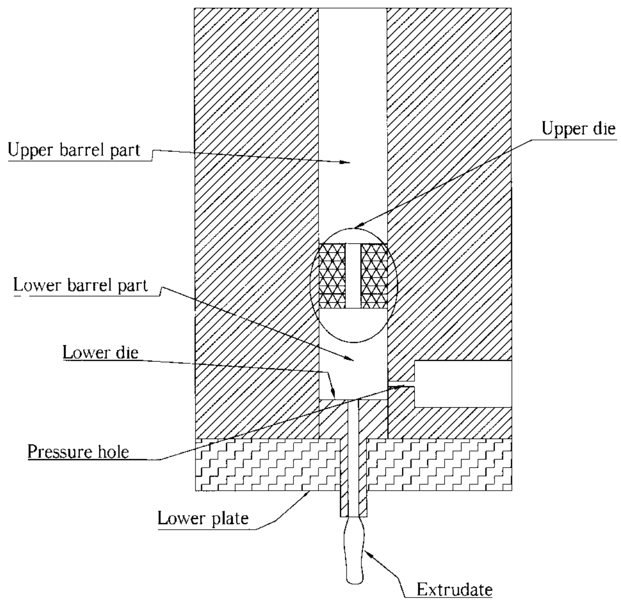


Figure 1 The barrel/die system in the rheometer apparatus (dimensions units in mm).

and dimensions) is shown in Figure 1; all the components were fitted into an AGS-500D (Shimadzu) tensile testing machine. The barrel was specially designed so that two dies could be firmly located along the barrel length at the same time. The rheometer used two die locations (see Figure 1): one at the bottom of the barrel (hereafter called *lower die location*) and the other 35 mm above the surface of the first die (hereafter called *upper die location*). In this work, fives dies with different cross-sectional geometries were used, the die geometries and dimensions are shown in Table I. It should be noted that this experimental arrangement was intentionally designed so that it is similar to the design of conventional extrusion processes, whose designs involve converging and diverging die faces.[9] In this work, the experimental apparatus was set up in two different systems with respect to the die design, as follows:

- Experimental system I: varying the geometry of the upper die: In this system, die number 1 was firmly fixed at the lower die location, and the upper die location used die numbers 2 to 5.
- Experimental system II: varying the geometry of the lower die: In this system, die number 1 was firmly fixed at the upper die location, and the lower die location used die numbers 2 to 5.

In this section, the same piston speed (10 min/mm) was used to solely examine the effect of die geometry on the flow patterns in the rheometer. A small pressure hole was located between the two die locations to detect the occurrence of die entrance pressure drop, which was measured with a Pin-Spring pressure sensor.[10] The apparatus temperature was controlled with a Eurotherm 018 temperature controller.

Experimental Procedure

A colored layer technique was employed for flow pattern investigations. The experimental procedure was initiated by loading alternate unpigmented (brown color of rubber vulcanizate) and pigmented (white color compound) discs of rubber compound (starting with the white one) into the upper barrel, with the lower barrel and the upper die being filled with unpigmented rubber before starting the extrusion. The rubber was partially extruded at a temperature below that at which vulcanization would occur (i.e., 80°C). The residual material in the barrel and die was then vulcanized for 30 min, the temperature of the apparatus being then raised to 160°C. The rod of vulcanized compound was removed from the barrel, cooled, sectioned, and polished, and the flow patterns were investigated. The flows were visualized as a function piston displacement for experimental systems I and II.

