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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: 000–000, 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 i[n] in all cases except degraded rubb[er] evidence of vortices in the die cor[ner]. [6] conducted flow marker experim[ents] rubber compounds, including na[tural] (R), SBR, and EPDM, in the barre[l] y rheometer using a wide range of [shear rates] ey found that the radial flow simpl[y] rd to the capillary die as the ram [en]e barrel, with no evidence of second[ary] batsompop et al.[1,4] investigated th[e] s of NR compounds developed in[dustrially] and the capillary of a rheometer[.] ented rubber compound and found [that] patterns in the barrel of the capilla[ry] were complex and a function of pi[gm]ent, with the general style of the [flow] being independent of die design (s[ha]rp entry angle). The complex flows result[ed] rial adjacent to the piston that was [ad]e the polymer, with the surface flo[w] the face of the piston and down t[he] the barrel. Although the die desi[gn] fect the general style of the flow pa[tte]rn in the barrel, it influenced the flow patte[rn], that is, the flow patterns in the die with the occurrence of flow irregul[arities] the die and melt fracture at the die [entrance] henomenon was related to the pres[sure] hich was in turn affected by die des[ign] d at the die entrance. Sombatsomp[ong][7] proposed a novel design of a co[nstant] rate rheometer, which features the of moving either the piston or the studied the flow properties and flow[ing] LDPE melt with these two modes [of] operation. They found that the mo[de] ter operation had an effect on the flo[w] and flow patterns being monitored.

In polymer extr[usion] ble flow of polymer melts through cap[illaries] cause various phenomena in the ext[rudate] as extrudate swelling and fracture.[5-8] t extrudate swell is an important para[meter] mining the size and the quality of the [polymer] ymer products, and the extrudate swe[ll] d for assessing the polymer elasticity[during] trusion. Extrudate swell has been [studied] ied, primarily with capillary rheomet[ers] chanism and degree of swelling of the [polymer] e usually explained in terms of elasti[city] r effect of residence time on the appli[ed] the extrudate swell of a polymer me[lt] aried by shear rate, temperature, fille[r] 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 and dimensions) is shown in Figure 1; all the components were fitted into an AGS-500D (Shi-

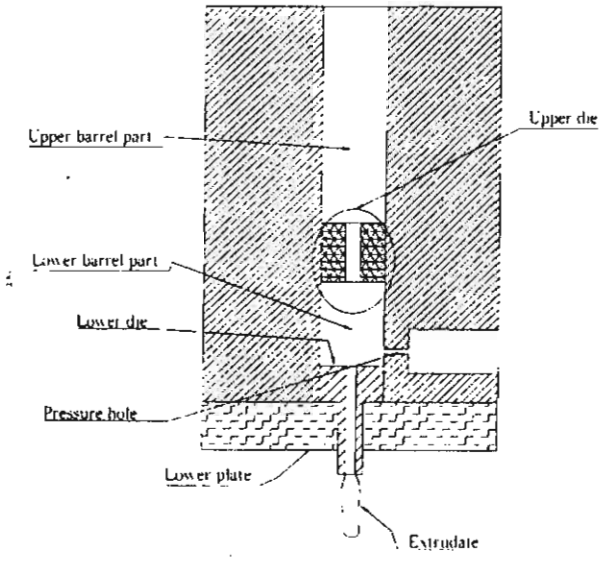


Figure 1 The barrel/die system in the rheometer apparatus (dimensions units in mm).

madzu) 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.

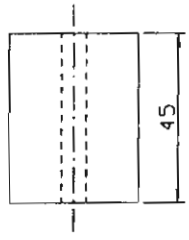
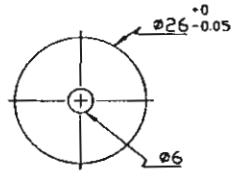
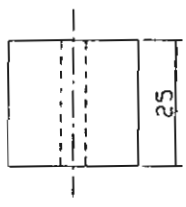
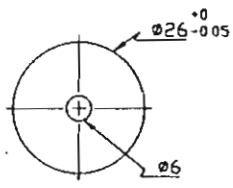
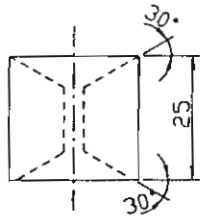
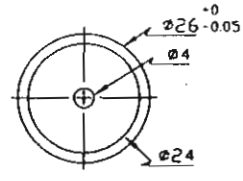
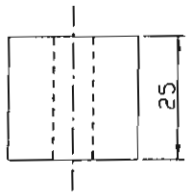
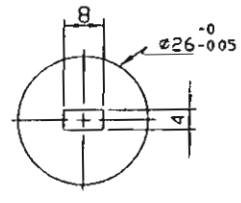
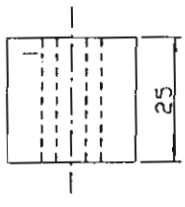
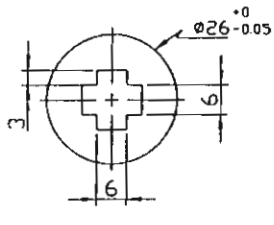
For extrudate swell studies, only experimental system I was used. The measurement of percentage increase in the extrudate diameter at the die exit was compared with the die diameter (6 mm) used. By trial and error, the piston speed used was adjusted for each die such that the pressure drop between the 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 flow moving radially inward towards the upper die. This result indicates that the flow patterns in

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		

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 work[1] dealing with the flow patterns developed using circular dies with different entry angles.

