http://dx.doi.org/10.1590/0104-1428.1696

Flow Visualization & Extrudate Swell Behavior of Natural Rubber Compound in Annular Die Capillary Rheometer

Naret Intawong, Auttapong Wiratket, Pholkrit Meechue
Department of Industrial Engineering, Faculty of Engineering, Rajamangala University of Technology Lanna, Thailand

Abstract: This research aims to study the flow pattern and extrudate swell properties of a Natural Rubber (NR) compound in a constant shear rate capillary rheometer, using two types of annular die: convergent annular and divergent annular. Results revealed that flow patterns that occurred in the barrels of both types of annular dies were significantly different, especially the vortex flow at the barrel wall and at the die entrance. This difference in flow pattern significantly affected both diameter swell and thickness swell of the NR compound. It was also found that thickness swell was higher than diameter swell in every test condition. This difference could be explained by analysis of the complex flow pattern at the die entrance.

Keywords: *Die design, extrudate swell ratio, polymer rheology, flow pattern, capillary rheometer.*

Introduction

In polymer's extrusion process, control of product size is more difficult than in other processes. That is, when melt polymer is extrudate from a die, it assumes a free flow pattern and changes its size and form from the die's form. This change results from the flow pattern and flow history of melt polymer flowing in the extrusion system. This change in polymer size after extrusion is known as the extrudate swell[1]. The impact of polymers extrudate swell has gained a lot of interest because it is a variable that can be controlled without causing damage to polymer product, both on products either their surface or their distortion. Generally, factors that affect of polymers extrudate swell mostly come from processing conditions. These factors are processing temperature, die pressure drop, and apparent shear rate during the production process[2].

In addition, current research has also found that the magnetic field has a direct impact on the swelling property of melt polymer. Sombatsompop et al. [3-5] studied the effect of the electrical magnetic field on the swell property of melt polymer by designing and constructing an electro-magnetic die with adjustable magnetic density. Their study found that the extrudate swell of polymer increased with the increase of magnetic flux density, particularly polymer with benzene as repeating unit in its molecular chain, such as polystyrene (PS). These findings were further developed by Intawong and co-worker[6] who compared the effects of the magnetic field on the swelling property of PS melt between steel die and stainless die. It was found that the overall extrudate swell ratios from the steel die were approximately 10% higher than those from stainless die. Due to application of the magnetic field, using steel die caused an increase in extrudate swell ratio, while using stainless die resulted in a decrease in extrudate swell ratio.

Understanding extrudate swell behavior of polymer enables to control of polymer's swell in real production industry, such as pipe extrusion process or extrusion blow moulding, which generally uses annular die. Control of the size of polymer products produced from annular die is quite complicated because the extrudate polymer expands in both external diameter and thickness, called diameter swell and thickness swell, respectively. Many existing studies attempt to explain various factors that affect swell behavior of melt polymer extrudate from annular die. For instance, results from the studies of swell behavior in annular die, both by mathematic model and computer simulation[7-10] and by experimental method[11-13] all indicate that the causes of diameter swell and thickness swell are shear stress, shear stress distributions, and extensional stress that stretch the melt polymer molecular chain while it is flowing in the die. More important is the deformation history experience of melt polymer due to annular die design that takes into account several variables of die contraction ratio, die gap opening, die land length, and converging or diverging type of annular die. Nevertheless, certain studies show that change in flow pattern of melt polymer during extrusion process significantly affects amount of swell. For example, Wood et al.[14] studied flow pattern of natural rubber compound in different types of capillary rheometer using a colored layer technique. He found that velocity profiles and flow pattern are important variables in determining flow properties of polymer. Sombatsompop et al.[15] studied flow pattern of natural rubber compound in two types of capillary rheometer: mobile piston and mobile barrel. They found the swell of natural rubber compound changed in accordance with the flow pattern and was influenced by convergent flow angle. Intawong, et al.[16] the studied flow pattern of natural rubber compound in circular rotating die capillary rheometer and found that die rotation caused complex flow pattern at the die entrance. The circumferential flow caused repetitious circulation at the die entrance. Hence, polymer had longer residence time. This resulted in

***Corresponding author: Naret Intawong**, Department of Industrial Engineering, Faculty of Engineering, Rajamangala University of Technology Lanna, 128 Huay Kaew Road, Chiang Mai, 50300, Thailand, e-mail: naret_i@yahoo.com*

decreasing the swell of rubber compound in accordance with the increase of rotation speed.