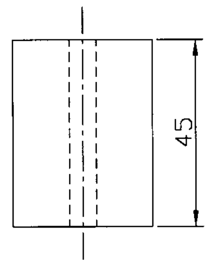
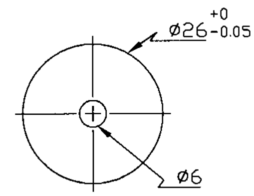
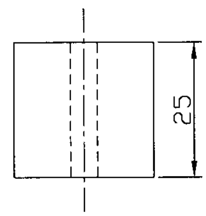
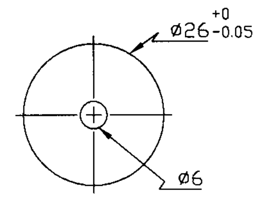
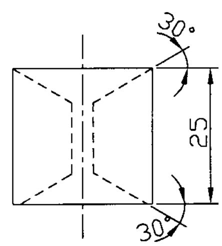
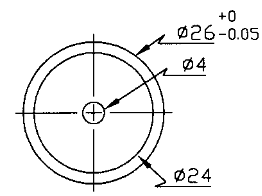
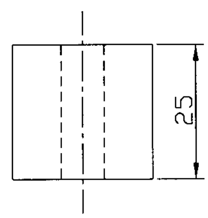
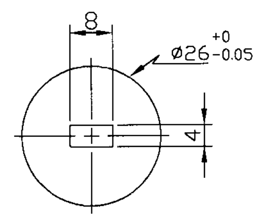
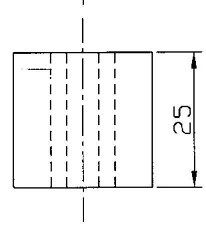
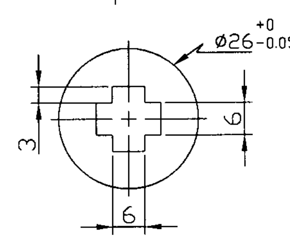
For extrudate swell studies, only experimental system I was used. The measurement of percentage increase in the extrudate diameter at the die exit was compared with the die diameter (6 mm) used. By trial and error, the piston speed used was adjusted for each die such that the pressure drop between the lower and upper die locations was the same, thereby eliminating the pressure effect on the measurement.

RESULTS AND DISCUSSION

Flow Pattern Investigations

Figures 2a–2d show the flow patterns of the rubber in the barrel with different piston displacements for various dies (experimental system I). The flows were found to be very complex, particularly at the lower barrel, and were a function of piston displacement. The flow patterns in the upper barrel were very similar for all the dies, with the flow patterns being parabolic in shape and the

Table I Die Design and Dimensions

Die Number	Type of Cross-section	Side View of Die	Top View of Die
1	Circular		
2	Circular		
3	Tapered circular		
4	Slit		
5	Crossed		

flow moving radially inward towards the upper die. This result indicates that the flow patterns in the upper barrel were independent of the die design, which is in good agreement with previous work.[1] Together these results clearly indicate that the flow that developed in the barrel was a function of piston displacement and its complexity was influenced mainly by the action of the piston/barrel system. Although the flows in the barrel were similar for all dies, by consideration of the central flow layer for a given piston displacement it can be seen that the sample from die number 2 (Figure 2a) exhibited the fastest flow, whereas that from die number 5 (Figure 2d) showed the slowest. This result involved the pressure drop occurring at the die entrance. Die number 2 was likely to produce relatively higher pressure drop at the entrance region than die number 5 because of the size and die entry geometry. This behavior was supported by the results of previous