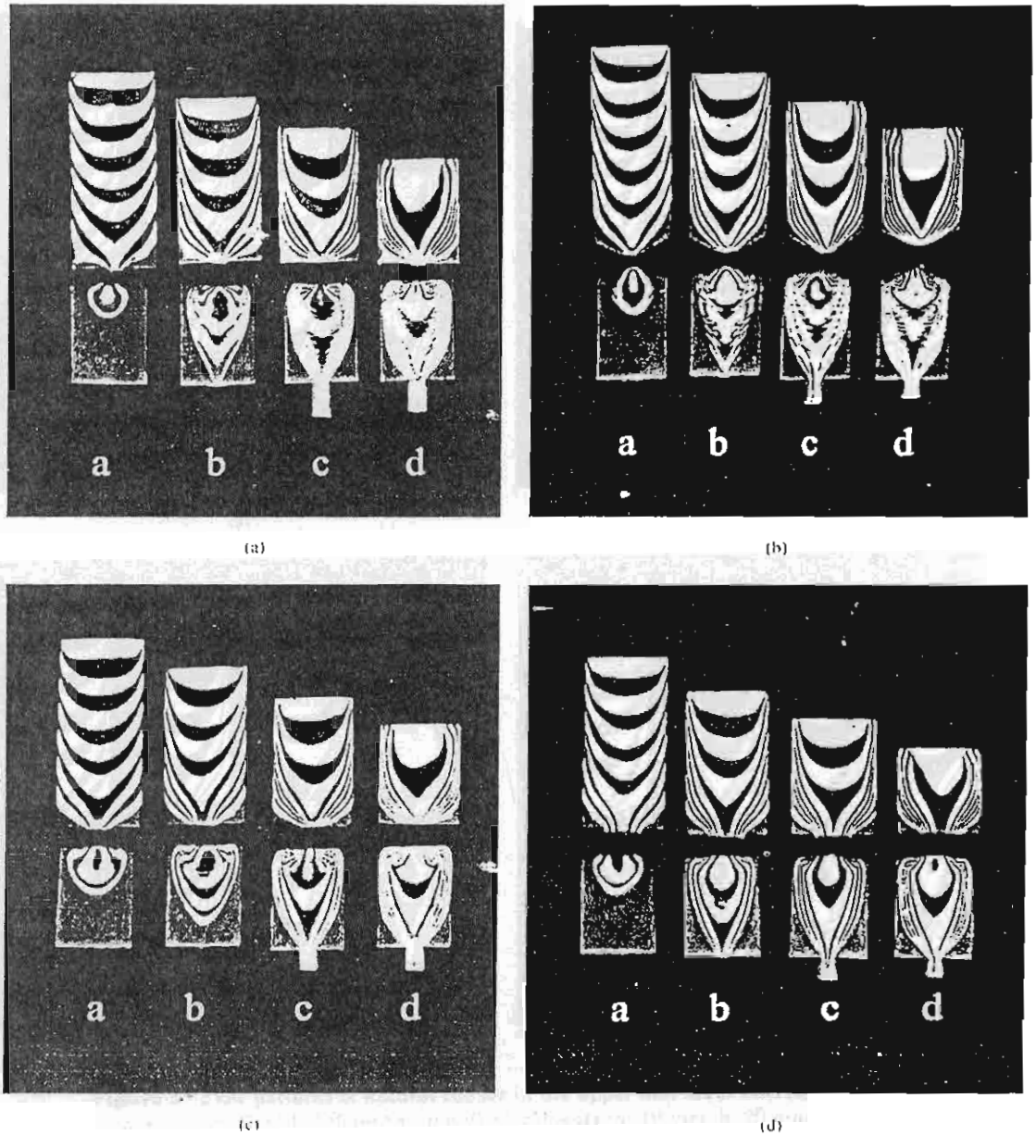


Figure 2 Flow patterns of natural rubber in the upper and lower barrels for experimental system I with different piston displacements (a: 10 mm; b: 20 mm; c: 30 mm; d: 40 mm); (2a): die number 2; (2b): die number 3; (2c): die number 4; (2d): die number 5.

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 lower barrel. The melt deceleration was due to the difference in the melt velocity within the upper

