รายงานฉบับสมบูรณ์

โครงการวิจัย ทุนพัฒนานักวิจัย RSA 16/2545
โครงการ พฤติกรรมบวมตัวเชิงรัศมีของพอลิเมอร์หลอมเหลวในเครื่องคาปิลารี่รีโอมิเตอร์

โดย

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พฤษภาคม 2547

สัญญาเลขที่ RSA 16/2545

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สนับสนุนโดยสำนักงานกองทุนสนับสนุนการวิจัย

(ความเห็นในรายงานนี้เป็นของผู้วิจัย สกว. ไม่จำเป็นต้องเห็นด้วยเสมอไป)

กิตติกรรมประกาศ

หัวหน้าโครงการฯ ขอแสดงความขอบคุณต่อสำนักงานกองทุนสนับสนุนการวิจัย (สกว.) เป็นอย่างยิ่งในการให้การสนับสนุนทุนวิจัยในการดำเนินการวิจัย หัวหน้าโครงการขอขอบคุณ นายนเรศ อินต๊ะวงษ์ นักศึกษาปริญญาเอก และนายศุภกิจ เศกศิริ นักศึกษาปริญญาโท คุณวุฒิสิทธิ์ ย่อชัย ผู้ช่วยนักวิจัย สำหรับความช่วยเหลือในการดำเนินการวิจัย และคุณปรียานุช รัชตะหิรัญ หัวหน้างานคลังคณะพลังงานฯ ที่ให้การช่วยเหลือในการบริหารการเงินของโครงการฯ

รศ.ดร. ณรงค์ฤทธิ์ สมบัติสมภพ
หัวหน้าโครงการ
สายวิชาเทคโนโลยีวัสดุ คณะพลังงานและวัสดุ
มหาวิทยาลัยเทคโนโลยีพระจอมเกล้าธนบุรี

สารบัญรายงาน

Abstract

Project Code: RSA 16/2545

Project Title: Radial Extrudate Swell Profiles of Flowing Polymer Melts in Capillary Rheometers

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Project Period: 1st November 2001 - 31st May 2004

Objectives:

1. To design and develop a parallel co-extrusion technique for simultaneous measurements of radial extrudate swell and velocity profiles of polymer melts flowing in a capillary Rheometer.
2. To investigate the effects of test conditions such as polymer type, die configurations and designs, die temperature, shear rate and electromagnetic flux density on radial extrudate swell and velocity profiles of polymer melts flowing in the capillary Rheometer.
3. To relate the flow behaviour, radial extrudate swell and velocity profiles of polymer melts in the capillary rheometer

Methodology, Results and Discussion

This research project determined the radial extrudate swell and velocity profiles of polystyrene melt flowing in a capillary Rheometer using a novel technique, so called "parallel co-extrusion technique (PCT)". The PCT technique allowed simultaneous measurements of extrudate swell and melt velocity across the die diameter. The test variables included shear rate, die designs and configurations, die temperature and use of electro-magnetized die.

Paper# 1: ***Journal of Applied Polymer Science,*** 87(10): 1713-1722 (2003)

Two capillary dies with different design configurations were used, one being single flow channel and the other being dual flow channel. It was found in this work that the power law index (n value) was the main parameter to determine the output rate ratio and the extrudate swell between the large and small holes for dual flow channel die, the greater the n value the lower the output rate ratio and thus decreased extrudate swell ratio. The differences in the extrudate swell ratio and flow properties for PS and LLDPE melts resulted from the output rate ratio and the molecular chain structure respectively. The extrudate swell was observed to increase with

wall shear rate. The discrepancies in the extrudate swell results from single and dual dies for a given shear rate were caused by differences in the flow patterns in the barrel and die, and the change in the melt velocities flowing from the barrel and in the die to the die exit.

Paper#2: ***Polymers for Advanced Technologies***, 14 (10): 699-710 (2003)

A dual channel die with mixed circular/slit flow channels in a constant shear rate rheometer were used. In single channel die, the extrudate swell of both PS and LLDPE melts in circular flow channel die was greater than that in slit flow channel, whereas, in dual channel die the slit channel exhibited a higher extrudate swell ratio, the results being explained by revealing the flow patterns of the melt in the barrel and die of the rheometer. The dimensionless size of the vortex flows near the entrance, and the extent of dis-entanglement of molecular chains on entering the die were the important factors for the differences in the extrudate swell ratios of the melts at the die exit influenced by the die designs used.

Paper#3: ***Polymers for Advanced Technologies*** (accepted)

The magnetic flux density, barrel diameter, extrusion rate and die temperature were varied to examine the extrudate swell behaviour and flow properties of a polystyrene melt. The results suggest that the maximum swelling of the polystyrene melt with application of the electro-magnetic field could be enhanced up to 2.6 times whereas that without the electro-magnetic field was 1.9 times. The barrel diameter of 30mm was found to be a critical value in case that the die swell ratio and flow properties of the PS melt were significantly affected by the magnetic flux density. The die swell at wall shear rates less than $11.2s^{-1}$ was caused by the magnetic torque, whereas at higher wall shear rates it was dependent on the shearing force. For a given magnetic flux density, the maximum increase in the die swell ratio as a result of the magnetic torque was calculated to be approximately 20%. Increasing the die temperature from 180 to 200°C reduced the overall die swell ratio and suppressed the effect of the magnetic flux density.

Paper#4: ***Polymer Engineering and Science***, (accepted)

A new experimental technique to *simultaneously* measure radial die swell and velocity profiles of polystyrene melt flowing in the capillary die of a constant shear rate Rheometer was. The proposed technique was based on parallel co-extrusion of colored melt-layers into uncolored melt-stream from the barrel into and out of the capillary die. The radial velocity profiles of the melt were measured by introducing relatively light and small particles into the melt layers, and the times taken for the particles to travel for a given distance were measured. The proposed experimental technique was found to be both very simple and useful for the simultaneous and accurate measurement of radial die swell and velocity profiles of highly viscous fluids in an extrusion process. The radial die swell and velocity profiles for PS melt determined experimentally in this work were accurate to 92.2% and 90.8%, respectively. The overall die swell ratio of the melt ranged from 1.25 to 1.38. The overall die swell ratio was found to increase with increasing piston speed (shear rate). The radial extrudate swell profiles could not be reasoned by the shear rate change, but were closely linked with the development of the velocity profiles of the melt in the die. The die swell ratio was high at the center (~1.9) and low (~0.9) near the die wall. The die swell ratio at the center of the die reduced slightly as the piston speed was increased.

Paper#5: ***Polymer Engineering and Science,*** (submitted)

An electro-magnetized capillary die was used to monitor the changes in the radial die swell profiles of the melt. The magnetic flux density applied to the capillary die was varied in a parallel direction to the melt flow, and all tests were performed under the critical condition at which sharkskin and melt fracture did not occur in the normal die. The experimental results suggest that the overall die swell for all shear rates increased with increasing magnetic flux density to a maximum value and then decreased at higher densities. The maximum swelling peak of the melt appeared to shift to higher magnetic flux density, and the value of the maximum swell decreased with increasing wall shear rate and die temperature. The effect of magnetic torque on the die swell ratio of PS melt was more pronounced when extruding the melt at low shear rates and low die temperatures. For radial die swell and velocity profiles, the radial swell ratio for a given shear rate decreased with increasing r/R position. There were two regions where the changes in the die swell ratio across the die diameter were obvious with changing magnetic torque and shear rate, one around the duct centre and the other around r/R of 0.65-0.85. In summary, the changes in the overall die swell ratio of PS melt in a capillary die were influenced more by the swelling of the melt around the centre of the die.

Conclusion and suggestions to further work

The extrudate swell was observed to increase with wall shear rate. The discrepancies in the extrudate swell results from single and dual dies for a given shear rate were caused by differences in the flow patterns in the barrel and die, the dimensionless size of the vortex and the change in the melt velocities flowing from the barrel and in the die to the die exit. The extrudate swell and velocity profiles of PS melt were affected by the magnetic field to the die. The barrel diameter of 30mm was found to be a critical value in case that the die swell ratio and flow properties of the PS melt were significantly affected by the magnetic flux density.

The Parallel Co-extrusion Technique (PCT) was found to be both very simple and useful for the simultaneous and accurate measurement of radial die swell and velocity profiles of highly viscous fluids in an extrusion process. The radial die swell and velocity profiles for PS melt determined experimentally in this work were accurate to 92.2% and 90.8%, respectively. The overall die swell for all shear rates increased with increasing magnetic flux density to a maximum value and then decreased at higher densities. The effect of magnetic torque on the die swell ratio of PS melt was more pronounced when extruding the melt at low shear rates and low die temperatures. The changes in the overall die swell ratio of PS melt in a capillary die were influenced more by the swelling of the melt around the centre of the die.

In the next step, the Parallel Co-extrusion Technique (PCT) should be used for measurements of radial extrudate swell and velocity profiles of flowing polymer melts in real processes like single and twin screw extruders, both with and without the application of the electromagnetic fields. The normal stress differences would be of interest under the applied magnetic field to the die used. (This is to perform in Advanced Research Scholar if granted).

Keywords: Capillary rheometer, Die/barrel designs, Polymer melts, Radial extrudate swell, Velocity profiles.

บทคัดย่อ

รหัสงานวิจัย:	RSA 16/2545
ชื่องานวิจัย :	พฤติกรรมบวมตัวเชิงรัศมีของพอลิเมอร์หลอมเหลวในเครื่องคาปิลารี่รีโอมิเตอร์
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วัตถุประสงค์ :

1. เพื่อออกแบบและพัฒนาเทคนิคการอัดรีดร่วมแบบขนาน (parallel co-extrusion) ในเครื่องคาปิลารี่รีโอมิเตอร์ สำหรับการวัดการบวมตัวเชิงรัศมี (radial extrudate swell profiles) และความเร็วยในการไหล (velocity profiles) ของพอลิเมอร์หลอมเหลวขณะไหล
2. เพื่อศึกษาพฤติกรรมการบวมตัวและความเร็วในการไหลเชิงรัศมีของพอลิเมอร์หลอมเหลวขณะอัดรีดในสภาวะต่าง ๆ เช่น ชนิดของพอลิเมอร์ ขนาดและรูปร่างของ die อุณหภูมิ อัตราเฉือน และสนามแม่เหล็กไฟฟ้า
3. เพื่อสร้างและเชื่อมโยงความสัมพันธ์ของพฤติกรรมการไหล รูปแบบการไหล และพฤติกรรมการบวมตัวเชิงรัศมี ของพอลิเมอร์หลอมเหลวในเครื่องคาปิลารี่รีโอมิเตอร์

วิธีการทดลอง ผลการทดลองและอภิปลายผลการทดลอง

งานวิจัยในโครงการพัฒนานักวิจัยนี้ เริ่มต้นจากการศึกษาอิทธิพลของการออกแบบหัวขึ้นรูปให้มีขนาดและรูปร่างต่าง ๆ เช่น หัวขึ้นรูปหน้าตัดกลมและหน้าตัดสี่เหลี่ยมผืนผ้า ระบบ die 2 ช่องการไหลทั้งที่หน้าตัดเหมือนกัน (เรียกว่า dual die) และหน้าตัดรูปร่างต่างกัน (เรียกว่า mixed cross-section die) และทำการวัดปริมาณการบวมตัวและสมบัติของพอลิเมอร์หลอมเหลว จากนั้น ได้ดำเนินการศึกษาเพื่อหาตรวจวัดปริมาณการบวมตัวเชิงรัศมี (radial die swell profiles) และรูปแบบความเร็วการไหลเชิงรัศมี (velocity profiles) ของพอลิสไตรีนในเครื่องทดสอบ capillary rheometer โดยได้มีการพัฒนาระบบการวัดแบบใหม่ที่เรียกว่า การอัดรีดร่วมแบบขนาน (Parallel Co-extrusion Technique; PCT) ซึ่งเทคนิคนี้สามารถตรวจวัดปริมาณการบวมตัวและความเร็วในการไหลของพอลิเมิอร์หลอมเหลวได้ตลอดพื้นที่หน้าตัดของหัวขึ้นรูป (die) ผลการวิจัย radial die swell profiles ที่เกิดขึ้นในโครงการพัฒนานักวิจัยนี้ ได้ถูกอธิบายโดยใช้ผลการวัดค่า radial velocity profiles ที่ได้ทำการวัดพร้อม ๆ กันภายใต้สภาวะการทดสอบต่าง ๆ เช่น การปรับเปลี่ยนอัตราเฉือน ขนาดและอุณหภูมิของ die ขนาดของห้องหลอมเหลว (barrel) และเป็นครั้งแรกที่ได้มีการใช้หัวขึ้นรูป (die)ที่มีความเป็นสนามแม่เหล็กไฟฟ้าในระบบการอัดรีดพอลิเมอร์ เป็นต้น ผลการวิจัยโดยสรุปพบว่า

- ระบบ parallel co-extrusion technique (PCT) สามารถใช้วัดปริมาณการบวมตัวและความเร็วในการไหลแบบแจกแจงเชิงรัศมีในเวลาเดียวกัน (simultaneously radial die swell and velocity profiles) ได้อย่างถูกต้องแม่นยำ และให้ค่าความถูกต้องของผลการวัดได้สูงกว่า 93%