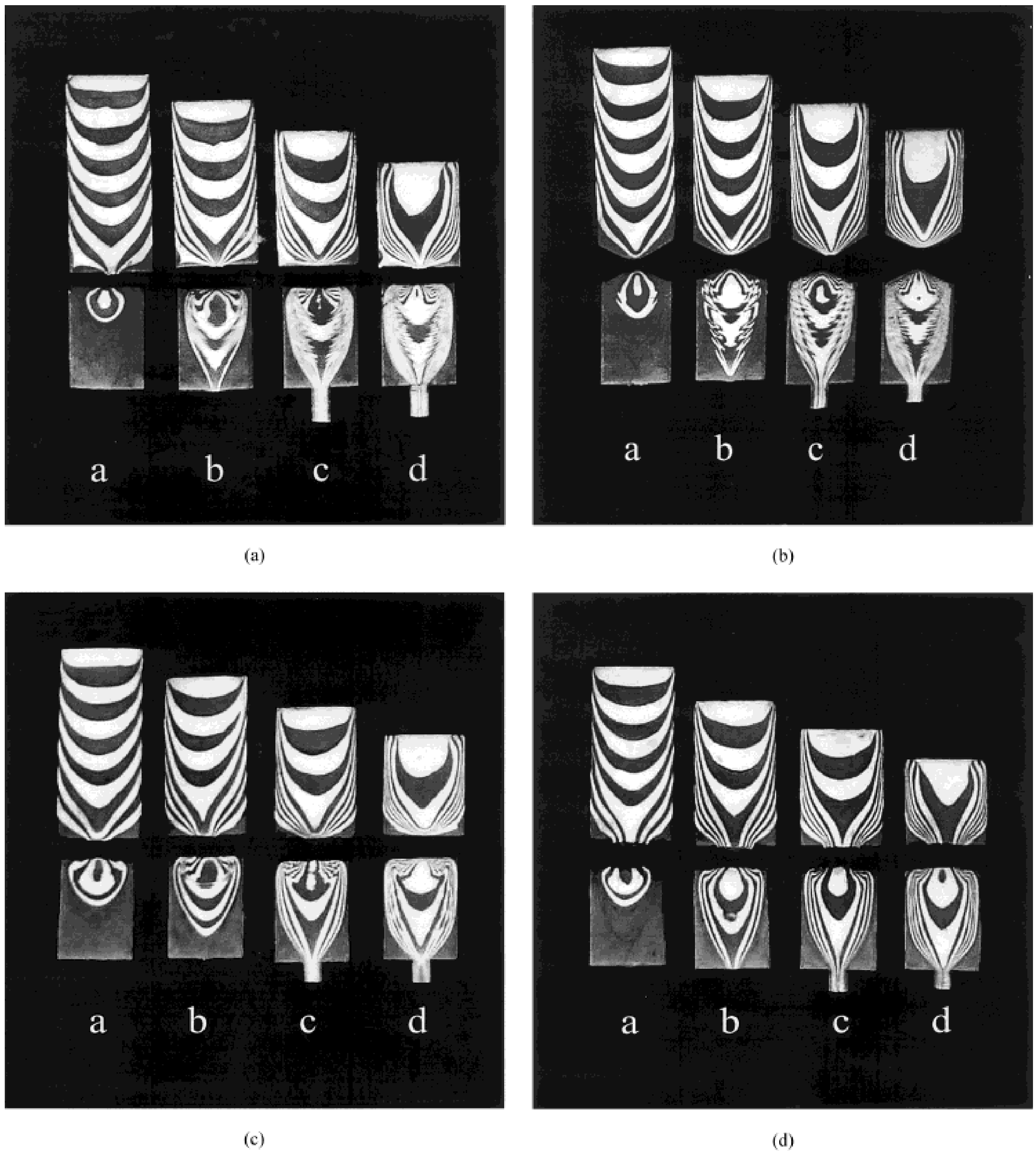


Figure 2 Flow patterns of natural rubber in the upper and lower barrels for experimental system I with different piston displacements (a: 10 mm; b: 20 mm; c: 30 mm; d: 40 mm): (2a): die number 2; (2b): die number 3; (2c): die number 4; (2d): die number 5.

work[4] dealing with the flow patterns developed using circular dies with different entry angles.

In the lower barrel, the flow patterns were different with all the dies. The flows with die number 2 were the most complex, whereas those with die number 5 were the least. The flows increased in complexity as the piston displacement increased, and the regular structure of the striations disappeared. The complex flows were related to the velocity of the flows developed at the upper barrel and die sections as already described. It was thought that the relatively fast flow of the material with die number 2 was rapidly subjected to a deceleration as it entered the