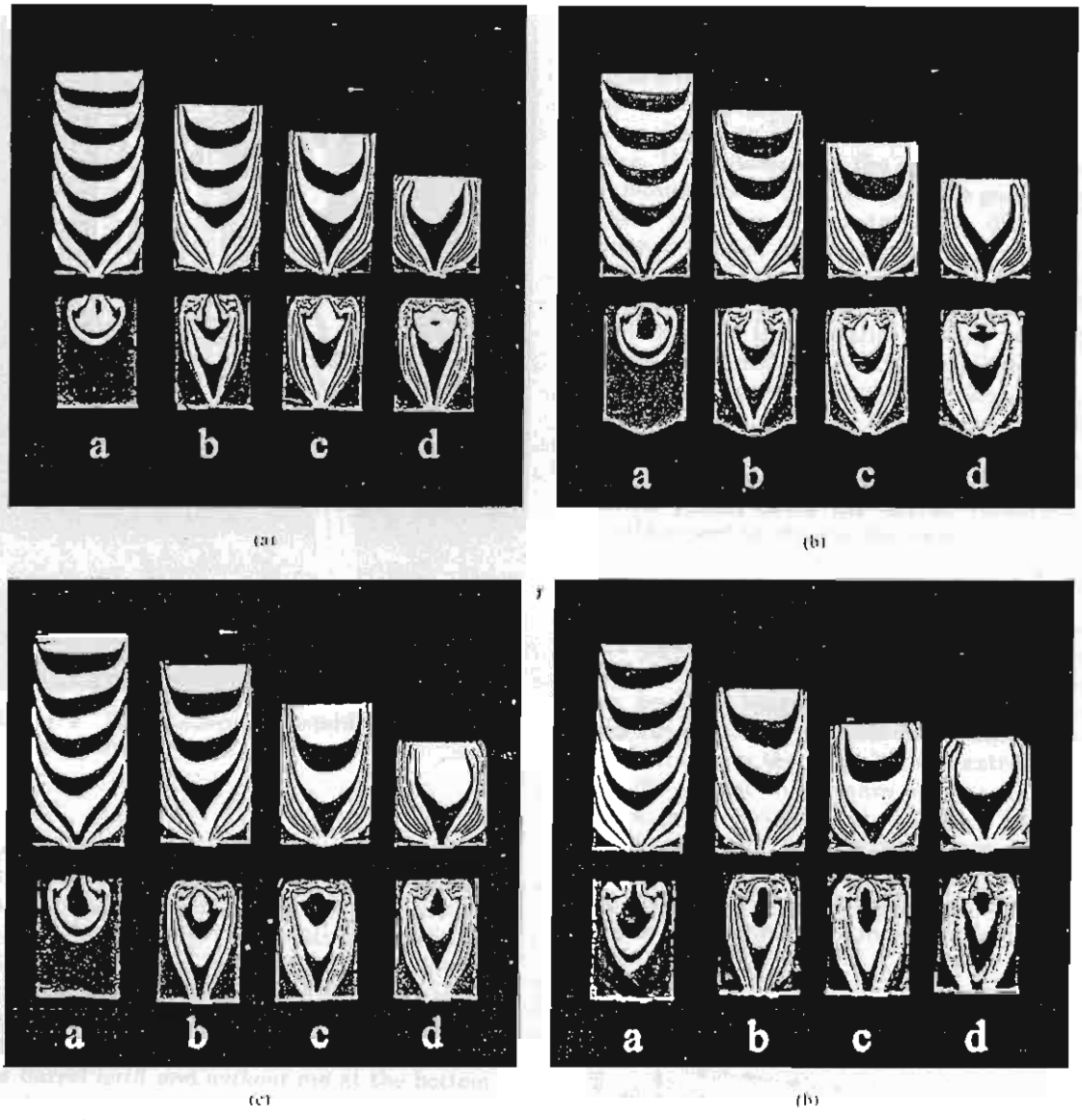


Figure 3. Flow patterns of natural rubber in the upper and lower barrels for experimental system II with different piston displacements (a: 10 mm; b: 20 mm; c: 30 mm; d: 40 mm). (3a): die number 2; (3b): die number 3; (3c): die number 4; (3d): die number 5.

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, when considering the flows in Figures 2a–2d, that the flows inside the lower barrel were affected

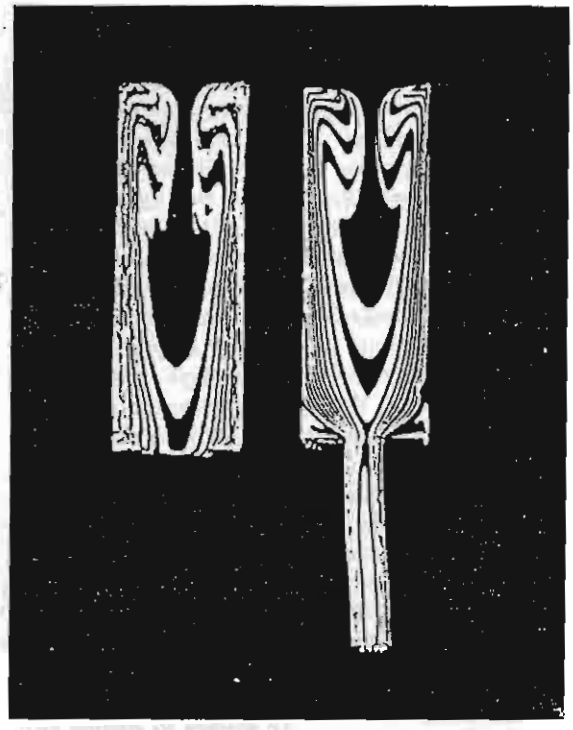


Figure 4 Flow patterns of natural rubber without (left) and with (right) die.

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 the piston moved down the barrel. The obtained results (not shown here in this article) indicated

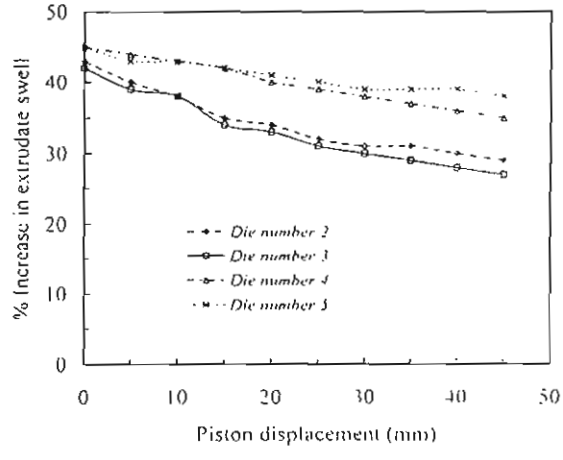


Figure 5 Percentage increase in extrudate swell for different piston displacements (using experimental system I).