- ระยะทางจากทางออกของ die ถึงจุดสมดุลย์ (equilibrium swelling) ของพอลิเมอร์หลอมเหลวลดลงเมื่อเพิ่มปริมาณอัตราเฉือน
- ปริมาณการบวมตัวของพอลิเมอร์หลอมเหลวตลอดพื้นที่หน้าตัดตามแนวรัศมีของ die ไม่เท่ากัน โดยพอลิเมอร์บริเวณตรงกลาง die จะมีปริมาณการบวมตัวที่สูงกล่าวในตำแหน่งอื่นๆ ทั้งนี้เนื่องมาจากการเปลี่ยนแปลงความเร็วในการไหลอย่างทันทีของพอลิเมอร์หลอมเหลวในตำแหน่งตรงกลางของ die ทำให้เกิดเป็นการขยายตัวอย่างรวดเร็วของสายอัดรีดพอลิเมอร์ (rapid extrudate expansion)
- การเปลี่ยนแปลงปริมาณการบวมตัวของพอลิเมอร์หลอมเหลวโดยรวม (overall die swell) ขึ้นโดยตรงกับอัตราอัดรีดที่ใช้ ในขณะที่ ปริมาณการบวมตัวของพอลิเมอร์หลอมเหลวเชิงรัศมีโดยเฉพาะในตำแหน่งตรงกลาง die (radial die swell at the centre of the die) ให้ผลในทิศทางตรงข้าม แต่สามารถอธิบายอย่างชัดเจนและอธิบายได้เชิงปริมาณ โดยการใช้ผลการวัดการเปลี่ยนแปลงรูปแบบความเร็วในการไหล (velocity profiles) ของพอลิเมอร์หลอมเหลว
- ปริมาณการบวมตัวของพอลิเมอร์หลอมเหลวเพิ่มสูงขึ้นมากกว่า 25% เมื่อใช้หัวขึ้นรูป die ที่มีค่าความหนาแน่นของสนามแม่เหล็กไฟฟ้าถึงจุดหนึ่ง จากนั้น ปริมาณการบวมตัวเริ่มลดลง โดยแนวโน้มการเปลี่ยนแปลงนี้สอดคล้องกับผลการวัด velocity profiles ของพอลิเมอร์หลอมเหลว โดยการเปลี่ยนแปลง radial die swell profiles และ velocity profiles สามารถอธิบายด้วย magnetic torque ที่เกิดขึ้นกับระบบพอลิเมอร์หลอมเหลวขณะไหลตัว นอกจากนี้ ยังพบว่า อิทธิพลของสนามแม่เหล็กไฟฟ้าที่มีต่อปริมาณการบวมตัวของพอลิเมอร์หลอมเหลวนี้ ขึ้นอยู่กับขนาดของห้องหลอมเหลว (barrel) ที่ใช้ โดยพบว่าที่ขนาดของห้องหลอมเหลว (barrel) ที่ 30 มม. ให้ผลของสนามแม่เห็นไฟฟ้าสูงสุด (ศึกษาขนาดของห้องหลอมเหลว (barrel) ระหว่าง 25 – 40 มม.)
- ผลงานวิจัยในโครงการทุนพัฒนานักวิจัยนี้ มีความสำคัญอย่างยิ่งกับ*กระบวนการผลิตประเภท* co-extrusion ตัวอย่าง เช่น หากทราบพฤติกรรมการบวมตัวของพอลิเมอร์เชิงรัศมี (radial extrudate swell profiles) เราสามารถควบคุมตำแหน่งและปริมาณของพอลิเมอร์ชั้นต่างๆ ในชิ้นงานที่ผลิตจาก co-extrusion process ได้อย่างถูกต้องและแม่นยำ
- จุดเด่นของผลงานวิจัยในโครงการทุนพัฒนานักวิจัยนี้ มีด้วยกัน 2 ข้อที่แตกต่างจากงานวิจัยอื่นๆ คือ ข้อแรก เป็นการพัฒนาเทคนิคที่สามารถทำการวัดค่า die swell และ melt velocity ได้พร้อมๆ กันในเชิงรัศมีตลอดพื้นที่หน้าตัดหัวขึ้นรูป (radial direction) ในเครื่อง capillary rheometer โดยเทคนิคใหม่ที่พัฒนาขึ้นนี้มีชื่อเรียกว่า Parallel Co-extrusion Technique (PCT) และข้อที่สอง การนำสนามแม่เหล็กไฟฟ้าเข้ามาร่วมใช้ในหัวขึ้นรูป (electro-magnetic die) เป็นครั้งแรกในการวัดในเครื่อง capillary rheometer

บทสรุปและข้อเสนอแนะและงานวิจัยในอนาคต

งานวิจัยนี้มีข้อสรุปที่ชัดเจนว่า ระบบ parallel co-extrusion technique (PCT) สามารถใช้วัดปริมาณการบวมตัวและความเร็วในการไหลแบบแจกแจงเชิงรัศมีในเวลาเดียวกันได้อย่างถูกต้องแม่นยำและให้ค่าความถูกต้องของผลการวัดได้สูงกว่า 93% ระยะทางจากทางออกของ die ถึงจุดสมดุลย์ (equilibrium swelling) ของพอลิเมอร์หลอมเหลวลดลงเมื่อเพิ่มปริมาณอัตราเฉือน ปริมาณการบวม

ตัวของพอลิเมอร์หลอมเหลวตลอดพื้นที่หน้าตัดตามแนวรัศมีของ die ไม่เท่ากัน โดยพอลิเมอร์บริเวณตรงกลาง die จะมีปริมาณการบวมตัวที่สูงกล่าวในตำแหน่งอื่น ๆ และสามารถอธิบายโดยการใช้ผลการวัดการเปลี่ยนแปลงรูปแบบความเร็วในการไหล (velocity profiles) ของพอลิเมอร์หลอมเหลว และพบว่าปริมาณการบวมตัวของพอลิเมอร์หลอมเหลวเพิ่มสูงขึ้นมากกว่า 25% เมื่อใช้หัวขึ้นรูป die ที่มีค่าความหนาแน่นของสนามแม่เหล็กไฟฟ้าถึงจุดหนึ่ง จากนั้น ปริมาณการบวมตัวเริ่มลดลง โดยสามารถอธิบายด้วย magnetic torque ที่เกิดขึ้น นอกจากนี้ ยังพบว่า อิทธิพลของสนามแม่เหล็กไฟฟ้าที่มีต่อปริมาณการบวมตัวของพอลิเมอร์หลอมเหลวนี้ ขึ้นอยู่กับขนาดของห้องหลอมเหลว (barrel) ที่ใช้ โดยพบว่าที่ขนาดของห้องหลอมเหลว (barrel) ที่ 30 มม. ให้ผลของสนามแม่เห็นไฟฟ้าสูงสุด

งานในอนาคต คือการใช้และพัฒนาเทคนิค Parallel Co-extrusion Technique (PCT) ซึ่งเป็นเทคนิคใหม่ ในการวัดค่า die swell และ melt velocity ในเครื่องมือผลิตผลิตภัณฑ์พอลิเมอร์ประเภท single screw extruder ซึ่งนับเป็นครั้งแรกที่ในวงการการผลิตและขึ้นรูปพอลิเมอร์ในการใช้หัวขึ้นรูปที่มีความเป็นสนามแม่เหล็กระหว่างทำการอัดรีดในกระบวนการอัดรีดแบบเกลียวหนอน และเป็นการศึกษาอิทธิพลของสนามแม่เหล็กที่มีต่อพอลิเมอร์ขณะหลอมเหลวและขณะไหล นอกจากนี้ ควรมีการใช้หัวขึ้นรูป (die) ที่มีความเป็นสนามแม่เหล็กไฟฟ้าที่สามารถปรับค่าความหนาแน่นสนามแม่เหล็ก (magnetic flux density) และทิศทางของสนามแม่เหล็ก (magnetic flux lines) ได้ และวัดค่า normal stress ที่เกิดขึ้นภายใน die ที่มีความเป็นสนามแม่เหล็กไฟฟ้า เพื่อใช้อธิบายผลการวัดค่า die swell และ melt velocity ซึ่งจากการค้นคว้าผลงานวิจัยในอดีต ยังไม่พบหลักฐานการปรากฏการวัดค่า normal stress ภายใต้สนามแม่เหล็กไฟฟ้านี้

คำสำคัญ คาปิลารี่รีโอมิเตอร์ พอลิเมอร์หลอมเหลว พฤติกรรมการบวมตัว ความเร็วในการไหล

หน้าสรุปโครงการ (Executive Summary)

โครงการวิจัยทุนพัฒนานักวิจัย RSA16/2545 นี้มีเป้าหมายเพื่อออกแบบและพัฒนาเทคนิคการอัดรีดร่วมแบบขนาน (parallel co-extrusion) ในเครื่องคาปิลารี่รีโอมิเตอร์ สำหรับการวัดการบวมตัวเชิงรัศมี (radial extrudate swell profiles) และความเร็วในการไหล (velocity profiles) ของพอลิเมอร์หลอมเหลวขณะไหล ในสภาวะต่างๆ เช่น ชนิดของพอลิเมอร์ ขนาดและรูปร่างของ die อุณหภูมิ อัตราเฉือน และสนามแม่เหล็กไฟฟ้า และเชื่อมโยงความสัมพันธ์ของพฤติกรรมการไหลรูปแบบการไหลและพฤติกรรมการบวมตัวเชิงรัศมีของพอลิเมอร์หลอมเหลวในเครื่องคาปิลารี่รีโอมิเตอร์ โครงการวิจัยนี้มีระยะเวลาดำเนินการที่ได้รับทุนวิจัยคือ ตั้งแต่ 1 พฤศจิกายน 2544 ถึง 31 ตุลาคม 2547 รวมเป็นเวลา 3 ปี ผลการดำเนินการวิจัยเป็นไปตามวัตถุประสงค์ที่ตั้งไว้ โดยมีผลสำเร็จเป็นรูปธรรมคือ ผลงานวิจัยที่ตีพิมพ์ในวารสารวิชาการนานาชาติ 5 เรื่อง และผลงานวิจัยเสนอในที่ประชุมนานาชาติ 1 เรื่อง (โปรดดูรายละเอียดในหัวข้อ Research Output) และได้ผลิตผู้ช่วยวิจัยในระดับปริญญาเอกและโทอย่างละ 1 คน

งานวิจัยในโครงการวิจัยนี้ การศึกษาอิทธิพลของการออกแบบหัวขึ้นรูปให้มีขนาดและรูปร่างต่างๆ เช่น หัวขึ้นรูปหน้าตัดกลมและหน้าตัดสี่เหลี่ยมผืนผ้า ระบบ die 2 ช่องการไหลทั้งที่หน้าตัดเหมือนกัน (เรียกว่า dual die) และหน้าตัดรูปร่างต่างกัน (เรียกว่า mixed cross-section die) และทำการวัดปริมาณการบวมตัวและสมบัติของพอลิเมอร์หลอมเหลว จากนั้น ได้ดำเนินการศึกษาเพื่อหาตรวจวัดปริมาณการบวมตัวเชิงรัศมี (radial die swell profiles) และรูปแบบความเร็วการไหลเชิงรัศมี (velocity profiles) ของพอลิสไตรีนในเครื่องทดสอบ capillary rheometer โดยได้มีการพัฒนาระบบการวัดแบบใหม่ที่เรียกว่า การอัดรีดร่วมแบบขนาน (Parallel Co-extrusion Technique; PCT) ซึ่งเทคนิคนี้สามารถตรวจวัดปริมาณการบวมตัวและความเร็วในการไหลของพอลิเมิอร์หลอมเหลวได้ตลอดพื้นที่หน้าตัดของหัวขึ้นรูป (die) ผลการวิจัย radial die swell profiles ที่เกิดขึ้นในโครงการพัฒนานักวิจัยนี้ ได้ถูกอธิบายโดยใช้ผลการวัดค่า radial velocity profiles ที่ได้ทำการวัดพร้อมๆ กันภายใต้สภาวะการทดสอบต่างๆ เช่น การปรับเปลี่ยนอัตราเฉือน ขนาดและอุณหภูมิของ die ขนาดของห้องหลอมเหลว (barrel) และเป็นครั้งแรกที่ได้มีการใช้หัวขึ้นรูป (die)ที่มีความเป็นสนามแม่เหล็กไฟฟ้าในระบบการอัดรีดพอลิเมอร์เป็นต้น ผลการวิจัยโดยสรุปพบว่า ระบบ parallel co-extrusion technique (PCT) สามารถใช้วัดปริมาณการบวมตัวและความเร็วในการไหลแบบแจกแจงเชิงรัศมีในเวลาเดียวกัน (simultaneously radial die swell and velocity profiles) ได้อย่างถูกต้องแม่นยำ และให้ค่าความถูกต้องของผลการวัดได้สูงกว่า 93% ระยะทางจากทางออกของ die ถึงจุดสมดุลย์ (equilibrium swelling) ของพอลิเมอร์หลอมเหลวลดลงเมื่อเพิ่มปริมาณอัตราเฉือน ปริมาณการบวมตัวของพอลิเมอร์หลอมเหลวตลอดพื้นที่หน้าตัดตามแนวรัศมีของ die ไม่เท่ากัน โดยพอลิเมอร์บริเวณตรงกลาง die จะมีปริมาณการบวมตัวที่สูงกล่าวในตำแหน่งอื่นๆ ทั้งนี้เนื่องมาจากการเปลี่ยนแปลงความเร็วในการไหลอย่างทันทีของพอลิเมอร์หลอมเหลวในตำแหน่งตรงกลางของ die ทำให้เกิดเป็นการขยายตัวอย่างรวดเร็วของสายอัดรีดพอลิเมอร์ (rapid extrudate expansion) การเปลี่ยนแปลงปริมาณการบวมตัวของพอลิเมอร์หลอมเหลว