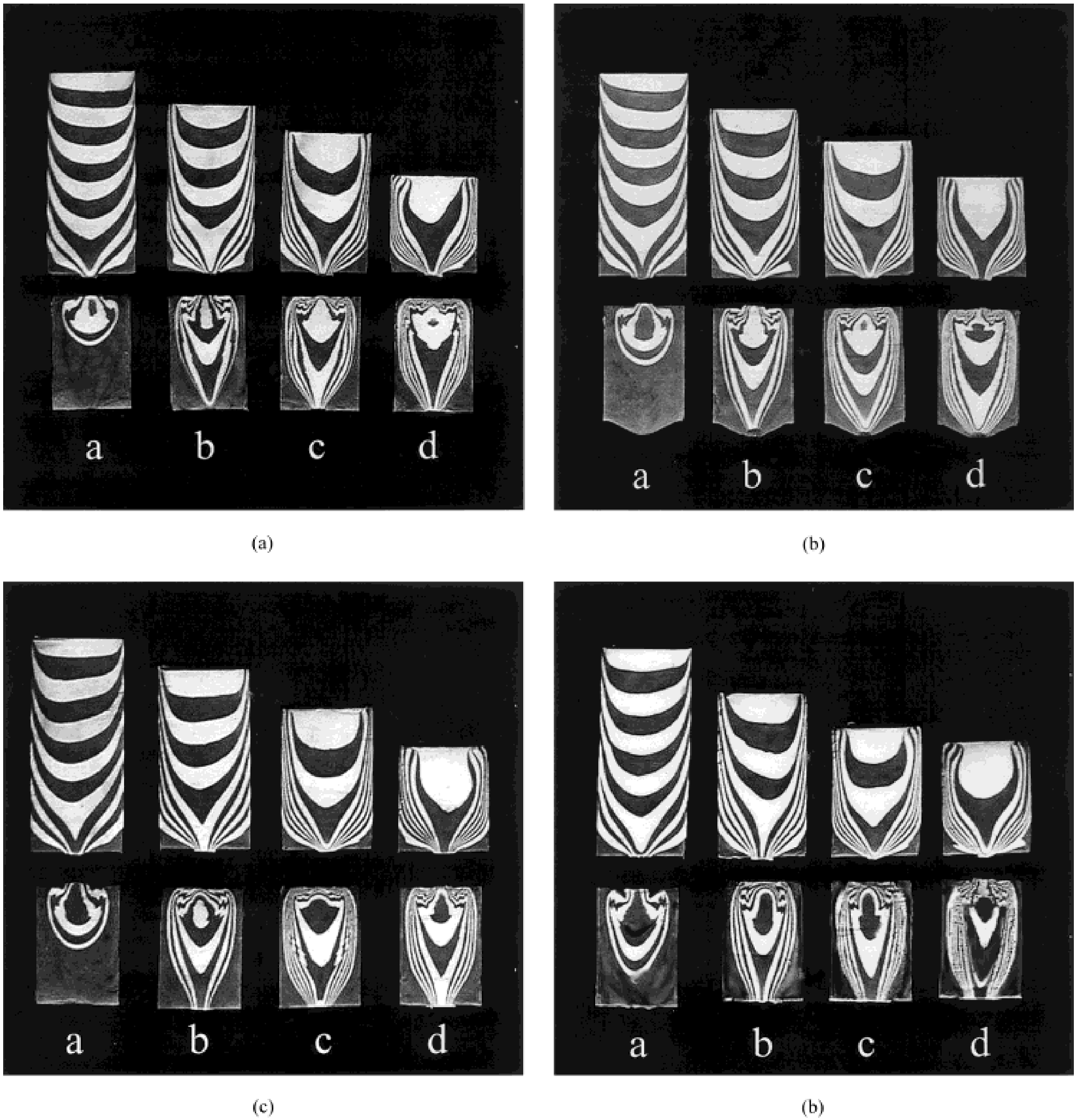


Figure 3 Flow patterns of natural rubber in the upper and lower barrels for experimental system II with different piston displacements (a: 10 mm; b: 20 mm; c: 30 mm; d: 40 mm). (3a): die number 2; (3b): die number 3; (3c): die number 4; (3d): die number 5.

lower barrel. The melt deceleration was due to the difference in the melt velocity within the upper die and the lower barrel. The material was then forced to diverge from the center and flowed towards the barrel wall before moving downward towards the capillary (lower) die. This effect was thought to decrease from die numbers 2 to 5 (Figures 2a–2d). Hence, the complexity of the flows inside the lower barrel were dependent not only on the piston displacement, but also on the geometry of the upper die used.