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.

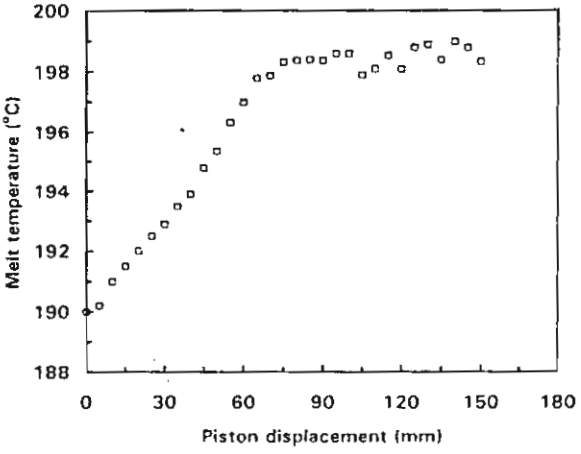


Figure 6 Temperature change of polypropylene melt in the rheometer for different piston displacements.

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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Polymer Journal, Vol. 33, No. 6, pp 491—494 (2001)

NOTES

A New Experimental Method for Determining Simultaneously True Radial Temperature Profiles of Polymer Melts under Isothermal Capillary Flow

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KEY WORDS Temperature Measurements / Shear Heating / Polymer Melts / Temperature Sensor /

In injection moulding and extrusion processes, the temperature of the polymer melt is dependent on the amount of heat generated in the polymer caused by viscous dissipation, the heat transferred through the barrel and screw, as well as the thermal properties of the polymer melt.[1] The transferred heat also effects the polymer melting behaviour, affecting the uniformity of the melt discharged from the process and its temperature. In extrusion, the melt temperature has a significant effect in the output rate of an extruder whereas melt temperature readings on the extruder are not absolute, but vary with the method of the measurement, the sensor type used, and specific conditions (such as bore diameter of hole and pressure levels).[2,3] It is very essential to measure the true temperature of the polymer melt during processing. Ideally, the properties of temperature sensor used for accurate temperature measurements in polymer processing are fast response time, non-intrusive measurement capability, real time display and robust design.[4] In the literature, various types and designs of temperature sensors have been introduced and used which can be categorized into the following types; Infrared (IR) probe, conventional thermocouple, optical pyrometers, ultrasonic probe, refractive index and temperature sensitive tracers.[5] It should be noted that where simultaneously radial temperature profiles in the melt are required the above temperature sensors are not very effective.[4]

As stated earlier that the temperature readings are dependent on the method of the measurement and the type of the sensor used. This is because different methods and sensor types present different deviations in melt temperatures being measured, this concerning shear heating and heat conduction effects between the melt and the sensor. The problem related with temperature measurement results from the complex dynamic and thermal state of the system to be measured. In most cases, the presence of a temperature sensor (probe) into the flowing polymer melt can cause two significant errors in the measurements.[6,7] Firstly, as a polymer melt is flowing past a stationary temperature sensor there will be frictional heat generated at the surface of the sensor, thus causing the temperature rise in the system. Secondly, there will also be a heat transfer (heat conduction) between the sensor root and its tip. As a result of these errors the measured temperature (T_m) needs to be corrected, this being carried out on the basis of eq 1.[8]

$$T_{true} = T_m + T_c - T_f \quad (1)$$

Where T_{true} is the true melt temperature, T_c is the heat conduction error, T_f is the shear heating error.

In this article, we aimed to introduce a new experimental apparatus (including design of the temperature sensor), and the experimental procedure in order to obtain the simultaneously true radial temperature profiles of PP melt under isothermal flow without the errors of T_f and T_c as stated above. Exemplary results with critical discussion were also given.

EXPERIMENTAL

Material and Experimental Equipment

The polymer used in this article was a polypropylene (PP, P-700J), supplied in granular form by Thai Polymer Propylene Co., Ltd., with a Melt Flow Rate of 16.

The experimental apparatus used for the study of the true temperatures of the flowing PP melt is shown in Figure 1, the apparatus design being similar to a conventional rheometer.[9] The barrel was specially designed such that it could be opened up into two halves which were bolted together, the barrel being fitted onto the base of a AGS-500 D (SHIMADZU) tensile testing machine. The fitting of the barrel onto the tensile testing machine allowed the possibility of the extrusion using the cross-head of the tensile machine. The barrel used was 315 mm long and 35 mm in diameter. A separate die plate and a die holder were bolted to the bottom of the barrel. A circular die with L/D of 55/4 was used. A small pressure hole was located near the die entrance for pressure measurement if desired. The apparatus temperature was controlled using a Eurotherm 018 temperature controller.