โดยรวม (overall die swell) ขึ้นโดยตรงกับอัตราอัดรีดที่ใช้ ในขณะที่ ปริมาณการบวมตัวของพอลิเมอร์หลอมเหลวเชิงรัศมี โดยเฉพาะในตำแหน่งตรงกลาง die (radial die swell at the centre of the die) ให้ผลในทิศทางตรงข้าม แต่สามารถอธิบายอย่างชัดเจนและอธิบายได้เชิงปริมาณ โดยการใช้ผลการวัดการเปลี่ยนแปลงรูปแบบความเร็วในการไหล (velocity profiles) ของพอลิเมอร์หลอมเหลว ปริมาณการบวมตัวของพอลิเมอร์หลอมเหลวเพิ่มสูงขึ้นมากกว่า 25% เมื่อใช้หัวขึ้นรูป die ที่มีค่าความหนาแน่นของสนามแม่เหล็กไฟฟ้าถึงจุดหนึ่ง จากนั้น ปริมาณการบวมตัวเริ่มลดลง โดยแนวโน้มการเปลี่ยนแปลงนี้สอดคล้องกับผลการวัด velocity profiles ของพอลิเมอร์หลอมเหลว โดยการเปลี่ยนแปลง radial die swell profiles และ velocity profiles สามารถอธิบายด้วย magnetic torque ที่เกิดขึ้นกับระบบพอลิเมอร์หลอมเหลวขณะไหลตัว นอกจากนี้ ยังพบว่า อิทธิพลของสนามแม่เหล็กไฟฟ้าที่มีต่อปริมาณการบวมตัวของพอลิเมอร์หลอมเหลวนี้ ขึ้นอยู่กับขนาดของห้องหลอมเหลว (barrel) ที่ใช้ โดยพบว่าที่ขนาดของห้องหลอมเหลว (barrel) ที่ 30 มม. ให้ผลของสนามแม่เห็นไฟฟ้าสูงสุด (ศึกษาขนาดของห้องหลอมเหลว (barrel) ระหว่าง 25 – 40 มม.) ผลงานวิจัยในโครงการวิจัยนี้มีความสำคัญอย่างยิ่งกับ*กระบวนการผลิตประเภท* co-extrusion ตัวอย่าง เช่น หากทราบพฤติกรรมการบวมตัวของพอลิเมอร์เชิงรัศมี (radial extrudate swell profiles) เราสามารถควบคุมตำแหน่งและปริมาณของพอลิเมอร์ชั้นต่าง ๆ ในชิ้นงานที่ผลิตจาก co-extrusion process ได้อย่างถูกต้องและแม่นยำ

จุดเด่นของผลงานวิจัยในโครงการทุนพัฒนานักวิจัยนี้ มีด้วยกัน 2 ข้อที่แตกต่างจากงานวิจัยอื่น ๆ คือ ข้อแรก เป็นการพัฒนาเทคนิคที่สามารถทำการวัดค่า die swell และ melt velocity ได้พร้อม ๆ กันในเชิงรัศมีตลอดพื้นที่หน้าตัดหัวขึ้นรูป (radial direction) ในเครื่อง capillary rheometer โดยเทคนิคใหม่ที่พัฒนาขึ้นนี้มีชื่อเรียกว่า Parallel Co-extrusion Technique (PCT) และข้อที่สอง การนำสนามแม่เหล็กไฟฟ้าเข้ามาร่วมใช้ในหัวขึ้นรูป (electro-magnetic die) เป็นครั้งแรกในการวัดในเครื่อง capillary rheometer

Research Output ที่ได้จากงานวิจัยในโครงการ

ผลงานวิจัยจากโครงการวิจัย RSA 16/2545 มีดังนี้

Publication Level	Number of Papers	Publication Status
International Journals	5	4 published, 1 submitted
International Conferences	1	Published

ชื่อเรื่องของผลงานวิจัย และแหล่งที่ตีพิมพ์เผยแพร่ และค่า Impact Factors

International Refereed Journals

1. Experimental Studies on Extrudate Swell Behaviour of PS and LLDPE Melts in Single and Dual Capillary Dies - ***Journal of Applied Polymer Science,*** 87(10): 1713-1722 (2003) [Impact Factor = 0.927]
2. Extrudate Swell Behaviour of PS & LLDPE Melts in a Dual Die with Mixed Circular/Slit Flow Channels in an Extrusion Rheometer- ***Polymers for Advanced Technologies,*** 14 (10): 699-710 (2003) [Impact Factor = 1.019]
3. Die Swell Ratio of PS Melt From an Electro-Magnetized Capillary Die in an Extrusion Rheometer: Effects of Barrel Diameter, Shear Rate & Die Temperature-***Polymers for Advanced Technologies*** (accepted) [Impact Factor = 1.019]
4. A Parallel Co-Extrusion Technique for Simultaneous Measurements of Radial Die Swell and Velocity Profiles of a Polymer Melt in a Capillary Rheometer - ***Polymer Engineering and Science,*** (accepted) [Impact Factor = 0.890]
5. Experimental Studies on Radial Extrudate Swell and Velocity Profiles of Flowing PS Melt in an Electro-magnetized Die of Extrusion Rheometer- ***Polymer Engineering and Science,*** (submitted) [Impact Factor = 0.890]

International Conference Papers

1. Sombatsompop N (2003) Extrudate Swell of Thermoplastic Melt Systems in a Magnetic Capillary Die, 2^{nd} ***Energy and Materials Symposia***, 13-14^{th} March , Kyoto, Japan.

ภาคผนวก

เอกสารแนบในภาคผนวกนี้เป็นสำเนาบทความวิจัยที่ตีพิมพ์ในวารสารวิชาการที่ได้รับจาก โครงการวิจัยฯ ตามลำดับ ดังนี้

1. Experimental Studies on Extrudate Swell Behaviour of PS and LLDPE Melts in Single and Dual Capillary Dies - ***Journal of Applied Polymer Science,*** 87(10): 1713-1722 (2003) [Impact Factor = 0.927]
2. Extrudate Swell Behaviour of PS & LLDPE Melts in a Dual Die with Mixed Circular/Slit Flow Channels in an Extrusion Rheometer- ***Polymers for Advanced Technologies,*** 14 (10): 699-710 (2003) [Impact Factor = 1.019]
3. Die Swell Ratio of PS Melt From an Electro-Magnetized Capillary Die in an Extrusion Rheometer: Effects of Barrel Diameter, Shear Rate & Die Temperature-***Polymers for Advanced Technologies*** (accepted) [Impact Factor = 1.019]
4. A Parallel Co-Extrusion Technique for Simultaneous Measurements of Radial Die Swell and Velocity Profiles of a Polymer Melt in a Capillary Rheometer - ***Polymer Engineering and Science,*** (accepted) [Impact Factor = 0.890]
5. Experimental Studies on Radial Extrudate Swell and Velocity Profiles of Flowing PS Melt in an Electro-magnetized Die of Extrusion Rheometer- ***Polymer Engineering and Science,*** (submitted) [Impact Factor = 0.890]

Experimental Studies on Extrudate Swell Behavior of PS and LLDPE Melts in Single and Dual Capillary Dies

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ABSTRACT: Extrudate swell behavior of polystyrene (PS) and linear low-density polyethylene (LLDPE) melts was investigated using a constant shear rate capillary rheometer. Two capillary dies with different design configurations were used, one being a single flow channel and the other being a dual flow channel. A number of extrudate swell related parameters were examined, and used to explain the discrepancies in the extrudate swell results obtained from the single and dual flow channel dies, the parameters including output rate and output rate ratio, power law index, wall shear rate, wall shear stress, melt residence time, pressure drop induced temperature rise, flow channel position relative to the barrel centerline, and the flow patterns. It was found in this work that the power law index (n value) was the main parameter to determine the output rate ratio and the extrudate swell between the large and small holes for the dual flow channel die: the greater the n value the lower the output rate ratio and thus decreased extrudate swell ratio. The differences in the extrudate swell ratio and flow properties for PS and LLDPE melts resulted from the output rate ratio and the molecular chain structure, respectively. The extrudate swell was observed to increase with wall shear rate. The discrepancies in the extrudate swell results from single and dual dies for a given shear rate were caused by differences in the flow patterns in the barrel and die, and the change in the melt velocities flowing from the barrel and in the die to the die exit. © 2002 Wiley Periodicals, Inc. J Appl Polym Sci 87: 1713–1722, 2003

Key words: swelling; rheology; extrusion; processing; polymer melts; thermoplastics

INTRODUCTION

It has been accepted that die swell or extrudate swell is an important parameter determining the size and the quality of the extruded-polymer products. In polymer extrusion, the final shapes of the polymeric extrudate are not easily determined because of the swell phenomenon occurring while the melt is being forced out of the shaping die. The mechanism and degree of swelling of the extrudate are usually explained in terms of elastic recovery and/or residence time upon the applied stresses.[1]

The extrudate swell of polymer melts has been widely studied, mostly being performed in capillary rheometers. The swelling of a polymer melt during the flow is affected by many parameters such as shear rate, temperature, fillers, and die size and design. Among these parameters, die design has still gained interests from many researchers[2–8] due to the fact that the extrudate swell is closely associated with the behavior of the polymer melt flow, which is in turn very complex and three-dimensional.[2] The components and complexity of the flow include shear (both convergent and divergent), elongation, reversal, and the rapid velocity gradient near the die exit,[3] these being believed to be influenced by the design of the die being used, especially dies with irregular shapes. Kiriakidis and Mitsoulis[4] performed studies on extrudate swell of a high-density polyethylene melt by using slit and capillary dies with respect to the effect of die-length/diameter (gap) using the finite element method (FEM) and an integral constitutive equation. They found that for any given shear rate, the swelling was greater with the capillary die than the slit ones, and the extrudate swell decreased drastically as the die-length/diameter (gap) ratio were increased. Liang[5] investigated the influence of the die entry angle on the flow behavior for rubber compounds. They found that the shear stress and the melt viscosity reduced with increasing die entry angle to a certain value and then increased. He also found that the swelling of the extrudate decreased linearly with a length/diameter (L/D) ratio due to an increase in the

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melt residence time in the die. Lee and Ho[6] studied the effects of melting temperature and die length on the die swell behavior of a polystyrene melt to design a profile die, the experimental results being compared to the theoretical ones. It was found that the die swell became smaller as the melting temperature and die length were increased, the results from the experiment being in good agreement with those from the theory. Recently, Sombatsompop and Dangtugee[7] examined the effect of using two dies located along a barrel on the extrudate swell and flow patterns of natural rubber in the barrel of a capillary rheometer using a colored tracer method. They found that the change in extrudate swell was associated with the degree of flow complexities occurring in the barrel, residence flow time, elastic characteristic, and the temperature rise during the flow. The flows with more complexity tended to reduce the degree of extrudate swell of the rubber. Sombatsompop and Dantugee[8] also found that the change in the extrudate swell was linearly influenced by the entrance pressure drop at low actual barrel/die diameters (D_B/D_D from 20/4 to 30/7 mm/mm), and was then associated with the change in material viscosity at high barrel/die diameters (D_B/D_D from 35/7 to 40/8 mm/mm). For a constant barrel diameter, the smaller the die diameter the greater the extrudate swell due to the increases in the extensional deformation and wall shear rate coupled with a reduction in the melt residence time.

This article is part of an ongoing research program on investigations into the extrudate swell of polymer melts during the flow in rheometer. It is continued from a series of published work[7,8] aiming to study the extrudate swell and flow properties of polystyrene (PS) and a linear low-density polyethylene (LLDPE) melts in a rheometer using capillary dies with single and dual flow channels, the differences in the extrudate swell results due to the die designs used being of our main interest. The work covered the measurements and discussion on extrudate swell ratio, output rate and output rate ratio, power law index, wall shear stress and wall shear rate, material residence time and pressure drop induced temperature rise, the flow patterns in the barrel and die of the rheometer as well as the effect of flow channel position relative to the barrel centerline.

EXPERIMENTAL

Raw materials

The two thermoplastic melts used in this article were a polystyrene (Styron 656D 267) supplied in granular form by Siam Polystyrene Co., Ltd (Thailand), and a LLDPE (El-Lene L2009F) supplied in granular form By Thai Polyethylene Co., Ltd. (Thailand).