The aforementioned research indicates that the flow pattern of melt polymer directly affects on the swell behavior of polymer. Therefore, this research aims to study a relationship between flow pattern development and die design that affects swell behavior of NR compound during its flow in a capillary rheometer. This is the first time of an attempt to study flow pattern in the barrel of capillary rheometer using two types of annular die: convergent annular die and divergent annular die. Results of the study reveal that flow pattern in both types of die is significantly different. Moreover, the flow pattern significantly affects both diameter swell and thickness swell of natural rubber compound.

Experimental

Materials

All tests in this work used natural rubber (NR: STR 5L), supplied from PAN INNOVATION LIMITED (Thailand). The formulation of the NR compound was shown in Table 1. The materials were compounded in accordance with the experimental procedure by Intawong et al.[16]. The compound was divided into two separate parts, one of which was pigmented with titanium oxide (TiO_2) to create a white compound.

Split barrel capillary rheometer.

Figure 1 shows a constant shear rate rheometer which was designed with split barrels in order to take out the NR vulcanized rod. The barrels were 40 mm in diameter and 150 mm long. A small pressure hole (ϕ1.5 mm) was located at the barrel's base, just 5mm above the annular die face and 2 mm from the barrel wall, and connected to an adapter to detect when die entrance pressure drop occurred. The entrance pressure was measured with a pressure transducer (Dynisco, Model PT460E-2CB-6, Franklin, MA) which had a pressure ranges from 0-100 bars. The measurement accuracy was ±1% f.s.v, and the repeatability was ±0.2% f.s.v. Temperature control consisted of four heater bars, a type-K thermocouple, and a DD6 temperature controller (Changchai Meter Bangkok, Thailand). The accuracy of temperature measurement was 2.5%, and the measurement temperature ranges were from 0-300°C. These were used for monitoring and controlling the temperature of the apparatus components. The split barrel capillary rheometer was designed so that the die system could be easily changed.

Table 1. The formulation of the NR compound.

Chemicals	Compound 1 (g)	Compound 2 (g)
Zinc Oxide	12	12
Stearic acid	6	6
Accelerator (Mercaptobenzthiazole,)	2.1	2.1
Titanium Oxide (TiO_2)	6	-
Sulphur	9	9
NR	300	300

Annular die design

The annular die geometry shown in Figure 2 consisted of a convergent annular die and a divergent annular die. Both of the annular dies were made of SKD61 tooling steel that can be used at elevated temperatures. The die had two main parts: The die body and a mandrel which was designed to have 4 spider legs in radial line in order to be assembled with the die body using transition fit technique at the tolerance of H7/k6. This technique fixes the mandrel firmly inside the die body in alignment. In the case of the divergent annular die, there were 2 mandrels: mandrel #1 and mandrel #2. Both were assembled with a M6x1 screw. The mandrel had diameters (a) of 18 mm, 16 mm and 14 mm which caused die gaps of 1 mm, 2 mm and 3 mm, respectively. The die body had 40 mm external diameter and 20 mm internal diameter, with a die inclination angle at the fixed degree of +45° (divergent annular die) and -45° (convergent annular die). Both dies had the same die land length of 28 mm.

Colored layer technique

Figure 3a shows the colored layer technique used to study the flow pattern of the NR Compound. The experiment began by putting discs of NR compound with diameters of 40 mm and 6 mm thickness into the barrels, alternating between white pigmented NR compound (white color of NR+ Titanium Oxide compound) and unpigmented NR compound (brown color of NR compound). After that, the NR compound discs were extrudate with a mobile piston and passed through the annular die which was located at the bottom of the barrel. The rubber was partially extruded at a temperature below vulcanization point, which in this study was 110°C. In addition, after adding rubber into the barrel to its full extent, the experiment had to wait for 15 minutes for the rubber's temperature to become isothermal before extrusion[16]. The displacements of the piston was limited to stop moving at 30 mm, 60 mm, 90 mm and 120 mm from the barrel's top. After the piston stopped, the barrel's temperature was increased to 160°C to vulcanize the NR compound in the barrel for 15 minutes. It should be noted that while the piston moved to extrudate the NR compound inside the barrel, both types of NR compound

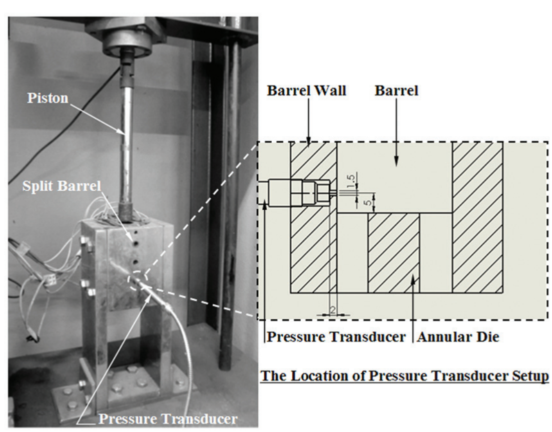

Figure 1. The split barrel constant shear rate capillary rheometer.