The temperature sensor used in this work was specially designed based on the construction of an interconnected series of Type-K thermocouples, the thermocouple wires (T_1 and T_2) being placed over each forming measuring junctions of the sensor (also in Figure 1). The

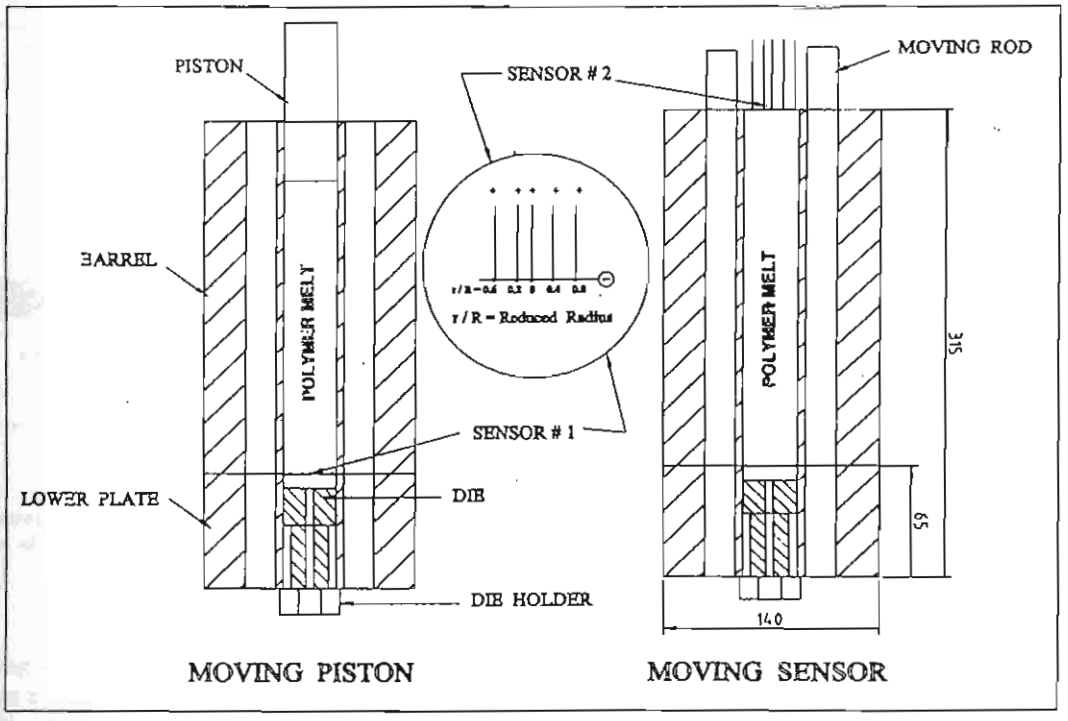

Figure 1. Experimental apparatus for temperature profile measurements (Left: moving piston mode; Right: moving sensor mode).

iameter of the wires was 0.6 mm. The design of the emperature sensor in this work was similar to that deeloped by Wood *et al.*[10] The measuring junctions were pot-welded so as to improve the mechanical strength of he measuring junctions of the sensor. It should be noted hat due to the design of the sensor whose measuring nctions were directly exposed to the melt, the tempera-re error from the conduction effect between the sensor nd the melt was automatically eliminated. Thus eq 1 as reduced to:

$$T_{true} = T_m - T_f \quad (2)$$

xperimental Procedure

In this work, the temperature measurements were arried out in two different ways as follows:

Moving piston (Figure 1-left): The melt was forced to flow past the stationary temperature sensor (sensor #1) and through the die, which were firmly located at the bottom of the barrel by action of the piston displacement, the piston being engaged to the cross-head of the tensile tester. The sensor was positioned at 10 mm above the die face. The melt temperatures were recorded as a function of time (or piston displacement).

Moving sensor (Figure 1-right): The temperature sensor (sensor #2) was attached to the moving rod of the apparatus which was firmly connected to the cross-head section of the tensile tester, this enabling the possibility of moving the sensor up and down along the stationary melt. In this case, the die exit was blocked. The sensor was carefully inserted so that the apparatus did not produce any leakage while moving the sensor past through the stationary melt. The melt temperatures were recorded as a function of time.

The moving piston mode gave the measured temperature (T_m) of the melt in the barrel while the moving sensor mode measured the temperature change due to the melt-sensor friction (T_f). By subtracting these two temperature results (at the same speed of the piston and sensor movements) the true melt temperature (T_{true}) were produced.

The experimental procedures were commenced by allowing the apparatus to warm up and the required temperature to stabilize. The die and die holder were inserted. The barrel was charged with the polymer using a charging tool to regularly tamp down the polymer. The piston was mounted, the motor starts and the piston drive was engaged in order to compress the polymer slightly. The polymer was left in the barrel for a period of thirty minutes in order to allow the polymer to melt and consolidate, as a consequence, could be considered to have a uniform temperature, this being referred to as the *isothermal* condition,[11] before taking the temperature measurements.

It should be noted that with the design of the sensor the temperature values are obtained *simultaneously* at various radial (reduced radius, r/R) positions across the duct, allowing radial temperature profile to be formed. All the measurements were taken using a high-speed data acquisition system coupled to a personal computer. The test temperature (referred to as initial bulk temperature of the melt) used was in the range of 190 to 230℃. The piston or sensor speed used was 50—500 mm min^{-1}.