The flow patterns of the rubber compound with various dies in experimental system II are shown in Figures 3a–3d. The flow patterns in the upper barrel were still the same for all cases, confirming that the flows in this region are solely dependent on the piston/barrel action, as observed in experimental system I (Figures 2a–2d). We also stated,

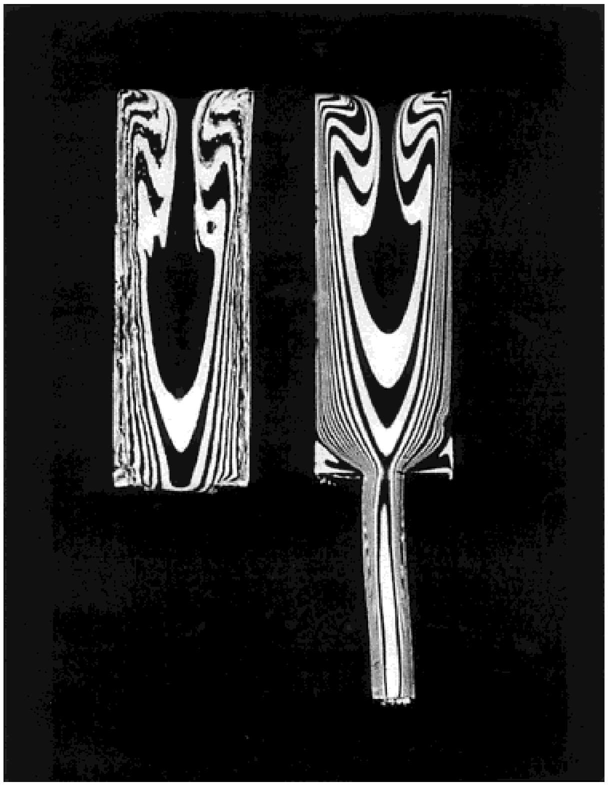


Figure 4 Flow patterns of natural rubber without (left) and with (right) die.

when considering the flows in Figures 2a–2d, that the flows inside the lower barrel were affected only by the upper die. If this was true, one would expect to obtain similar flow patterns in the lower barrel because the same die (die number 1) was utilized for all cases. The results in Figures 3a–3d show that the flows in the lower barrel are independent of the lower die geometry, thus confirming this expectation. An independent experimental result (Figure 4), showing the flow patterns in the barrel *with* and *without* die at the bottom of the barrel, also substantiate this expectation. Overall, these results indicate that the general style of flows patterns was independent of the die used.

Extrudate Swell Investigations

The results of percentage increase in extrudate swell for the rubber using experimental system I are shown in Figure 5. In those studies, the extrudate swell for each upper die was measured periodically as the piston moved down the barrel. For all cases, it can be seen that the diameter of the rubber extrudate was greater than that of the die and the increase in the extrudate diameter was ~30–45%. The percentage increase in extrudate swell with die number 3 was less than that with die number 5. The difference in the extrudate swell between these two dies was not caused by the pressure drop because the piston speed was altered to equalize the pressure drop for all dies. The difference in the extrudate swell can be explained using the flow patterns generated with these two dies. The material in die number 3 seemed to have greater residence time in the barrel because of the relatively higher degree of flow complexity, which occurred as a result of the radial flows of the material moving towards the barrel wall and inward the capillary die. The longer the residence time, the less the elastic characteristic, and thus the reduced swelling.