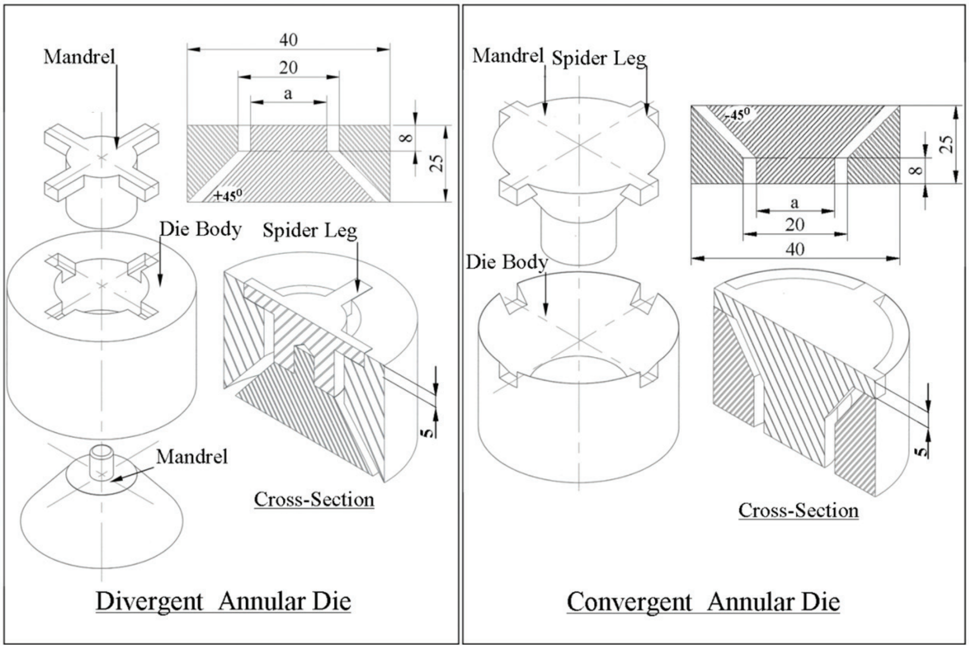

Figure 2. Die design and dimensions.