Experimental apparatus

The constant shear rate rheometer was used as employed in previous work.[7] The barrel, having 30 mm diameter and 150 mm long, was designed so that the die system could be easily changed. In this work, two circular dies with different flow channels were used, one being a single flow channel having L/D of 65.0/4.48 and the other being a dual flow channel having L/D of 65/4 and 65/2, the dies and their dimensions being shown in Figure 1. For the dual flow channel die, the channel L/D of 65/4 was referred to as *large hole* whereas that of 65/2 was referred to as *small hole.* In this work, the diameters of the two dies were intentionally designed to obtain the same cross-sectional area for comparison purposes, the sum of the cross-sectional area of the small and large holes in the dual channel die being equal to that of the single channel die. These two dies were also made of the same material (mild steel). A small pressure hole was located between the two die locations to detect the entrance pressure drop occurring, the entrance pressure being taken using the Pin-Spring pressure sensor.[9] The apparatus temperature was controlled using a Eurotherm 018 temperature controller, the apparatus temperature being of 180°C throughout this work.

Measurements and Calculations

Calculations of output rate (Q) and wall shear rate (γ_w)

Since the size of flow channels was varied depending on the design of the dies used, the output rate and wall shear rate for each flow channel at a given piston speed would be different, and they had to be calculated individually. For the single flow channel die, the total output rate was directly determined using the piston velocity and the barrel cross-section. In the case of dual flow channel die, the output rate of the melt through each flow channel (large and small holes) was determined based on the power law equations [eqs. (1) and (2)] and the fact that for a given piston speed the entrance pressure drop at small hole (ΔP_S) was equal to the entrance pressure drop at large hole (ΔP_L) as shown in eq. (3):

$$\tau = K\gamma^n \tag{1}$$

$$\Delta P = \frac{2KL}{R}\left(\frac{4Q}{\pi R^3}\right)^n \tag{2}$$

$$\Delta P_S = \Delta P_L \tag{3}$$

This gives

$$\frac{2KL_S}{R_S}\left(\frac{4Q_S}{\pi R_S^3}\right)^n = \frac{2KL_L}{R_L}\left(\frac{4Q_L}{\pi R_L^3}\right)^n \tag{4}$$

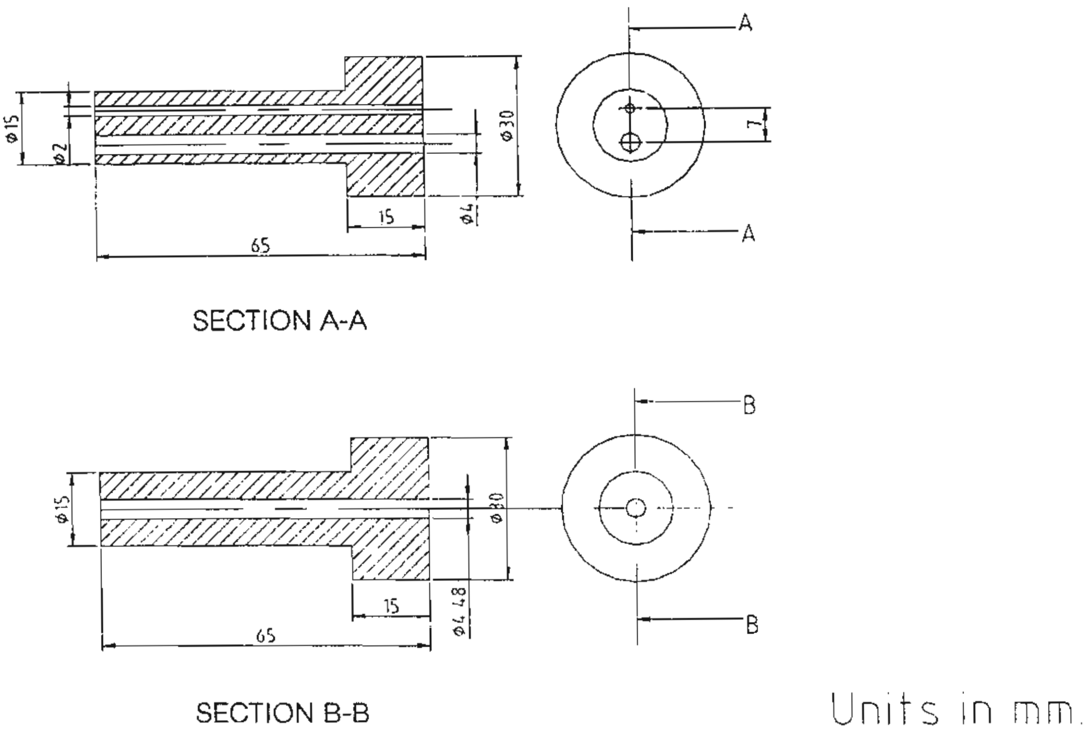

Figure 1 Die design and their dimensions (units in mm).

where K is the power law constant, n is the power law index, L and R are the length and radius of the die, respectively, and the subscripts S and L indicate small and large holes in dual flow channel die, respectively.

Equation (4) was reduced to give eqs. (5) and (6):

$$\frac{Q_S^n}{R_S^{3n+1}} = \frac{Q_L^n}{R_L^{3n+1}} \tag{5}$$

$$\frac{V_S^n}{R_S^{n+1}} = \frac{V_L^n}{R_L^{n+1}} \tag{6}$$

It should be noted that the power law index (n) was experimentally determined using the single channel die and the n values of PS and LLDPE melts were found to be 0.39 and 0.46, respectively. In this work, the power law index values of the melts in the dual flow channel die were assumed to be the same as those obtained in the single flow channel die, the assumption being based on the work of Drozdex and Faller,[10] who suggested that the n values between single and dual flow channel dies were very similar. As a result, the average melt velocities in the dual die for PS and LLDPE melts are calculated and shown in eqs. (7) and (8), respectively.

$$\text{For PS melt} \quad V_L = 12.02 V_S \tag{7}$$

$$\text{For LLDPE melt} \quad V_L = 8.89 V_S \tag{8}$$

By determining the average melt velocity in the small hole (V_S) and (V_L), the output rates at each channel (Q_S and Q_L) could be then determined, the sum of these two parameters equaling the total output rate (Q_t). The values of Q_t, Q_S, and Q_L were then used to determine the wall shear rate in single flow channel, small and large holes of dual flow channels respectively, by using eq. (9). Rabinowitsch corrections were applied to the shear rate data due to the use of different polymer melts.

$$\gamma_w = \frac{(3n+1)}{4n} \frac{4Q}{\pi R^3} \tag{9}$$

Extrudate swell measurement

In this work, the extrudate swell ratio (B) of the polymer melts was directly measured by calculating the ratio of diameter of the extrudate to that of the die, the extrudate diameter being based on the size of the extrudate diameter in the fully swollen (approximately 2 inches away from the die exit).[7] The test temperature was 180°C. By trial-and-error experiment, the critical shear rates for the onset of extrudate distortions (melt fractures) during extrusion were found to

TABLE I
The Output Rate and Wall Shear Rate of the Single and Dual Flow Channel Die for PS and LLDPE Melts

Total output rate (Q_i) in single die (10^{-9} m^3/s)	Wall shear rate in single die (s^{-1})		Output rate in dual die (10^{-9} m^3/s)				Wall shear rate in dual die (s^{-1})			
			PS		LLDPE		PS		LLDPE	
	PS	LLDPE	Small (Q_S)	Large (Q_L)	Small (Q_S)	Large (Q_L)	Small (γ_S)	Large (γ_L)	Small (γ_S)	Large (γ_L)
58.6	9.3	8.6	1.2	57.4	1.6	57.1	1.5	9.1	2.1	9.1
117.8	18.6	17.3	2.4	115.5	3.2	114.7	3.1	18.4	4.1	18.3
235.2	37.1	34.4	4.8	230.5	6.5	228.2	6.1	36.7	8.2	36.3
588.5	92.7	86.0	11.9	576.6	16.2	572.3	15.2	91.8	20.6	91.1
1177.7	185.6	172.2	23.9	1153.8	32.4	1145.4	30.5	183.6	41.2	182.3

be about 200 s^{-1} for both polymer melts used. In order to accurately measure the extrudate swell the wall shear rate used in this work ranged from 1 to 185 s^{-1} (corresponding to the piston speeds of 5–100 mm/min).

Entrance pressure drop (δP_{ent}) and wall shear stress (τ_w)

The entrance pressure drop was measured under the test conditions at which the extrudate swell measurements were taken. The wall shear stress (τ_w) of the polymer melt was calculated from the measured entrance pressure drop (ΔP_{ent}) using eq. (10).[8] In this work, Bagley's corrections were not applied to the shear stress data generated, the shear stress data solely being used for comparative reasons to illustrate the magnitude of the changes observed in the flow characteristics of the materials as a function of the design of the dies used.

$$\tau_W = \frac{R\Delta P_{ent}}{2L} \tag{10}$$

Melt residence time

It is widely accepted that the residence time (t_r) of the melt flowing in the die, being related to the relaxation of the polymer molecules, is one of the major factors that influences the extrudate swell. The residence time can be readily determined based on the die dimensions (L/D) and the wall shear rate (γ_w), this being expressed as[11]

$$t_r = 8\,\frac{(L/D)}{\gamma_w} \tag{11}$$

Pressure drop induced melt temperature rise

For a simple melt flow, the temperature increase (ΔT) of a polymer melt can be easily calculated using the entrance pressure drop (ΔP_{ent}) obtained under the conditions at which the extrudate swell was measured and the material characteristics are as shown in eq. (12)[12]:

$$\Delta T = \frac{\Delta P_{ent}}{\rho \cdot C_p} \tag{12}$$

where ρ is the melt density (1.03 g/cm^3 for PS and 0.92 g/cm^3 for LLDPE) and C_p is the specific heat of the melt (1970 J/kg K for PS and 3206 J/kg K for LLDPE).

It should be noted in this work that the melt temperature rise due to the viscous heating of the melt was not taken into account since the work was conducted under low shear rates. Previous work[13] on measuring true temperature rise of polymer melts resulting from shear heating effect has clearly suggested that at low shear rates (less than 200 s^{-1}) the temperature rise difference of the melt due to shear heating effect was very small, this being about 0–2°C. Besides, the determinations of ΔT value proposed in this work were only used to explain the differences in the extrudate swell between two different die systems (single channel and large hole in dual channel die) which had very similar (and low) shear rates. Therefore, the melt temperature rise caused by shear heating effect in this particular case could be neglected.

RESULTS AND DISCUSSION

Determinations of output rate, output rate ratio, and wall shear rate

Table I shows the output rate and wall shear rate of the single and dual flow channel dies for PS and LLDPE melts. It can be seen that the wall shear rates for PS and LLDPE melts in the single flow channel die were not the same, this being due to different n values as shown earlier. The wall shear rates for the single flow channel die was similar to that for the large hole in the dual flow channel die. In the dual flow channel die, the output rate and wall shear rate for the large hole was much greater than those for the small hole. In general, one may expect to obtain the opposite results due to the dimensions of the flow channel, the smaller

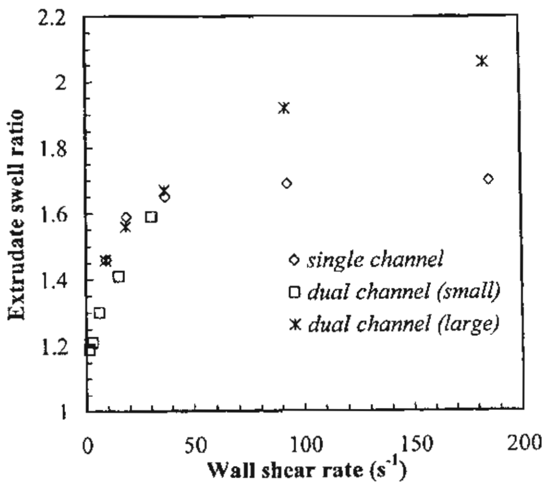

Figure 2 Extrudate swell of PS melt for single and dual flow channels.

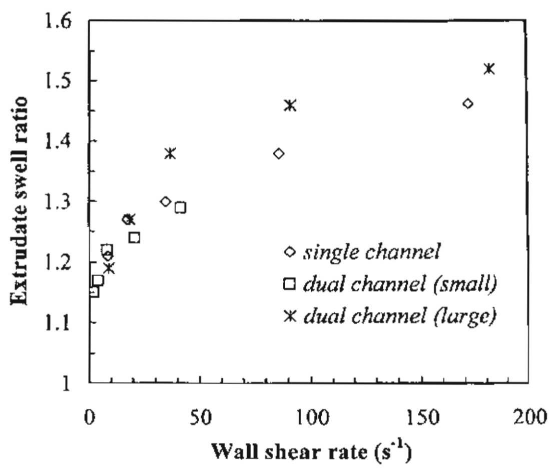

Figure 3 Extrudate swell of LLDPE melt for single and dual flow channels.

the flow channel diameter the greater the wall shear rate.[10] In this case, the output in the large hole of the dual channel die seemed to be the dominant factor to control and determine the wall shear rate. It was also interesting to note that the average output rate ratio (Q_L/Q_S) for PS and LLDPE melts were different, the Q_L/Q_S value in all output rates being 48.4 for PS and 35.3 for LLDPE, due to the fact that, according to eqs. (7) and (8), the output rate ratio was associated with the pseudoplastic characters (power law indexes) of these two fluids: the greater the power law index the lower the output rate ratio. This was in good agreement with the results reported by Drozdex and Faller.[10] In this work, we also found that the output rate ratio was linked with the extrudate well ratio of the melt, which is discussed later.