Another interesting aspect to consider was the change in extrudate swell during the movement of the piston down the barrel. Generally, one would expect to observe the same value of extrudate swell as the piston proceeds down the barrel, with the assumption that the piston speed remained constant with no change in frictional effect between the piston and the barrel. However, this was not the case. As can be seen in Figure 5, the swelling magnitude decreased with piston displacement. One possible factor that may cause the decrease in the magnitude of extrudate swell was the change in entrance pressure drop as the piston proceeded down the barrel. With this possibility in mind, we periodically measured the entrance pressure drop (using die number 1) as

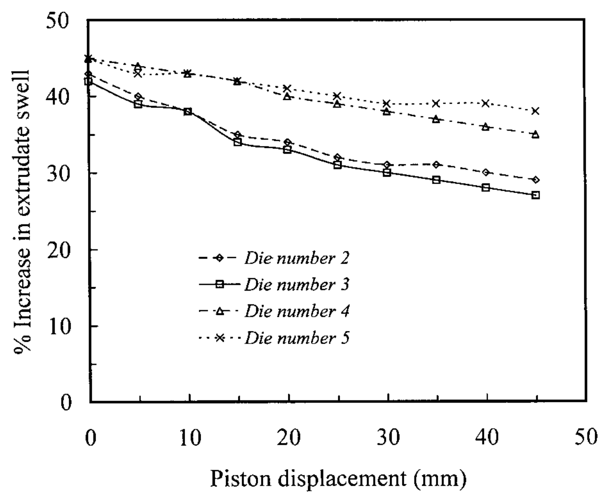


Figure 5 Percentage increase in extrudate swell for different piston displacements (using experimental system I).

the piston moved down the barrel. The obtained results (not shown here in this article) indicated that the entrance pressure drop did not change at all with piston displacement. Therefore, the change in the extrudate swell was not caused by the entrance pressure drop.

In our opinion, if both the flow patterns and extrudate swell changed with piston displacement, these two phenomena should have a relation to one another; that is, the decrease in percentage increase in extrudate swell is associated with the flows and other flow-related parameters. The extrudate swell should also be related to the flow developed only *in the lower barrel* because the general style of the flows in the upper barrel was the same in all cases. If this was true, the extrudate swell value obtained from different upper dies would be different, the extrudate swell value being different with die geometries (see Figure 5). The extrudate swells produced by die numbers 2 and 3 were very similar because of their similarities in flow patterns; this similarity was also the case for the extrudate swelling for die numbers 4 and 5. The following flow-related parameters are thought to be linked with the decrease in percentage increase of extrudate swell (as shown in Figure 5):

- Residence time: The residence time of the material in the lower barrel, as a result of the flow patterns already shown, increased as the piston proceeded down the barrel because the material did not move directly towards the lower die but tended to flow radically across the barrel diameter before moving towards the die. It is widely known[8] that a polymer melt with greater residence time will exhibit less elastic characteristic and thus reduced swelling.
- Diverging flow: As indicated by the flow pattern results in Figure 2, diverging flow was observed in the lower barrel. This pattern resulted in a recoil reaction of the polymer molecules from the upper die flow and tended to reduce the swelling. This result is linked with the work by Orbed and Delay[11] who suggested that a straight die generated greater swelling of the extrudate than a diverging die.
- Temperature rise during the flow: It has been widely accepted that melt temperature change results in a change in viscous and elastic characters of a polymer melt.[9] In relation to this work, as the piston proceeded, the viscous effect became greater because of an increase in melt temperature resulting from the shear heating during the flow. This result was confirmed by experimentally measuring the temperature (5 mm above the entrance using die number 1) of the polypropylene melt in the same rheometer (see Figure 6); the measuring conditions were the same as those used for the extrudate swell measurement and the details of temperature sensing system are given elsewhere.[12,13] It should be noted that, in this case, we used the polypropylene melt instead of the rubber because of some limitations in strength of the temperature sensor used. Previous work[14] has shown that the temperature change for the polypropylene melt was related to the flows occurring in the barrel of a capillary rheometer of a natural rubber compound. It can be seen that the overall melt temperature increased ~8–9°C with a piston displacement of 70 mm. Above this displacement, the temperature rise stabilized. The increase in melt temperature was due to the shear heating effect during the flow.[13] The increase in the temperature during the flow led to a reduction in the elastic characteristic and extrudate swelling.