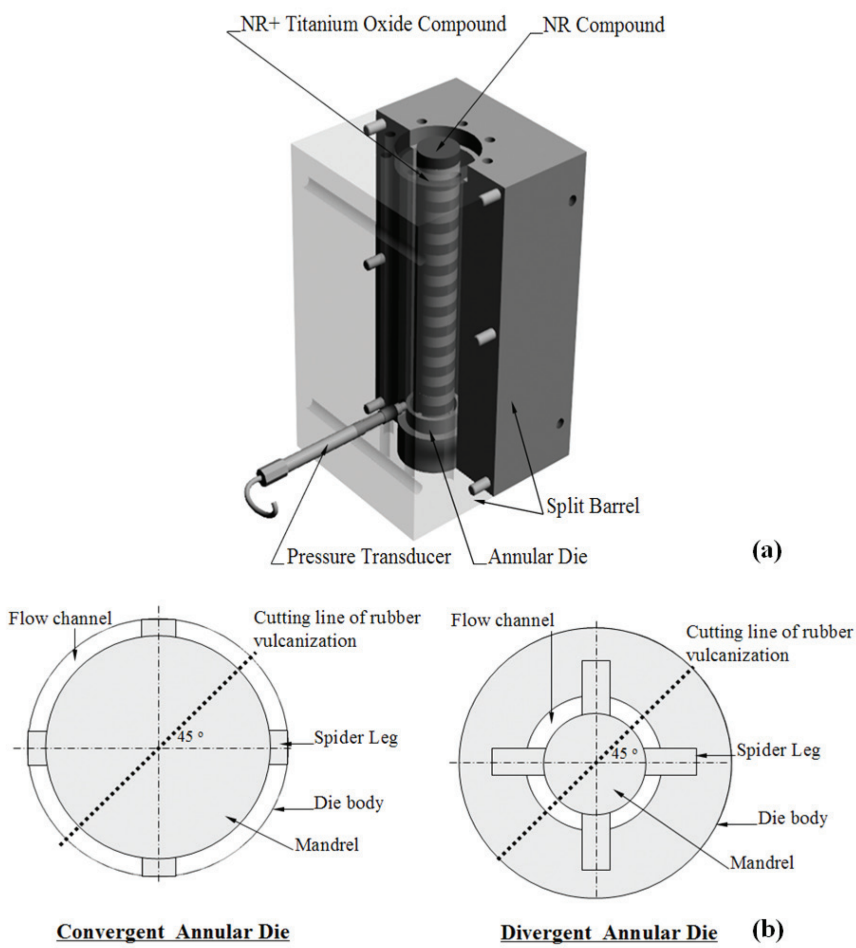

Figure 3. An experimental arrangement for the Colored Layer Technique (a) the tops of divergent annular die and convergent annular die show the cutting line of rubber vulcanization rod) (b).

changed their flow pattern simultaneously throughout the extrusion length. After extrusion stopped, the flow pattern of the rubber compound inside the barrel was still pressed by the piston. Thus, no layer of rubber compound could change its shape or realign. The sudden increase of barrel's temperature after the parison's movement (from 110°C to 160°C) also caused the curing mechanism to begin immediately and continue until the compound was completely vulcanized. Then, the barrel was opened to take out the rod of vulcanized compound out and the rod was sectioned as shown in Figure 3b. In the figure, the tops of the divergent annular die and convergent annular die show the cutting line of the vulcanized rubber. The vulcanized compound was cut into 2 equal pieces at 45° between of the center lines of the flow channel zone. The NR compound had full development of the flow without obstruction from the spider legs. After cutting, the vulcanized compound pieces were fine-polished. The flow pattern that occurred in the barrel was recorded for further study.

Flow properties measurement

The flow properties of the NR compound were studied using a divergent annular die and a convergent annular die with a die gap of 1 mm. The wall shear stress (τ_w) was calculated from the entrance pressure drop (ΔP_{ent}) measured under the test conditions in which extrudate swell measurements were taken using Equation 1. The wall shear rate ($\gamma^{\bullet}_w$) calculated from the average velocity ($\overline{v}$) as defined by the piston speed of the capillary rheometer and Power-law index (n), was determined using Equation 2[17].

$$\tau_w = \frac{H\Delta P_{ent}}{2L} \qquad (1)$$

$$\gamma^{\bullet}_w = -\left(1/n+2\right)2\overline{v}/H \qquad (2)$$

Where H = Die gap, L= Die land length

Measurements of the extrudate swell ratio

The outside diameter of the NR compound parison was measured indirectly method using a video-camera (1.3 MP USB DIGITAL MICROSCOPE 20X to 400X) to visualize the extrudate leaving the die exit. Results were recorded and displayed in real time using a personal computer. The outside diameter of the NR compound parison was carefully measured by replaying the recorded flow on a computer. It should be noted that the recording position of the NR compound parison was at 25 mm from the die exit in every experimental condition in order to avoid a draw-down effect. The NR compound parison was photographed and thickness was measured with *IMAGE J* computer program. The outside diameter and thickness of the NR compound parison were used to calculate swell values. The diameter swell ratio (B_D) and the thickness swell ratio (B_T) of the NR compound were calculated from Equation 3 and Equation 4 respectively**.** The diameter swell ratio was determined by ratio of the parison's diameter (D_p) to the outside diameter of the annular die (D_o). The thickness swell ratio was determined by the ratio of the parison's thickness (h_p) to the die gap of the annular die (h_o)[12].

$$B_D = \frac{D_p}{D_d} \qquad (3)$$

$$B_T = \frac{h_p}{h_d} \qquad (4)$$

Results and Discussion

Flow Properties of NR compound

Figure 4 shows a relationship between wall shear stress and wall shear rate of NR compound with and without TiO_2 extrudate through divergent annular die and convergent annular die with a die gap of 1 mm. In general, it was found that both types of NR compound showed non-Newtonian properties in pseudoplastic fluid flow behavior. At the same shear rate, it was found that the difference in shear stress of both types of NR compound was no more than 4.5%. It could be concluded that a mixture with TiO_2 did not affect overall flow property of NR compound[16]. Results from the measurement of flow property indicated that the wall shear stress from divergent annular die was higher than the wall shear stress from convergent annular die by an average of 6.5 % at every shear rate. This difference was consistent with results of previous research[14,18,19] which similarly described that the different flow property was affected by die design. That is, different flow channels of die directly affect the flow pattern of melt polymer flowing at the die entrance. For example, it can cause turbulent flow and vortex flow. These different flow patterns also have a direct effect on flow property. In this research, regarding flow pattern of NR compound at the entrance of divergent annular die, a vortex flow was found at the barrel wall more than in convergent annular die. This occurrence of vortex flow resisted the flow and caused difficulty for the NR compound flow. More extrusion force was needed, which could be evident in higher shear stress. This explanation is supported by results from an inspection of flow pattern