Extrudate swell behavior

Figures 2 and 3 show the relationship of extrudate swell changes against wall shear rate for the PS and LLDPE melts, respectively, using the single and dual flow channel dies. Generally, it can be seen that the extrudate swell increased with shear rate for both polymer melts and for all dies used. When comparing these two polymer melts, the PS melt had a greater swelling ratio than the LLDPE melt for both dies used. It was postulated for the dual flow channel die that this was linked with the output rate ratio as mentioned earlier: the greater the output rate ratio the higher the swelling in the dual channel die. In this case, the output rate ratio of the PS melt was greater than that of the LLDPE. When considering the extrudate swell in single and dual channel dies, it was found that the extrudate swell ratio for all dies was very similar at low shear rates (less than 40 s^{-1}, whereas the greater difference in the swelling ratio was observed at high shear rates (from ~40 to 185 s^{-1}). In the case of a small hole in the dual flow channel die, the shear rate to be reached was limited due to the operable range of the system. Possible explanations for the discrepancies in the extrudate swell results from single and dual (large) channel dies at higher shear rates are discussed as follows.

Differences in the flow properties

It has been known that variations of the extrudate swell are directly related to the change in the flow properties. In this work, the relationships between wall shear stress and wall shear rate of the two polymer melts were constructed under the conditions under which the extrudate swell was measured, the re-

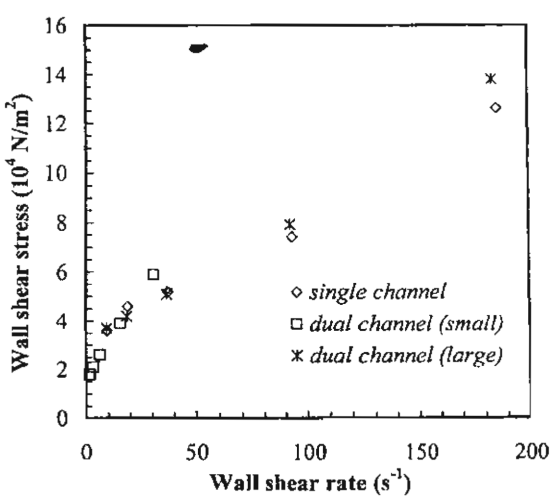

Figure 4 Flow properties of PS melt for single and dual flow channels.

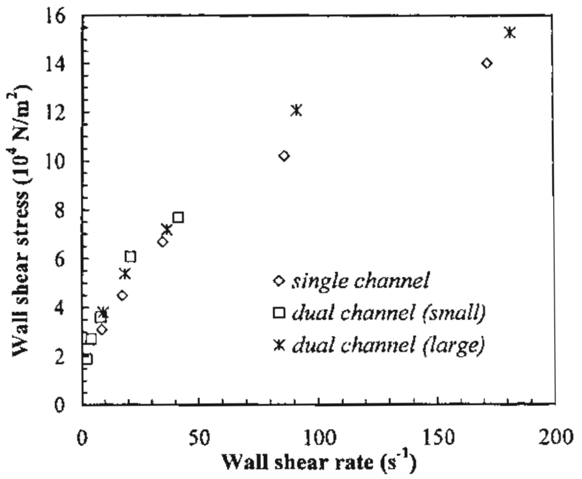

Figure 5 Flow properties of LLDPE melt for single and dual flow channels.

sults being shown in Figures 4 and 5 for PS and LLDPE melts, respectively. It can be seen that both polymer melts exhibited the pseudoplastic non-Newtonian behavior. The differences in the flow properties between PS and LLDPE melts for any given shear rates were due to differences in molecular chain structure. For both polymer melts, the differences in the flow properties between the single and dual channel dies were not as much as those found in the extrudate swell results (in Figs. 2 and 3). Therefore, the discrepancies in the extrudate swell results between single and dual channel dies in this case were not caused by the changes in the flow properties of the melts.

Entrance corrections

One would expect that the differences in the extrudate swell results might be caused by the entrance losses since different die sizes were used. In the general case, Bagley's methods were applied to correct the die entrance pressure drop, and the flow curves produced with different die sizes would then be superimposed, the true shear stress values for a given true shear rate from various die sizes used being less different.[2] In relation to the results in this work, if Bagley's corrections were applied, the flow data from the single and dual channel dies would be even more similar. Therefore, the entrance losses did not cause the extrudate swell differences.

Differences in melt residence time

It may be expected that if the melt has more time to flow in the die the degree of extrudate swell would reduce due to the stress relaxation.[7] In relation to the results in Figures 2 and 3, the residence times of the materials flowing in the single die should be greater than those in the large hole of the dual die due to less swelling of the extrudate. The residence time results are listed in Table II. The residence times of the PS and LLDPE melts were very similar for the two dies used except for the results from the small hole of the dual flow channel die. These results suggested that the discrepancies in the extrudate swells of the melts in the single and dual channel dies did not result from the differences in the residence time of the melts. Another interesting point to prove this explanation to be true was that, at low shear rate tests the differences in the residence time of the materials were very large (comparing between small and large holes), but the extrudate swell ratios were very similar.

It should also be noted that the differences in the residence times between PS and LLDPE melts were due to the differences in the n value (or flow properties of the two polymers) that were used to calculate the wall shear rates in eq. (11). In these particular circumstances, the power law indexes of PS and LLDPE, calculated from the flow curves in Figures 3 and 4, were found to be 0.39 and 0.46, respectively.

Pressure drop induced temperature changes

From previous work,[14,15] the melt temperature was usually expected to increase due to the shear heating and the development of the flow patterns of the material during the flow. The increase in the melt temperature would then result in the reduction of the elastic characters of the melt and thus reduced swell-

TABLE II
The Calculated Residence Times of PS and LLDPE Melts for Single and Dual Flow Channels

Total output rate	Single channel die		Dual channel die			
			PS		LLDPE	
(10^{-9} m^3/s)	PS	LLDPE	Small	Large	Small	Large
58.6	12.5	13.5	173.3	14.2	123.8	14.3
117.8	6.2	6.7	83.8	7.1	63.4	7.1
235.2	3.1	3.4	42.6	3.5	31.5	3.6
588.5	1.3	1.4	17.1	1.4	12.6	1.4
1177.7	0.6	0.7	8.5	0.7	6.3	0.7

TABLE III
The Calculated Temperature Rise of PS and LLDPE Melts for Single and Dual Flow Channels

Total output rate (10^{-9} m^3/s)	Temperature rise (°C) Single channel die PS	Single channel die LLDPE	Dual channel die PS	Dual channel die LLDPE
58.6	1.0	0.6	1.2	0.8
117.8	1.3	0.8	1.4	1.2
235.2	1.5	1.3	1.6	1.6
588.5	2.1	1.7	2.6	2.7
1177.7	3.6	2.7	4.5	3.4

ing of the extrudate. In this article, the temperature rises of the PS and LLDPE melts were determined using the entrance pressure drop, which were experimentally measured under the conditions used for the extrudate swell measurement, and the temperature results are shown in Table III. It can be seen that the maximum melt temperature rise ranged from 0.6 to 4.5°C, the differences in the melt temperature rise between the single and dual channel dies being very small (being less than 1°C in all cases). As a result, the melt temperature change was not the reason for the discrepancies in the extrudate swell due to the die designs being discussed.

The flow channel position relative to the barrel centerline

An independent investigation on the effect of flow channel position of dies relative to the barrel centerline on the flow properties and extrudate swell of the melts was conducted using a series of dies having the same size (L/D = 30/1.5) with a die temperature of 190°C. Each flow channel of dies was positioned at different points across the barrel diameter, the centerlines of the each die being 0.0, 0.75, 1.5, and 2.25 mm away from those of the barrel, and the flow properties and extrudate swell ratio being produced. The results are shown in Table IV. It can be seen that the flow properties and the extrudate swell of both LDPE and PS melts for the same shear rates did not change significantly with the flow channel position, the differences in the obtained results for each flow channel position being within the experimental errors (±2%). Therefore, it was suggested that the position of the die channel across the barrel diameter was not the cause of the differences in the extrudate swell results produced by dual and single flow channels. It should be noted that due to the limitation of the experiment regarding the size of the die flow channel and barrel diameter in the rheometer, the same L/D (i.e., 65/4.8) ratio of dies as in single flow channel die could not be used. The flow property results reported in this section aim only to explain the discrepancies of the extrudate swell results due to the single and dual flow channels qualitatively.

Proposed possible causes to the discrepancies in the extrudate swell from single and dual (large *hole) flow channel dies*

In this section, we offer some possible explanations for the discrepancies of the extrudate swell results due to

TABLE IV
Flow Properties and Extrudate Swell of PS and LLDPE Melts at Various Die Positions Across the Barrel Diameter: (a) PS Melt and (b) LLDPE Melt

(a) PS melt

Wall shear rate (s^{-1})	Wall shear stress (10^5 N/m^2) Die position away from the centre (mm) 0.0	0.75	1.5	2.25	Extrudate swell ratio Die position away from the centre (mm) 0.0	0.75	1.5	2.25
18	1.1	1.0	1.0	0.9	1.3	1.4	1.4	1.5
36	1.2	1.2	1.2	1.1	1.5	1.5	1.5	1.6
179	1.8	1.7	1.8	1.7	1.9	1.6	1.8	1.9
360	2.2	2.1	2.2	2.1	2.0	1.8	1.9	2.1
719	2.7	2.6	2.7	2.6	2.2	2.1	2.2	2.2
1078	2.9	2.9	2.9	2.9	2.3	2.2	2.2	2.3

(b) LLDPE melt

Wall shear rate (s^{-1})	Wall shear stress (10^5 N/m^2) Die position away from the centre (mm) 0.0	0.75	1.5	2.25	Extrudate swell ratio Die position away from the centre (mm) 0.0	0.75	1.5	2.25
18	0.9	0.8	1.5	1.2	1.3	1.3	1.3	1.3
36	1.4	1.5	1.9	1.7	1.4	1.4	1.4	1.4
179	2.7	2.9	3.0	2.9	1.5	1.5	1.5	1.5
360	3.5	3.7	3.8	3.7	1.5	1.5	1.6	1.5
719	4.4	4.5	4.3	4.4	1.7	1.7	1.7	1.7
1078	4.5	4.7	4.6	4.6	1.8	1.8	1.8	1.8

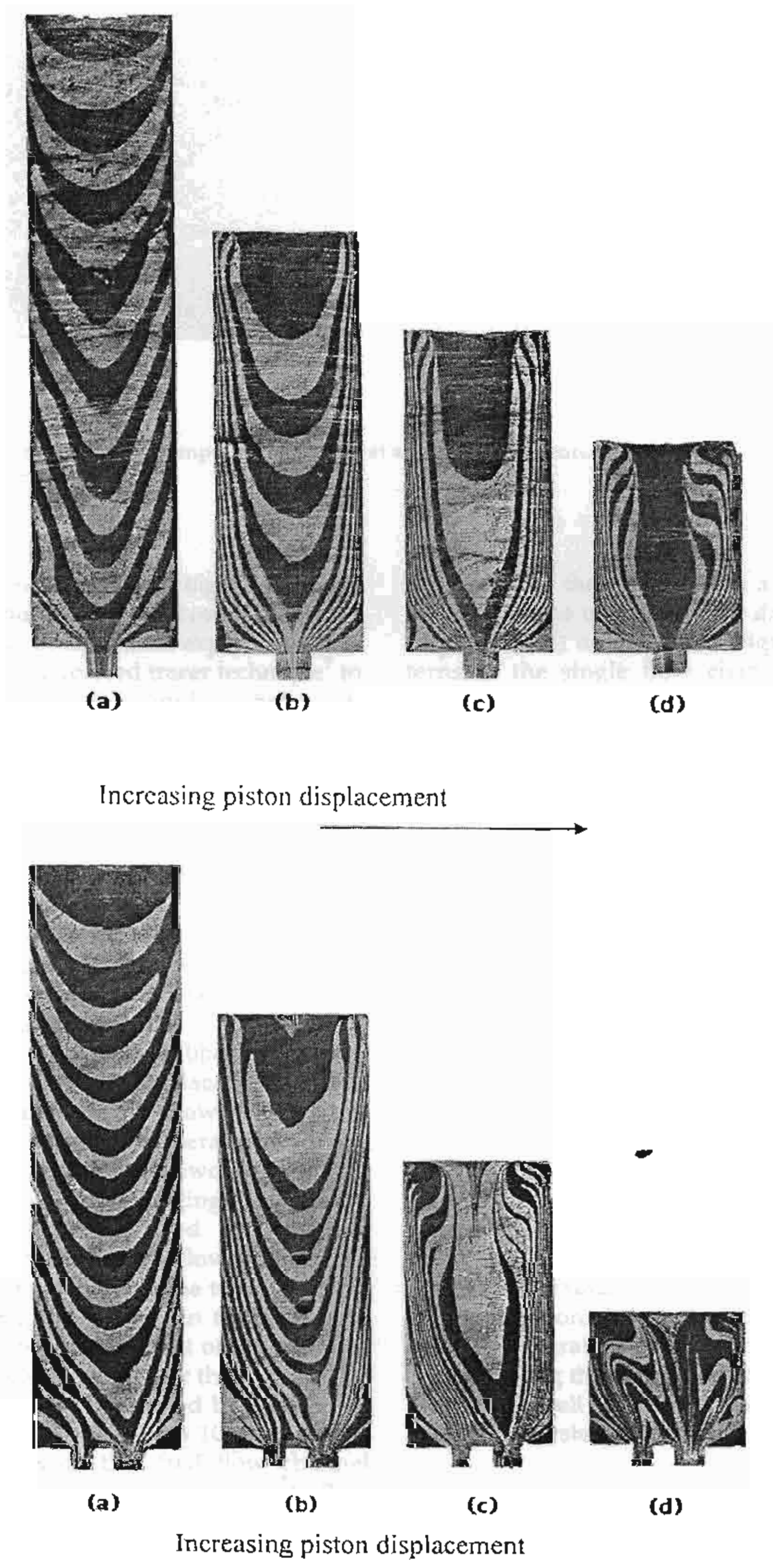

Figure 6 The flow patterns for NR compound in the barrel of capillary rheometer at different displacements down the barrel. [Piston displacements for (a)–(d): 10, 40, 70, and 100 mm.] Top: single flow channel; bottom: dual flow channel