RESULTS AND DISCUSSION

Figure 2 shows the selected temperature result for PP melt, at various radial points across the duct, as a func-

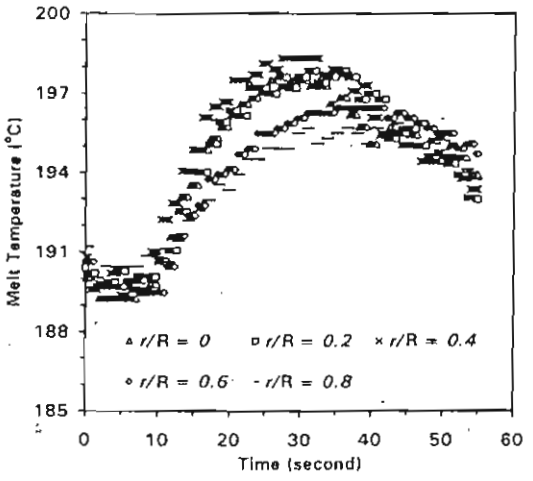

Figure 2. Temperature profiles and time for PP melt at a initial bulk temperature of 190℃ with piston speed of 100 mm min^{-1} (moving piston).

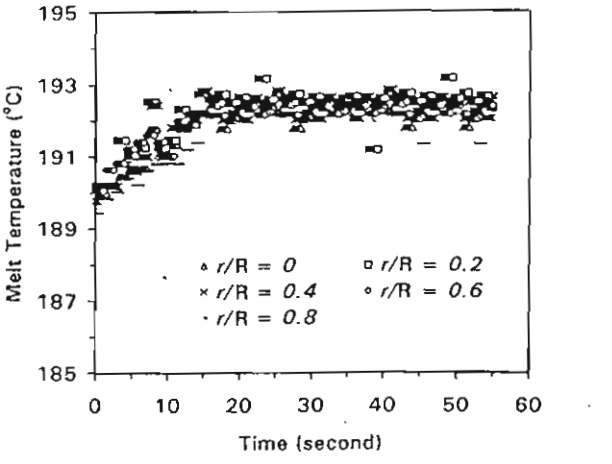

Figure 3. Temperature profiles and time for PP melt at a initial bulk temperature of 190℃ with sensor speed of 100 mm min^{-1} (moving sensor).

ion of time using sensors #1 (moving piston) at a piston peed of 100 mm min^{-1}. It was found that the melt temerature for any radial position initially increased and eached a maximum before it decreased as the extrusion ime proceeded. The increase in melt temperature durng the flow was as a result of shear heating effect. The xplanation for this behavior can be obtained in previous ork.[11] The maximum temperature rise (ΔT_{max}) was approximately 8—9℃.

The selected temperature profiles as a function of time r PP melt measured by moving the sensor #2 past the tationary melt at a sensor speed of 100 mm min^{-1} is hown in Figure 3. It can be seen that the melt temperare firstly increased about 3℃ up to 15 s and then stailized. The increase in melt temperature in the initial tage was solely due to the friction between the sensor nd the melt, the longer the sensor movement the higher e temperature rise. The plateau value of the temperare was caused by the fact that the maximum friction etween the melt and the sensor has reached.

One interesting point to be taken into consideration as the temperature differences for each junctions. In e case of moving piston, the melt temperature around e duct center ($r/R=0.0$—0.6) was greater than that ar the barrel wall, this being associated with shear ating and conduction effects and the velocity profiles ose details can be found elsewhere.[12] This was not en in the case of moving sensor, the melt temperature each r/R junction being very similar. The difference tween these two operation modes may probably be used by type of velocity profiles being developed and s shear heating during the measurements. In moving ton mode, the flow pattern of the melt was likely to be rabolic in form,[13] high melt velocity around the center the duct and low melt velocity near the barrel wall. is then resulted in relatively high shear heating (thus mperature increase) around the center. For moving sor mode, the velocity of all the measuring junctions s the same; therefore, the shear heating was indiffer-

The temperature data from Figure 2 were re-used to ain plots between the melt temperature as a function

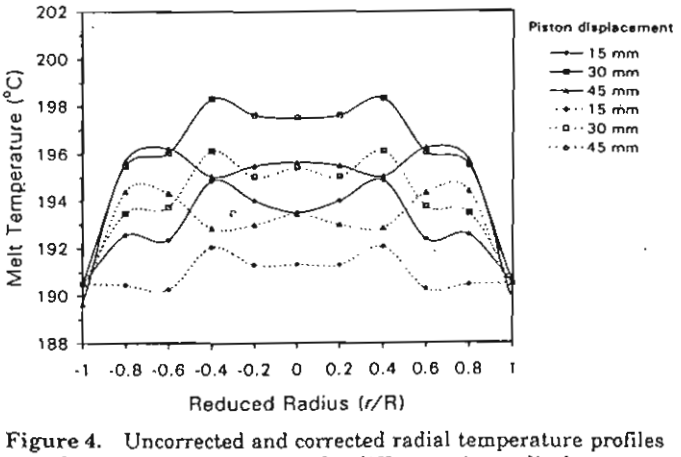

Figure 4. Uncorrected and corrected radial temperature profiles as a function of r/R positions for different piston displacements (Solid line: uncorrected value and Dashed line: corrected value).

of radial (r/R) position as shown in Figure 4, the data being presented for different piston displacements. The solid and dashed lines represent the uncorrected and corrected radial temperature profiles of the PP melt. In general, it was observed that the melt temperature profiles changed with piston displacement, the explanation for the changes in melt temperature profiles with piston displacement being given elsewhere.[12] It can be seen that the corrected (true) temperature profiles was different from that of the uncorrected ones, the corrected temperature being lower than the uncorrected as one would expect.