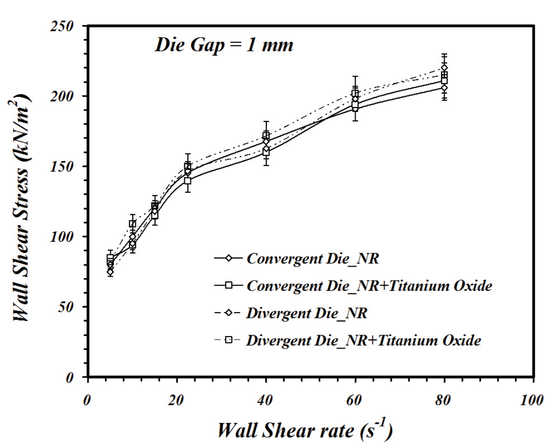

Figure 4. Flow curves of NR compound with and without addition of Titanium Oxide pigment from convergent and divergent annular die with the die gap of 1 mm.

comparing the two types of NR compound, which will be further discussed in the flow pattern analysis section.

Flow pattern development

Figure 5a shows flow pattern development of the NR compound at the extrusion range of 30 mm to120 mm in the barrel of the capillary rheometer with a shear rate of 1.5 s^{-1}, using convergent annular die with a die gap of 1 mm. Figure 5b shows flow pattern development of NR compound with the same test conditions of divergent annular die. Results indicate that the flow pattern of the NR compound in both types of die developed with an extrusion range which could be divided into 2 patterns: Axial Flow (AF) and Radial Flow (RF)[16]. The axial flow of both types of die had the same flow pattern. The flow pattern changed in accordance with extrusion range of the mobile piston. The NR compound had a parabolic-like flow pattern. That is, the melt flow velocity at the center of the barrel was higher than that at the barrel wall and flowed in the same direction as the mobile piston. Previous researches[16,20] has described a direct relation between a NR compound's flow pattern in the barrel and velocity profiles. The shape of the NR melt layer which

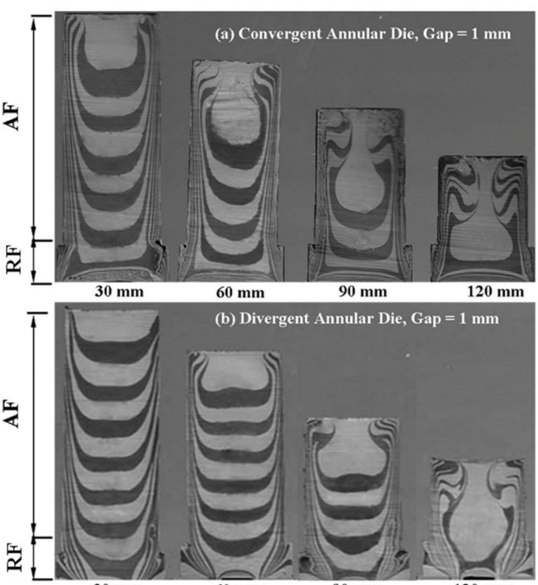

Figure 5. Flow patterns of NR compound in the barrels of convergent annular die system (a) and divergent annular die system (b) with different piston displacements [30 mm,60 mm, 90 mm, 120 mm] and shear rate of 1.5 s^{-1}.

developed into a parabolic-like flow pattern is a velocity profile of the melt layer flow in the barrel. Flow velocity could be calculated from the parabolic-shaped profile of melt layers and by directly counting the number of melt layer left in the barrel[18].

For the experimental results presented in this research, the assumptions of the previous researches mentioned above were applied to describe differences of velocity that occurred in both types of die. In general, regarding number of NR compound layer left in the barrel compared between convergent annular die and divergent annular die at die gap of 1mm and various shear rates and extrudate rates, number of melt layer left in the barrel of convergent annular die was less than number of melt layer left in the barrel of divergent annular die. Experimental results found at every shear rate are shown in Table 2. That is, the melt flow velocity of NR compound in the barrel of convergent annular die would be higher than that in the barrel of divergent annular die. This could be evaluated from a comparison of melt layer remaining in the barrel of both types of die at the same extrusion speed and extrusion displacement, such as at the shear rate of 1.5 s^{-1} die gap 1 mm and piston displacement of 60 mm. It was found that the convergent annular die had 9 layers of melt remained in the barrel whereas the divergent annular die had 13 layers of melt remaining in the barrel. Fewer layers remaining in the barrel of the divergent annular die indicate lower flow velocity at every test condition. This is because of different development of radial flow (RF) above the entrance of both types of die.