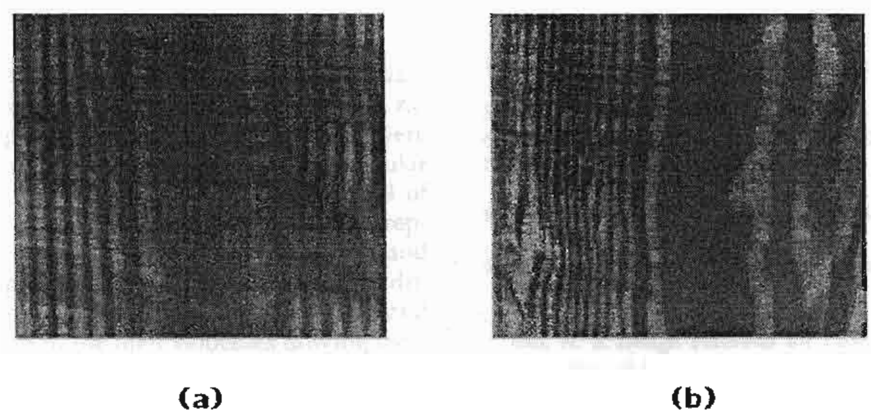

Figure 7 The flow patterns for rubber compound in the die at a piston displacement of 100 mm. (a) Single flow channel and (b) large hole of dual flow channel.

the single and dual flow channels at high shear rates (from 40 to 185 s^{-1}), this being carried out through the flow visualization. An independent experiment was then carried out using the colored tracer technique[7] to reveal the flow patterns in the capillary rheometer, especially at the die entrance. It should be noted that, in this case, we used a natural rubber (NR) compound for the flow pattern studies instead of either PS or LLDPE melt due to some limitations in the visualization technique used. Previous work[16] has indicated that the flow patterns for the NR and thermoplastic melts like LDPE, PS, and PP melts in the capillary rheometer were very similar in terms of qualitative consideration. The details of the experimental procedure including the preparation of the rubber compound can be found elsewhere.[1,7]

The flow pattern results for the rubber compound *in the barrel* at different piston displacements using single and dual flow channels are shown in Figure 6, which clearly indicates that the general style of the flow patterns in the barrel for the two die systems were very similar, the flows changing with piston displacement. This was supported by previous work,[1,7] which suggested that the flow pattern development in the barrel occurred due to the moving action of piston along the barrel.[1] In the dual flow channel (Fig. 6, bottom), the amount of the material flowing in the large hole was greater than that in the small hole, this being also confirmed by the output rate results as given in Table I. At a 100 mm piston displacement, the flow in the dual flow channel became very complex compared to the single flow channel (Fig. 6, top). It was interesting to observe that in all cases, the central velocity of the material flowing in the barrel in the dual flow channel die appeared to be higher than that in the single flow channel die, especially at high piston displacements. These complex and fast flows *in the barrel* of dual flow channel then resulted in a nonsymmetric flow patterns of the material *in the die* (at 100 mm piston displacement) as shown in Figure 7, the flow patterns in the single flow channel [Fig. 7(a)] being symmetric and stable whereas that in the large hole of dual flow channel [Fig. 7(b)] was not. Nonsymmetric and unstable flows in the dual flow channel die may be caused by the fast central flow of the melt in the barrel (Fig. 6, bottom) when entering the die, the velocity profiles of the melt in this die being then rearranged. In relation to the extrudate swell results, it was postulated that the material around the center of the barrel would also continue such high velocities along the die length (thus causing the flow complexity in the die), and would then decelerate as the melt exited from the die in order to equalize the velocity of the extrudate on the die exit. This sudden change in the melt velocities would then cause an increase in the cross-sectional area and thus greater extrudate swell, this phenomenon being the case for the large hole of dual flow channel. This explanation was supported by Chirstodoulou et al.[17]

Beyond the scope of this paper, the present work should be continued to quantitatively measure the exit pressure in order to calculate normal stress differences, recoverable shear strain as well as the velocity profiles along the die, which can be used to explain the extrudate swell behavior of the material for these particular die systems.

CONCLUSION

This article investigated the effect of flow channel characteristics of capillary dies on the extrudate swell of PS and LLDPE melts in a constant shear rate capillary rheometer. It was found that the power law index was the main parameter to determine the output rate ratio between the large and small holes in the dual flow channel die, the greater the power law index the lower the output rate ratio, and the lower extrudate swell ratio. The differences in the extrudate swell ratios and flow properties for polymer melts were caused by the output rate ratio and the molecular chain structure, respectively. The extrudate swell of the melts increased with wall shear rate. The discrepancies in the extrudate swell results from single and dual dies for a given shear rate were caused by differences in the flow patterns in the barrel and die, and the sudden change in the melt velocities flowing from the barrel and in the die to the die exit.

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Extrudate Swell Behavior of PS and LLDPE Melts in a Dual Die with Mixed Circular/Slit Flow Channels in an Extrusion Rheometer

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ABSTRACT

Extrudate swell behaviors of polystyrene (PS) and linear low-density polyethylene (LLDPE) melts in a dual channel die, having mixed circular/slit flow channels, in a constant shear rate rheometer were examined. The extrudate swell ratio for PS melt was observed to be higher than that for LLDPE melt for all cases, this being associated with the differences in molecular structures that could be described in terms of power law indexes and secondary flows near the die entrance. In single channel die, the extrudate swell of both PS and LLDPE melts in circular flow channel die was greater than that in slit flow channel, whereas, in dual channel die the slit channel exhibited a higher extrudate swell ratio, the results being explained by revealing the flow patterns of the melt in the barrel and die of the rheometer. It was found that the dimensionless size of the vortex flows near the entrance, and the extent of disentanglement of molecular chains on entering the die were the important factors for the differences in the extrudate swell ratios of the melts at the die exit influenced by the die designs used. Copyright © 2003 John Wiley & Sons, Ltd.

KEYWORDS: swelling; extrusion; processing; rheology; melt

INTRODUCTION

In extrusion processes, as a polymer melt is forced from a barrel through a die, the extrudate size is not equal to the die size. The ratio between the extrudate and the die size is known as "die swell" or "extrudate swell". Extrudate swell is an important parameter for determining the size and the quality of the extruded-polymer products, and it is useful for assessing the polymer elasticity in extrusion. Extrudate swell has been widely studied, mostly in capillary rheometers. The mechanism and degree of swelling of the extrudate are usually explained in terms of elastic recovery or residence time, the extrudate swell of a polymer melt being varied by shear rate, temperature, fillers, and die design [1].

When designing a die for polymer extrusion processes, the fundamental flow properties and the phenomenon of elastic swell of the molten

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polymer must be taken into account. Kiriakidis and Mitsoulis [2] performed studies on extrudate swell of a high-density polyethylene (PE) melt using slit and capillary dies with respect to the effect of die-length/diameter using the finite element method (FEM). They found that for any given shear rate, the swelling was greater with the capillary die than the slit one, and the extrudate swell decreased drastically as the die-length/diameter ratio was increased. Lee and Ho [3] studied the effects of die length on the die swell of a polystyrene (PS) melt to design a profile die. It was found that the die swell became smaller as the die length was increased, the results from the experiment being in good agreement with those from theory. Eggen and Hinrichsen [4] studied the effect of the die entrance angle and die length on the extrudate swell and on the onset of extrudate distortion in capillary extrusion. They found that the elongation component at the entrance region mainly influenced the extrudate distortion. Sombatsompop and Dangtangee [5] studied the extrudate swell and flow visualization of natural rubber in a capillary rheometer having two dies located along a barrel using a colored tracer method. The change in extrudate swell was found to be associated with the degree of flow complexities in the barrel, residence time, elastic characteristic and the temperature rise. They also found that the change in extrudate swell was linearly influenced by entrance pressure drop at low actual barrel/die diameters (D_B/D_D from 20/4 to 30/7 mm/mm), but was associated with the change in material viscosity at high actual barrel/die diameters (D_B/D_D from 35/7 to 40/8 mm/mm) [6]. For a constant barrel diameter, the smaller the die diameter the greater the extrudate swell due to the increases in the extensional deformation, shear rate, and a reduction in the melt residence time. Most recent work by Sombatsompop and O-Charoen [7] investigated extrudate swell behavior of PS and linear low-density polyethylene (LLDPE) melts in a rheometer having two circular dies with different designs (single and dual channels). They found that the power law index was the main parameter required to determine the output rate ratio and the extrudate swell in dual flow channel dies. The discrepancies in the extrudate-swell results from single and dual dies were caused by differences in the flow patterns in the barrel and die, and a sudden change in the melt velocities flowing from the barrel and into the die to the die exit.

It is reasonable to further examine the extrudate swell ratios and flow properties of PS and LLDPE melts with more complex die designs in the extrusion process, in order to obtain an in-depth understanding of flow behavior of polymer melts. In this article, two die configurations were used, one with single flow channel and the other with two (dual) flow channels, in order to assess the extrudate swell and flow properties of PS and LLDPE melts in a constant-shear rate extrusion rheometer. The single channel die used was either circular or slit in geometry, whereas the dual channel die comprised of both circular and slit flow channels within the same die. This was the first time that a dual die having two different channel geometries was used to study the extrudate swell behavior during the flow in an extrusion rheometer.

EXPERIMENTAL

Materials

A PS (Styron 656D 267), supplied in granular form by Siam Polystyrene Co., Ltd (Bangkok, Thailand), and a LLDPE (El-Lene L2009F), supplied in granular form by Thai Polyethylene Co., Ltd (Bangkok, Thailand) were used.

Experimental Apparatus

The constant shear rate rheometer was used as employed in previous work [7]. The barrel used had a 30 mm diameter (D) and 150 mm length (L). Two different die systems were used, and their designs and dimensions are shown in Figure 1:

(1) Single channel die: Two separate dies with different flow channel geometries were used, one being circular in cross-section and the other being slit in cross-section. The circular channel die had a L/D of 65.0/6.0. The slit channel die was 17.3 mm in width, 1.7 mm in height and 65 mm in length.
(2) Dual channel die: This die had two different flow channels (circular and slit cross-sections) within the same die. The circular channel had a L/D of 65/4 and the slit die was 12.2 mm in width, 1.2 mm in height and 65 mm in length.

All dies were made of mild steel (ST37). A small pressure hole was located near the die entrance to monitor entrance pressure drop occurring using a photo-conductive light pressure sensor [8]. The apparatus temperature was controlled using a Eurotherm 018 temperature controller, the apparatus temperature being 180 °C throughout this work.