The work was extended to study the effects of either piston or sensor speed (50—500 mm min^{-1}) and initial bulk temperature (190—230℃) of the melt on the ΔT_{max} obtained from the two operation modes, the results being listed in Table I. For moving piston system, it was observed that the ΔT_{max} initially increased with piston speed and then decreased at the higher piston speeds. The increase in the ΔT_{max} was due to the shear heating effect whereas the decrease in ΔT_{max} was caused by the residence time effect, the residence decreasing with increasing piston speed. Increasing initial bulk temperature from 190 to 230℃ increased the ΔT_{max} from 7.1 to

Table I. Effects of piston and sensor speeds, and initial bulk temperature of the melt on ΔT_{max} for PP melt

Initial bulk temperature	Piston or sensor speed	ΔT_{max} for Moving piston	ΔT_{max} for moving sensor	Corrected ΔT_{max}
℃	mm min^{-1}	℃	℃	℃
190	50	7.1	3.9	3.2
	100	8.3	3.1	5.2
	200	8.3	2.8	5.5
	300	5.7	3.3	2.4
	500	5.4	3.2	2.1
210	50	8.5	3.8	4.7
	100	8.2	3.1	5.1
	200	9.7	3.0	6.7
	300	10.2	3.4	6.8
	500	7.5	2.8	4.7
230	50	10.9	3.8	7.1
	100	10.7	3.4	7.3
	200	11.1	3.1	8.0
	300	10.7	3.4	7.3
	500	10.6	3.0	7.6

11.1℃. In moving sensor system, it can be seen that the error from melt-sensor friction was so large, being in the order of 2.8—3.9℃ which was approximated to be 30% of the measured temperature. It was interesting to observe that the changes in ΔT_{max} was independent of sensor speed and initial bulk melt temperature. This result suggested that although the velocity of the flowing melt across the duct was different, one single correction (by moving sensor) was only required. For instance, the temperature profile at 100 mm min^{-1} sensor speed could be used for correcting the temperature profile generated at 100 mm min^{-1} piston speed (although the melt of the latter profile had different melt velocities across the duct., *i.e.*, the melt velocity profile is usually assumed to be parabolic in nature). After correcting the ΔT_{max} (Table I) the true ΔT_{max} values were in the range of 2—8℃ depending on the test speed used.

The findings in this paper had significantly practical implications in that, in order to obtain more accurate temperature value of the flowing polymer melt by insertion of the temperature probe into the melt stream, the melt-sensor friction correction had to be taken into consideration quantitatively.

CONCLUSION

This paper offered a new method of obtaining the simultaneously true radial temperature profiles of PP melt during an isothermal flow. It was found that the use of unsheathed thermocouple network and the moving of temperature sensor past the stationary melt could eliminate the conduction and shear heating errors between the temperature sensor and the polymer melt respectively. The error of the melt-sensor friction was approximated to be 30% of the measured melt temperature. Under the test conditions in this work, the true maximum melt temperature was found to be in the range of 2—8℃. Increasing the piston speed with higher initial bulk temperature of the PP melt tended to increase the true ΔT_{max} to the optimum value, too high piston speed resulting in a decrease in the true ΔT_{max}. The polymer melt-sensor friction was found to be independent of the sensor speed and initial bulk temperature of the PP melt.

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Effect of Die Design on Flow Visualization & Die Swell of NR in a Capillary Rheometer

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In polymer processing, the properties of the end-product are very dependent on the materials used and the design of the processing equipment. Unstable flows occurring during processing can result in low-quality products. It has been evident that the complex flows occurring are due to the design of the equipment and the material characteristics, and the pressure & temperature that occur[1]. The most common technique used for the determination of the flow properties of polymer melts is that of the capillary rheometer. It has been observed that the flow property results produced in the capillary rheometer depend on the design of the apparatus, the same die giving different results when used in different apparatus designs[2]. The differences in the results are associated with the flow patterns occurring[2]. As a result, studies on flow patterns of polymer melts in the capillary rheometer have been widely carried out[1-4]. Song *et al*[5] conducted flow marker experiments of various rubber compounds, including NR, SBR and EPDM in the barrel of a capillary rheometer using a wide range of die designs. They found that the flow radially simply moved inward to the capillary die as the ram moved down the barrel, no secondary flows occurring. Sombatsompop *et al*[1,4] investigated the flow patterns of NR compounds developed inside the barrel and the capillary of a rheometer by the use of pigmented rubber compound, and found that the flow patterns in the barrel of the capillary rheometer

Submitted 21/11/43

were complex and were a function of piston displacement, 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, used to extrude the polymer, surface flowing across the face of the piston and down the centre of the barrel. Although the die design did not affect the general style of the flow patterns in the barrel, but it influenced the flow patterns in the die, the flow patterns in the die being linked with the occurrence of flow irregularities along the die and melt fracture at the die exit[4]. This was related to the pressure drop, which was in turn affected by die design, developed at the die entrance. Sombatsompop and Intawong[6] have proposed a novel design of a constant shear rate rheometer that featured the possibilities of moving either the piston or the barrel, and the flow properties and flow patterns of a LDPE melt from these two modes of rheometer operation were studied. It was found that the mode of rheometer operation had an effect on the flows being observed.