Figure 6 shows flow patterns of NR compound in the barrel of convergent annular die and divergent annular die at a shear rate of 1.5 s^{-1}, die gap of 1 mm and piston displacement of 60 mm. It can be seen that RF flow pattern development in both types of die occurred at the range of 20 mm above the die entrance. The direction of the flow was from center of the barrel to both barrel walls before flowing to the die, This pattern is called "cross flow"[18]. Moreover, this cross flow also caused two forms of Vortex Flow: Vortex Flow at Wall (VFW) and Vortex Flow at Die Entrance (VFE) in both types of die. The main difference was that in the convergent annular die, the VFE occurred most at the die entrance with no VFW at the barrel wall (Figure 6a). In divergent annular die, on the other hand, the VFW occurred most at the barrel wall with little VFE at the die entrance (Figure 6b). The different Vortex Flow of the two types of die significantly affected the flow velocity of the NR compound. This was because the VFW that occurred at the barrel wall of the divergent annular die obstructed the cross flow pattern development of the axial flow (AF) from the center of the

Table 2. Number of NR compound layer left in the barrel, compared between convergent annular die (CV) and divergent annular die (DV) (die gap = 1mm) at various wall shear rates and piston dissepiments.

Piston dissepiments	Number of melt layer remained in the barrel *vs.* shear rate							
	1 s^{-1}		1.5 s^{-1}		2 s^{-1}		2.5 s^{-1}	
	DV	CV	DV	CV	DV	CV	DV	CV
30 mm	16	15	14	13	12	11	10	9
60 mm	15	13	13	9	11	10	8	7
90 mm	9	7	7	5	5	4	3	2
120 mm	3	2	2	2	2	1	1	1

barrel to the radial flow (RF). This obstruction made it difficult for the compound to flow in the die channel. This is evident from the low-sloped parabolic form of flow layer remaining in the middle of the barrel. In the case of convergent annular die, no VFW occurred at the wall. This means the compound flowed easily and quickly in the die channel. Results clearly indicate the influence of vortex flow on flow velocity in both types of die. Moreover, this occurrence also affected extrudate swell properties of the NR compound, as will be discussed later.

Extrudate swell ratio of NR compound parison

Extrudate swell of the NR compound parison, both diameter swell and thickness swell, occurred in convergent annular die and divergent annular die with a die gap of 1 mm, 2 mm and 3 mm were shown in Figures 7a,b, respectively. Results of this study found that, the diameter swell and thickness swell of the NR compound in both types of die tended to increase with the increasing of shear rate, as expected. This is because the increase of shear rate (piston speed) led to the increase of shearing stress while the compound was flowing inside the die, as is clearly seen in Figure 4. Both diameter swell and thickness swell found in both types of die increased with the increase of the contraction ratio (decrease of the die gap) by about 10-15%. This is because the increase of the contraction ratio caused a greater degree of stretching in the direction of the flow and accumulation of elastic energy which would be released in the form of a greater swell rate[11].

These results also clearly found that the thickness swell was higher than the diameter swell in both types of die. In the convergent annular die, the amount of thickness swell was higher than the diameter swell by about 35% at every shear rate and die gap. In the divergent annular die, the thickness swell was higher than the diameter swell by about 28%. This could be explained by the flow pattern development in the rheometer barrel. That is, the flow pattern development in both types of die occurred from a change of flow direction from the center of the barrel to