Measurements and Calculations

Shear Rate (γ). Since the size of the flow channels was varied depending on the design of the dies used, the shear rate for each flow channel at a given piston speed would be different and had to be calculated individually:

(1) For single channel die, the shear rate was calculated from the total output rate (Q_t), directly determined from the piston velocity and the barrel cross-section, and the die channel dimensions.
(2) For dual channel die, the shear rate of the melt through each flow channel (both circular and slit) had to be calculated from the actual output for each flow channel (either $Q_{circular}$ or Q_{slit}), whose melt velocity had to be known. The calculations of the actual melt velocity for circular (V_C) and slit (V_S) channels were carried

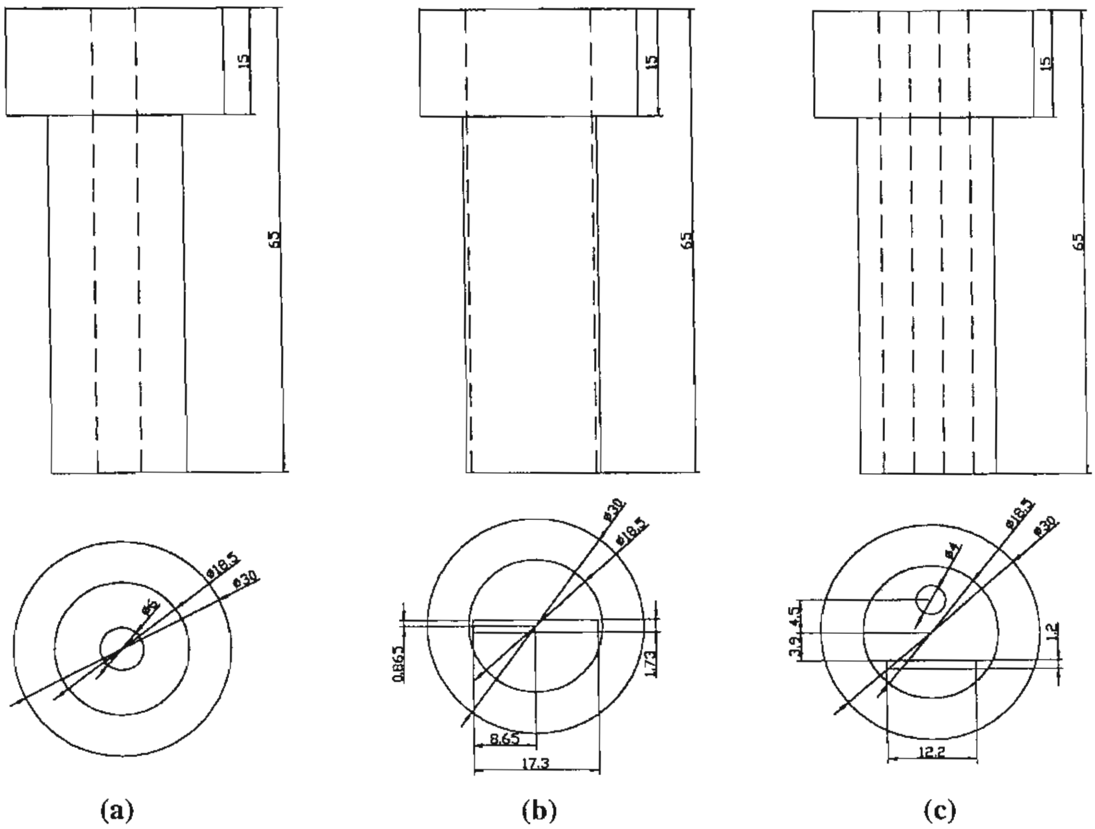


FIGURE 1. Die designs and dimensions (units in mm): (a) single channel die with circular cross-section; (b) single channel die with slit cross-section; (c) dual channel die with circular and slit cross-sections.

out through the power law equation expressed in eqs (1)–(3). The calculations of actual melt velocity for a given piston speed was based on the assumption that the entrance pressure drop at the circular channel (ΔP_C) was equal to that at the slit channel (ΔP_S). This assumption is supported by the work of Drozdek and Faller [9] as expressed in eqs (4) and (5).

$$\tau = K\gamma^n \tag{1}$$

For circular channel:

$$\Delta P = \frac{2KL_C}{R}\left(\frac{4Q_C}{\pi R^3}\right)^n \tag{2}$$

For slit channel:

$$\Delta P = \frac{2KL_S}{H}\left(\frac{6Q_S}{WH^2}\right)^n \tag{3}$$

$$\Delta P_C = \Delta P_S \tag{4}$$

In the dual die, this gives:

$$\frac{2KL_C}{R}\left(\frac{4Q_C}{\pi R^3}\right)^n = \frac{2KL_S}{H}\left(\frac{6Q_S}{WH^2}\right)^n \tag{5}$$

where K is the power law constant, n is the power law index (being determined using the single channel dies), L_C and R are the length and radius of the circular channel respectively, H, W and L_S are the height, width and length of the slit, respectively.

Equation (5) can be reduced to yield eq. (6):

$$\frac{(4V_C)^n}{R^{n+1}} = \frac{(6V_S)^n}{H^{n+1}} \tag{6}$$

Therefore, the actual melt velocity for circular and slit flow channels in a dual channel die for PS and LLDPE melts are calculated using eqs (7) and (8). For PS melt:

$$V_C = 24.3V_S \tag{7}$$

For LLDPE melt:

$$V_C = 11.1V_S \tag{8}$$

By determining the values of V_C and V_S from eqs (7) and (8) and knowing the total output rate (Q_t), the output rate at each channel (Q_C or Q_S) could then be determined, and so could the shear rate (eqs 9 and 10 for the calculations of true shear rate in circular and slit flow channels, respectively). Rabinowitsch corrections were applied to shear rate data due to the use of different polymer melts [1]. For circular channel:

$$\gamma = \frac{(3n+1)}{4n}\frac{4Q_C}{\pi R^3} \tag{9}$$

For slit channel:

$$\gamma = \frac{(2n+1)}{3n}\frac{6Q_S}{HW^2} \tag{10}$$

Wall Shear Stress (τ_w). The wall shear stress (τ_w) was calculated from the entrance pressure drop (ΔP_{ent}) which was measured under the test conditions at which the extrudate swell measurements were taken using eqs (11) and (12) for circular and slit flow channels, respectively. It should be noted that the pressure drop used for calculations of wall shear stresses for any given piston speed in circular and slit flow channels of dual channel die were the same [9]. In this work, Bagley's corrections were not applied to the shear stress data generated, as the shear stress data were solely used for comparative reasons to illustrate the magnitude of the changes observed in the flow characteristics of the materials as a function of the design of the dies used. For circular channel:

$$\tau_w = \frac{R\Delta P_{ent}}{2L} \tag{11}$$

For slit channel:

$$\tau_w = \frac{H\Delta P_{ent}}{2L} \tag{12}$$

Power Law Index (n). The power law index was calculated using eq. (13), which was related to the determination of the ratio (slope) of the shear stress and shear rate. The shear rate and shear stress data were obtained from eqs (9)–(12) [10].

$$n = \frac{\log[\tau]}{\log[\gamma]} \tag{13}$$

Extrudate Swell Ratio. During the experiment, the extrudate flowing out of the die appeared to be asymmetrical especially in the case of the slit flow channel. This made it very difficult to measure the dimension changes of the extrudate accurately. Therefore, the extrudate swell ratio (B) was determined using a weight difference method, whose calculation is expressed in eq. (14). The melt densities for PS and LLDPE melts were determined using a melt flow indexer at 180 °C, the experimental procedure following BS2782 Method 720A (1979). The melt densities of PS and LLDPE melts were 0.770 and 0.960 g/cm^3, respectively.

$$B = \frac{M}{\rho AL} \tag{14}$$

where A is the cross-sectional area of the die used (in mm^2), L is the length of the die used (in mm), M is the weight of the extrudate at L mm (in g), ρ is the melt density (in g/cm^3).

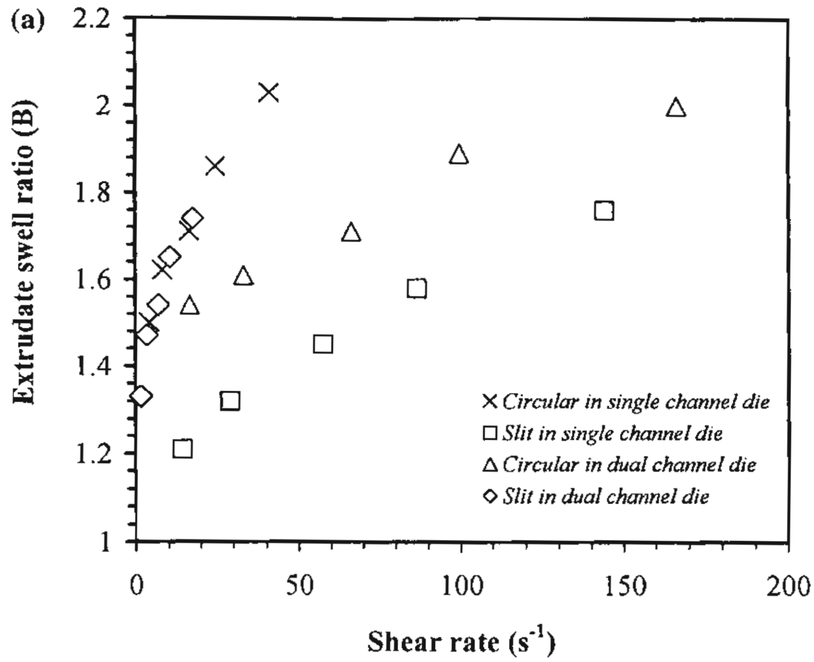
It should be noted that the extrudate samples collected for determining the swell ratio were in fully swollen state to ensure an equilibrium swelling, the extrudate being cut from the die lip approximately two inches away from the die exit [5]. Through trial-and-error experiments, the critical shear rates for the onset of melt fractures during extrusion were found to be about 180 and 150 sec^{-1} for PS and LLDPE, respectively. In order to accurately measure the extrudate swell, the shear rate used in this work ranged from 1 to 167 sec^{-1}.

RESULTS AND DISCUSSION

Figures 2(a) and 2(b) show the relationship between extrudate swell ratio (B) and the shear rate for PS and LLDPE melts respectively, using different flow channel and die configurations. It was generally found that the extrudate swell ratio increased with increasing shear rate as one would expect. Comparing the two melts, it is noticeable that the overall swell ratio for PS melt was greater than that for LLDPE melt, the extrudate swell ratios of PS and LLDPE melts ranging from 1.2 to 2.1 and from 1.1 to 1.7, respectively. The differences in the extrudate swell ratio of these two melts can be explained by two reasons, one being the differences in the power law index and the other being the size of vortex flow (stagnant flow) near the die entrance. As for the former reason, previous work [7] has suggested that a polymer melt with a greater power law index value tended to exhibit less swelling as it exited the die. Table 1 indicates that the power law index for LLDPE melt was slightly greater than that for PS melt. As for the latter case, experimental results by Dennison [11] and Binding [12] suggested, by visualizing the flow patterns for different types of polymer melts near the die entrance, that there were some secondary or stagnant flows observed for PS melt, but were not seen with LLDPE melt. Further work by Wong and Jaing [13] described that a polymer melt exhibiting secondary flows was bound to be more elastic, and thus resulting in greater swelling at the die exit. The above reasons should explain why the extrudate swell ratio for the PS melt was greater than that for the LLDPE melt found in this work. In addition, it should be noted that the sensitivity of the change of extrudate swell ratio to the shear rate for PS melt appeared to be greater than that for LLDPE melt. This was probably due to variations in the size of the secondary flows, as the shear rate was varied, near the die entrance of the PS melt, this not being the case for the LLDPE.

In terms of die design effect, the changes in the extrudate swell ratio for PS and LLDPE melts can also be explained in two separate cases as follows:

(1) *Case I—Single channel die.* This is quite a classical case in the literature [1–4, 14] where the extrudate swell of polymer melts has been investigated. In this article, the extrudate swell in the circular channel die was found to be much greater than that in the slit channel die, the effect being more pronounced for PS melt, the reason for which was discussed earlier. This



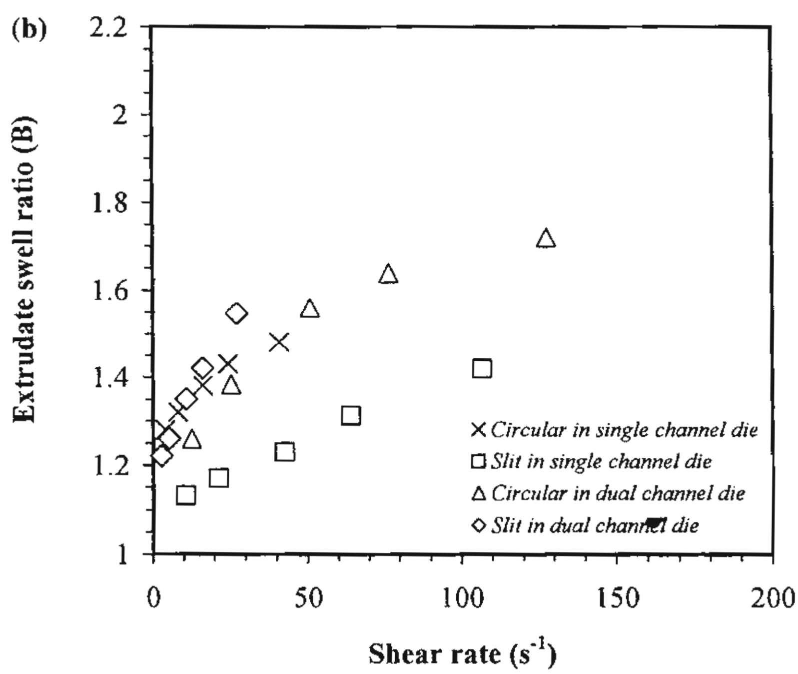


FIGURE 2. Extrudate swell ratio in single and dual channel dies: (a) PS melt; (b) LLDPE melt.

finding was in good agreement with the work of Kiriakidis and Mitsoulis [2] who examined the extrudate swell of a high-density polyethylene melt using slit and capillary dies. They found that for any given shear rate, the swelling was greater with the capillary die than the slit die.