Attempts to carry on the investigations into the flow patterns and die swell of NR compound in the barrel of a capillary rheometer, with respect to the effect of die geometry system, were made in this paper. This is the first time that the relationship between the flow patterns and die swell is established in a capillary rheometer as to the effect of die design. The arrangement of the rheometer apparatus (including barrel/die design and their dimensions) is shown in Fig. 1, all the components being fitted into an AGS-500D (SHIMADZU) tensile testing machine. It should be noted that the experimental arrangement in this work was designed so that it was similar to that used in conventional extrusion processes. The rheometer used two different dies for each time, one being located at the bottom of the barrel (referred to as Die#1) and the other (referred to as Die#2) being 35mm above the surface of Die#1. Die#1 was a

flat-entry die with 6mm in diameter and 43mm long. Only the design of Die#2 was varied, four different types of cross-section being used (see Fig. 1, Die #2a to #2d). A small pressure hole was located between the two dies (referred to as *lower barrel part*) to measure the pressure drop occurring between two dies, the pressure drop being taken using the Pin-Spring pressure sensor[7]. By trial and error, the piston speed was adjusted for each die such that the pressure drop between the two dies was the same during the flows. The apparatus temperature was controlled using a Eurotherm 018 temperature controller. All aspects of the sample preparation, including the formulation of NR compound, were used as used by Sombatsompop *et al*[1]. A coloured layer technique was employed to study the flow patterns in the barrel, the experimental procedure being detailed elsewhere[1]. In addition to the flow pattern study, die swell was investigated based on the measurement of percentage increase in the extrudate diameter at the die exit and compared with the die diameter (6mm).

Figure 2 shows the flow patterns of the rubber in the barrel of the rheometer with different piston displacements for various designs of Die#2. The flows were found to be very complex, specially at the lower barrel, and were a function of piston displacement. Although, the flow patterns in the upper barrel was very similar with all the dies, the flow patterns being parabolic in shape and the flow moved radially inward towards the die (Die#2), it can be seen that, by consideration of the central flow layer, the sample from Die#2d exhibited the fastest flow whereas that from Die#2a showed the slowest. This type of behaviour was also found in previous work [4] when using circular dies with different entry angles. In the lower barrel, the flow patterns were observed to be very different with all the dies. The flows with Die#2a was the most complex whereas those with Die#2d was the least. The flows increased

in complexity as the piston displacement increased, the regular structure of the striations disappearing. The complex flows arose because, as the material entering the lower barrel was forced from the central flow and became diverged from the centre towards the barrel wall before flowing downward towards the capillary die. The complexity of the flows varied with die geometry (Die#2). This was thought to be associated with memory effect since it had flowed through different geometries of the die(Die#2)[8]. Hence, the complexity of the flows inside the lower barrel was dependent not only on the piston displacement, but also on the die geometry.

The results of die swell for the rubber using various designs of Die#2 are shown in Fig. 3, the die swell for each die being measured periodically as the piston moved down the barrel. Normally, one would expect to obtain the same value of die swell as the piston proceeds, if the piston speed remains constant. However, this was not the case. As can be seen that the die swell decreased with piston displacement. The change in die swell was thought to be associated with the flows developed in the lower barrel as detailed earlier. If this was true, the die swell value obtained from different dies would be different, the die swell value varying with die geometries (see Fig. 3). The decrease in die swell may result from three possible reasons. Firstly, the diverging flows occurred in the lower barrel. This resulted in a recoil reaction of the polymer molecules from the die flow and this tended to reduce the swelling, this view being supported by Orbey and Dealy[9] who suggested, without giving any flow pattern results, that a straight die generated greater swelling of the extrudate than a diverging die. Secondly, the residence time, as a result of the flow patterns as shown earlier, of the material in the barrel increased as the piston proceeded, this being because the material flowed radially across the barrel diameter before moving towards

the die. It is widely known that a polymer melt having greater residence time will exhibit less elastic characteristic and thus reduced swelling[8]. Finally, as the piston proceeded the viscous effect became greater due to an increase in melt temperature resulting from the shear heating during the flow[10]. This can be substantiated by using the temperature profile result, (being measured at 5mm above the entrance of Die#1), of a polypropylene melt in the same rheometer (see Fig. 4), the measuring conditions being the same as used for the die swell measurement and the details of temperature sensing system being given elsewhere[11]. It can be seen that the overall melt temperature increased with piston displacement. The increase in the temperature during the flow led to reductions in the elastic characteristic and die swell. Comparing the die swell of Die#2b with Die#2d, it was found that the swelling of Die#2b was less than that of Die#2d, this again being explained using the flow patterns generated using these two dies. The material in Die#2b seemed to have greater time flowing in the barrel due to the radial flows occurring towards the barrel wall, and this would result in less elastic characteristic and reduced swelling.

In summary, a coloured tracer technique was employed to visualise the flow patterns of NR compound in the barrel of a capillary rheometer with respect to the effect of die design. The flows in the upper barrel were found to change with piston displacement and were similar with all the dies whereas the complexity of the flows in the lower barrel was dependent not only on the piston displacement, but also on the die design. The complex flows inside the lower barrel arose due to the divergence of the flow front on entering the lower barrel, and the degrees of complexity of the flows were associated with material memory effect. The die swell was found to relate with the flows developed inside the barrel, the flows being influenced by the die design.

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Figure 3	Percentage die swell for different piston displacements
Figure 4	Temperature profiles of PP melt in the rheometer at different piston displacements

Upper barrel part
Lower barrel part
Die #1
Pressure hole
Lower plate
Extrudate
Die #2
Die#2a
Die#2b
Die#2c
Die#2d

Fig. 1

Die #2a

Die #2b