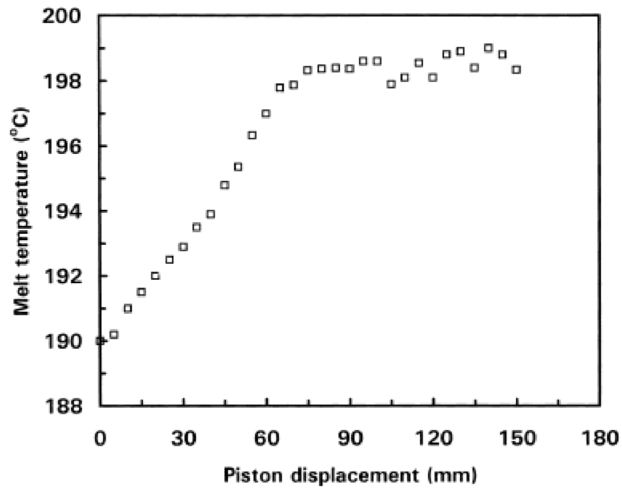


Figure 6 Temperature change of polypropylene melt in the rheometer for different piston displacements.

CONCLUSIONS

A colored tracer technique was employed to visualize the flow patterns of natural rubber compound in the barrel of a capillary rheometer with respect to the effect of die design. The following findings were noted:

- The flow in the upper barrel was dependent on the piston/barrel system and changed with piston displacement. The flow in the upper barrel was similar with all the dies, whereas the complexity of the flow in the lower barrel was dependent not only on the piston displacement but also on the geometry of the upper die design.
- The complex flows inside the lower barrel arose because of the divergence of the flow front inside the lower barrel, and the degrees of complexity or divergence of the flows were associated with melt velocity difference occurring inside the lower barrel.
- The flow patterns that developed in the whole barrel were independent of the die located at the bottom of the barrel.
- The change in extrudate swell was associated with the flows occurring inside the barrel in connection with residence time, elastic characteristic, and the temperature rise during the flow.

Finally, the findings in this paper clearly indicate that the general style of the flow patterns of natural rubber was greatly dependent on the die geometry that the rubber had previously flowed past.

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REFERENCES

1. Sombatsompop, N.; Tan, M. C.; Wood, A. K. Polym Eng Sci 1997, 37(2), 270.
2. Wood, A. K.; Read, A. G. G.; Lovegrove, J. G. A. Plast, Rubber Compos Process Appl 1989, 12(1), 15.
3. Liang, J. Z.; Huang, Y. Q.; Tang, G. J. Plast, Rubber Compos Process Appl 1992, 18(2), 311.
4. Sombatsompop, N.; Wood, A. K. Polym Eng Sci 1997, 37(2), 281.
5. Ma, C-Y.; White, J. L.; Weissert, F. C.; Isayev, A. I.; Nakajima, N.; Min, K. Rubber Chem Technol 1985, 58, 815.
6. Song, H. J.; White, J. L.; Min, K.; Nakajima, N.; Weissert, F. C. Adv Polym Technol 1988, 8(4), 431.
7. Sombatsompop, N.; Intawong, N-T. Mater Res Innovat 1999, 3(3), 150.
8. Bartos, O.; Holomek, J. Polym Eng Sci 1971, 11, 324.
9. Cogswell, F. N. Polymer Melt Rheology; George Godwin: London, 1981.
10. Sombatsompop, N.; Intawong, N-S.; Intawong, N-T. Polym Testing 2000, 19(5), 579.
11. Orbed, N.; Delay, J. M. Polym Eng Sci 1984, 24, 511.
12. Sombatsompop, N.; Panapoy, M. J Mater Sci 2000, 35, 6131.
13. Sombatsompop, N.; Chaiwatanapipat, W. Adv Polym Technol 2000, 19(2), 79.
14. Sombatsompop, N.; Wood A.; Godfrey I. SPE ANTEC Tech. Papers, 1997, 43(1), 388.