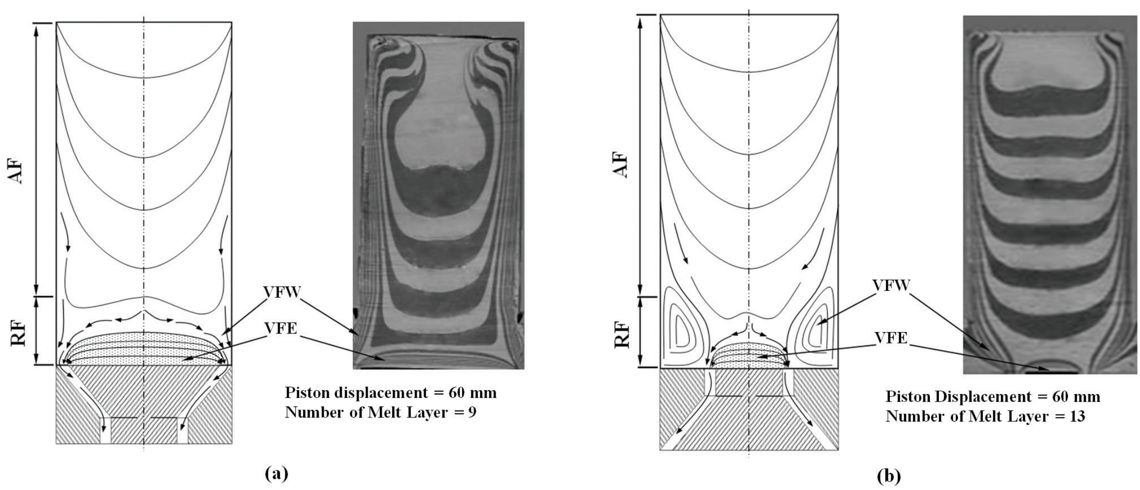

Figure 6. A schematic flow drawing for NR compound in the barrels of convergent annular die system (a) and divergent annular die system (b) with piston displacements of 60 mm.

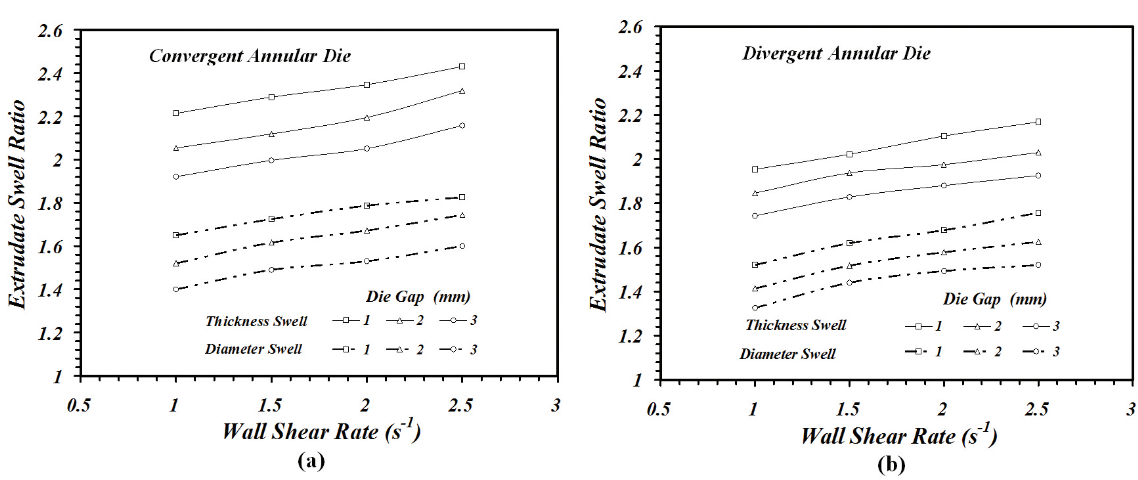

Figure 7. Diameter swell and thickness swell of NR compound flowed from the convergent annular die (a) and the divergent annular die (b) with different die gap [1 mm,2 mm, 3 mm].

the wall. This change stretched the molecular chain from the center of the barrel to the wall in a form of shear force, which was more pronounced around the top surface of the VFE. The VFE was, therefore, regarded as the parison's wall and caused the NR compound to accumulate more elastic energy than other areas. After passing through the die, NR compound in that area would become thickness of the parison. Hence, the accumulated elastic energy during NR compound flow would be used in recoiling of molecular chain and cause swell at the thickest side. This explanation corresponds with the work of Mu et al.[10], who studied shear stress distributions during the flow of melt polymer in annular die by using FES/BPNN/NSGA-II mathematical model. They found that the highest shear stress distribution occurred on the mandrel's surface. Work evidence that supports this explanation is that the divergent annular die had smaller (narrower) VFE than the convergent annular die. This implies that the amount of molecular chain stretching of the NR compound on VFE during flow from the center of the barrel to the wall of divergent annular die was lower than the flow in convergent annular die which had a larger (wider) VFE. This affected the amount of thickness swell as shown in Figure 7. It was found that the amount of thickness swell in the divergent annular die with the die gap of 1-3 mm was in a range of 1.74-2.16 only. On the other hand, the amount of thickness swell in the convergent annular die was higher in a range of 1.92-2.4 or over by an average of 10% at every shear rate.

Conclusion

The flow pattern and extrudate swell properties of NR compound were measured in a capillary rheometer using two types of annular die: convergent annular die and divergent annular die. Results of this study reveal that the flow pattern that occurred in the barrel of both types of die changed with the extrusion range. The flow pattern development in the die differed, particularly in the vortex flow occurring at the barrel wall and the die entrance. The different flow pattern in the barrel of both types of die significantly affected the diameter swell and the thickness swell of the NR compound. It was found that the amount of thickness swell was higher than diameter swell in every test condition. This difference could be explained with analysis of the complex flow pattern at the die entrance. Finally, the findings of this paper clearly indicate that the extrudate swell property of the NR compound is dependent on die geometry and die design.

Acknowledgements

The authors would like to thank the National Research Council of Thailand (NRCT) for financial support throughout this work.

References

1. Cogswell, F. N. - "Polymer Melt Rheology", George Godwin, London (1981). PMid:7267956.
2. Brydson, J. A. - "Flow Properties of Polymer Melts", Gorage Godwin, London (1981).
3. Sombatsompop, N. - J. Appl. Polym. Sci., **86**, p.509 (2002). http://dx.doi.org/10.1002/app.11015
4. Intawong, N.-T. & Sombatsompop, N. - Polym. Eng. Sci., **44**, p.2298 (2004). http://dx.doi.org/10.1002/pen.20257
5. Sombatsompop, N. & Sergsiri, S. - Polym. Advan. Technol., **15**, p.472 (2004). http://dx.doi.org/10.1002/pat.490
6. Intawong, N.-T.; Kimura, T.; Tamura, M.; Xiaojun, L. & Sombatsompop, N. - Polym. J., **37**, p.541 (2005). http://dx.doi.org/10.1295/polymj.37.541
7. Tanoue, S. & Iemoto, Y. - Polym. Eng. Sci., **39**, p.2172 (2004). http://dx.doi.org/10.1002/pen.11606
8. Müllner, H. W.; Eberhardsteiner, J. & Fidi, W. - Polym. Test., **26**, p.1041 (2007). http://dx.doi.org/10.1016/j.polymertesting.2007.07.003
9. Yousefi, A.-M.; Doelder, J.; Rainville, M.-A. & Koppi, K. A. - Polym. Eng. Sci., **49**, p.251 (2009). http://dx.doi.org/10.1002/pen.21251
10. Mu, Y.; Guoqun, M.; Yue, Z.; Xianghong, W. & Chengrui, Z. - Int. J. Adv. Manuf. Tech., **50**, p.517 (2010). http://dx.doi.org/10.1007/s00170-010-2556-z
11. Garcia, R. A. & Dealy, J. M. - Polym. Eng. Sci., **3**, p.158 (1982).
12. Koopmans, R. J. - Polym. Eng. Sci., **32**, p.1755 (1992). http://dx.doi.org/10.1002/pen.760322304
13. Garcia, R. A.; DiRaddo, R. W. & Ryan, M. E. - J. Non-Newton. Fluid., **60**, p.107 (1995).
14. Wood, A. K.; Read, A. G. & Lovegrove, J. G. A. - Plast. Rub. Proc. Appl., **12**, p.15 (1989).
15. Sombatsompop, N. & Intawong, N. T. - Mater. Res. Innov., **3**, p.150 (1999). http://dx.doi.org/10.1007/s100190050141
16. Intawong, N.-T.; Wongchaleo, C. & Sombatsompop, N. - Polym. Eng. Sci., **48**, p.1191 (2008). http://dx.doi.org/10.1002/pen.21074
17. Winter, H. H. & Fritz, H. G. - Polym. Eng. Sci., **26**, p.543 (1986). http://dx.doi.org/10.1002/pen.760260805
18. Sombatsompop, N. & Dangtungee, R. - J. Mater. Sci. Lett., **20**, p.1405 (2001). http://dx.doi.org/10.1023/A:1011695527853
19. Sombatsompop, N. & O-Charoen, N. - Polym. Advan. Technol., **14**, p.699 (2003). http://dx.doi.org/10.1002/pat.411
20. Sombatsompop, N. & Wood, A. K. - Polym. Eng. Sci., **37**, p.281 (1997). http://dx.doi.org/10.1002/pen.11670

Received: Mar. 1, 2014
Accepted: Apr. 8, 2014