(2) *Case II—Dual channel die.* As stated earlier, this was the first time that a dual die with two different flow channel geometries was used to investigate the extrudate swell ratio and the flow properties during melt extrusion. It is also important to note that this kind of rheometer system should be explained in connection with the synergetic effects between the circular and slit channels within the same die. From Figs 2(a) and 2(b), the extrudate swell ratios for the PS and LLDPE melts in the dual

TABLE 1. Power Law Index (n) for PS and LLDPE Melts in Circular and Slit Channels

Polymer melts	*Power law index (n)*	
	Circular	*Slit*
PS	0.39	0.23
LLDPE	0.46	0.34

channel die showed an opposite trend to those in the single channel die, and also to the previous work by Kiriakidis and Mitsoulis [2]. The extrudate swell ratio in slit flow channel found in this work was greater than that in circular flow channel for a given shear rate. It was also noticeable that the differences in the extrudate swell between these two channels was more pronounced for PS melt, the reason being discussed later. The possible causes for this observation can be explored as follows:

(a) *Differences in melt flow properties.* If the discrepancies in the extrudate swell ratio between the slit and circular channels were caused by changes in the flow properties one would expect to observe some differences in flow properties of the melts, between these two channels, to a similar extent as observed for extrudate swell ratio. Figures 3(a) and 3(b) show the variations in wall shear stress as a function of shear rate for PS and LLDPE melt respectively, using different flow channel die configurations. It can clearly be seen that the differences in the flow properties between these circular and slit channels were insignificant (this did not, however, mean that the flow properties of the melts in circular and slit channels were the same). Therefore, the flow property change was not the reason for such differences in the extrudate swell ratio as observed.

It is interesting to discuss the flow properties of PS and LLDPE melts in Figs 3(a) and 3(b), in terms of die design and polymer melt effects. It was observed that the design of the flow channel had less effect on the changes in flow properties as compared with the effect of polymer type. The discrepancies in flow properties of polymer melts due to die design has been well documented by other researchers [10, 15, 16]. For the effect of polymer type, the flow properties of the melts can be considered more clearly in terms of the power law index whose results have already been given Table 1. It can be seen that the power law indexes for PS and LLDPE melts were not the same due to differences in molecular structures, the n value for LLDPE melt being greater than that for PS melt for all cases. A higher power law index value means that the melt was more psuedoplastic non-Newtonian, which implied that the melt viscosity had a tendency to reduce more with applied shear rate. In the single channel die, the power law index in the circular flow channel was higher than that in the slit flow channel. The discrepancies in the power law indexes when using different designs of flow channels were probably caused by differences in the flow patterns

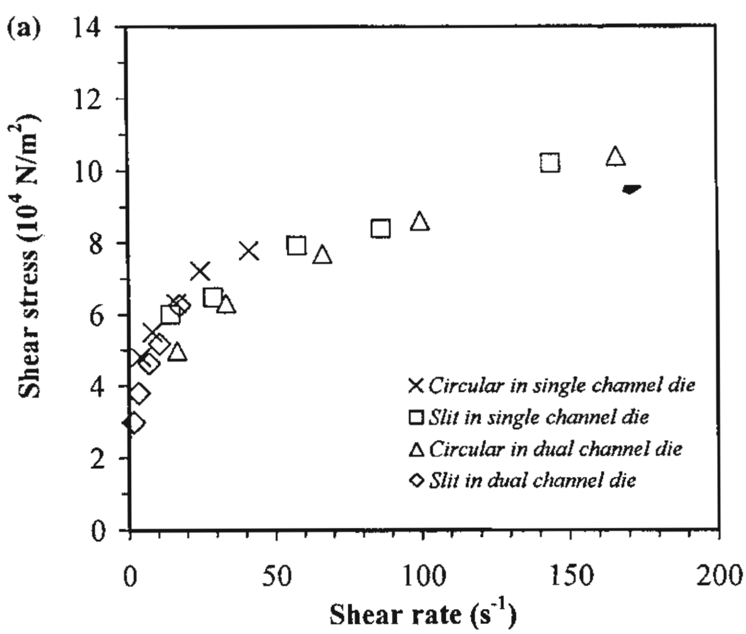


FIGURE 3. Shear stress v. shear rate in single and dual channel dies: (a) PS melt; (b) LLDPE melt.

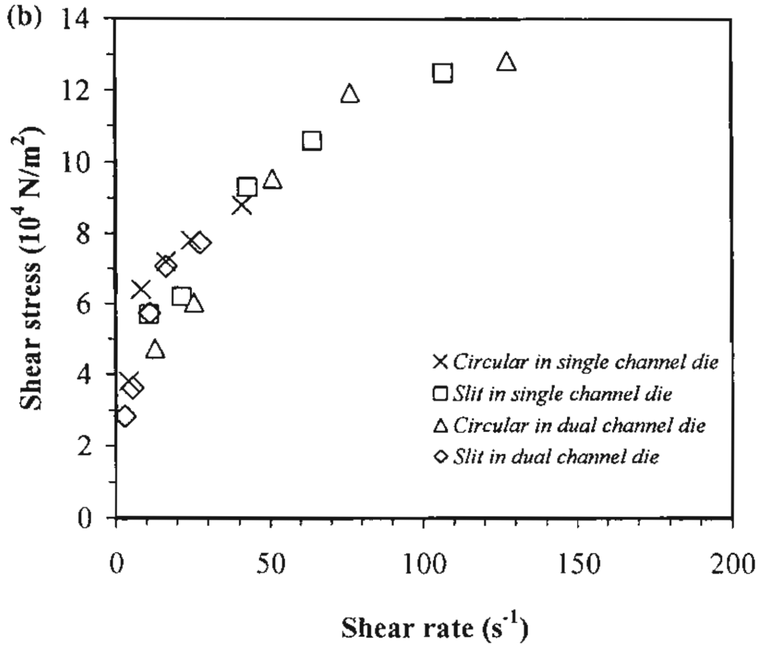


FIGURE 3. Continued

generated in each channel. It is widely known that a power law index varies with changing melt velocity profiles, which are directly influenced by the geometry of flow channels [1].

(b) *Entrance corrections.* One may expect that the differences in the extrudate swell results may be caused because Bagley's corrections were not used in this work, to analyze the true shear stresses, since different die geometries and sizes were utilized. In general, Bagley's corrections are applied to correct the die entrance pressure drop and the flow curves produced with different die sizes would then be superimposed, the true shear stress values for a given true shear rate from various die sizes used being less different [2]. In relation to this work, if Bagley's corrections had been applied, the flow property data of the melts for all flow channels used would either have been the same or even more similar to the existing ones (shown in Figs 3a and 3b). Therefore, the entrance corrections were not the case here.

(c) *Contraction ratio.* The dual die used was intentionally designed so that the cross-section areas of the circular and slit channels were the same. Therefore, the barrel-to-die contraction ratio was not the cause for the difference in the extrudate swell ratio between the circular and slit flow channels.

(d) *Residence time.* It is widely accepted that if a polymer melt has more time to flow in the die the extrudate swell ratio will reduce due to an increased stress relaxation time. In this work, the residence times of PS and LLDPE melts in circular and slit flow channels of dual die were determined, whose calculations can be obtained elsewhere [17], and the results are shown in Figs 4(a) and 4(b). The residence times of each melt at the two flow channels tended to be very similar if one has extrapolated the shear rate data. This suggests that the discrepancies in the observed extrudate swell ratio did not result from the residence time effect.

It is postulated in this work that a very high extrudate swell ratio for the slit flow channel in the dual die, as compared with that for the circular channel, may be explained in association with the flow patterns that had developed during the flows in the dual channel die. For this purpose, an independent experiment was carried out to reveal the flow patterns in the rheometer using the colored tracer technique [5]. In this case, a natural rubber (NR) compound was used for the flow pattern studies instead of either PS or LLDPE melt due to some limitations in the visualization technique used. Previous work [18] has clearly indicated that the flow patterns for the NR and thermoplastic melts like LDPE, PS and poly(propylene) (PP) melts in a capillary rheometer were very similar in terms of qualitative form. The detail of the experimental procedure including the preparation of the rubber compound can be found elsewhere [19]. Figure 5 shows the flow visualizations of NR compound in the dual channel die. It was observed that the flow

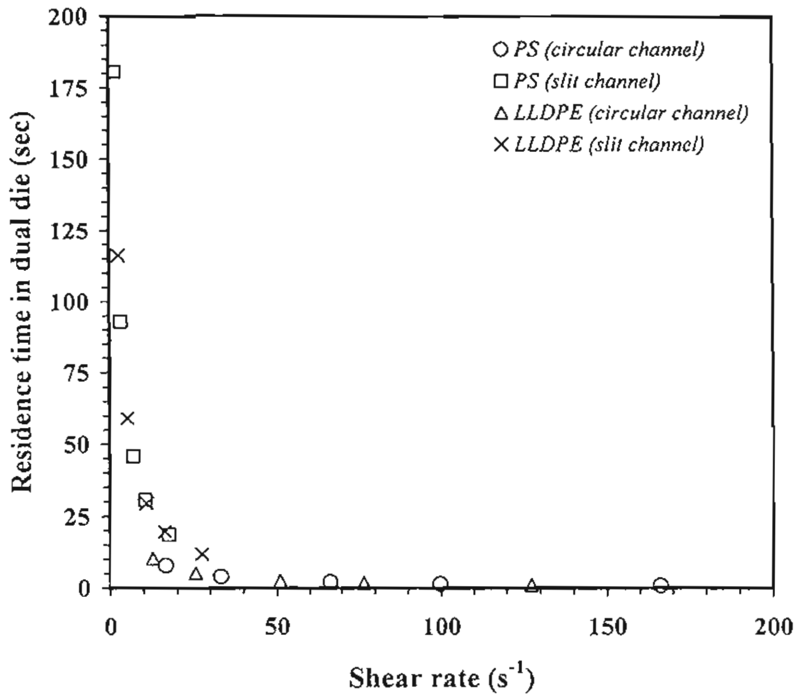


FIGURE 4. Melt residence times in dual channel die for PS and LLDPE melts.

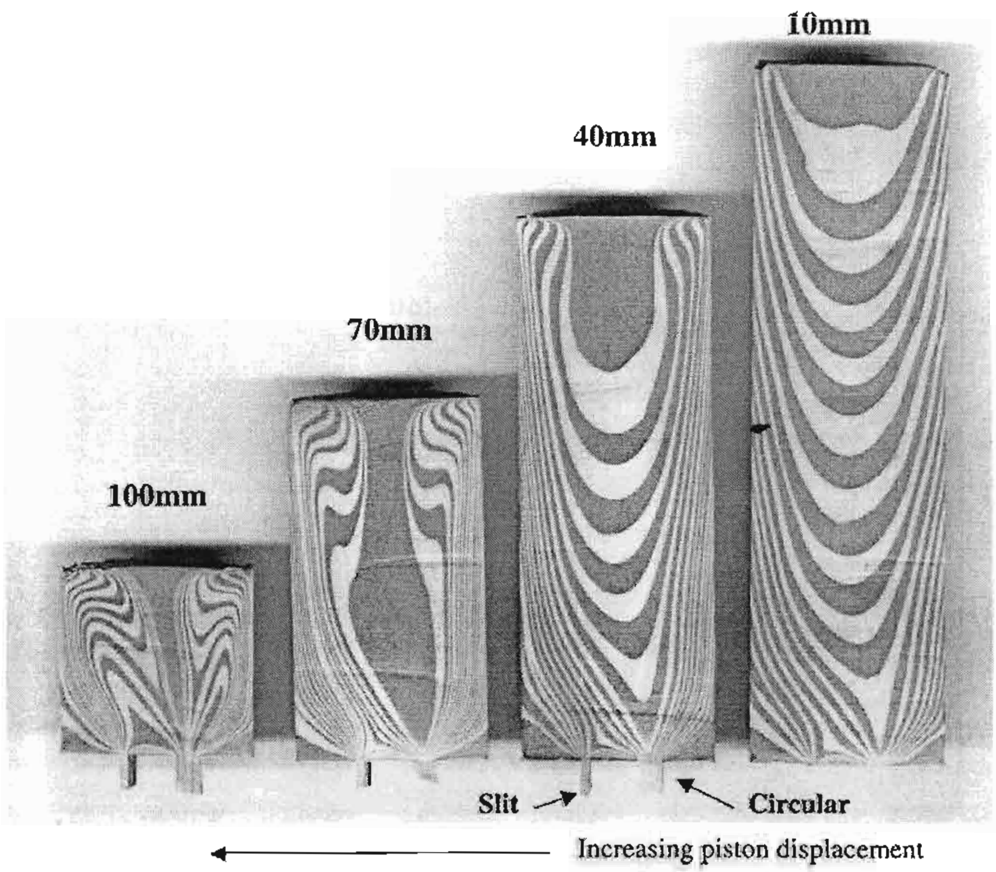


FIGURE 5. Flow patterns for NR compound in the barrel and die entrance for different piston displacements (10–100 mm down the barrel) in dual die containing circular and slit flow channels.

patterns were complex and changed with piston (or extrusion displacement), and the material at each radial position of the barrel flowed down the barrel, and moved radially towards the channels of the die—information on the flow patterns of NR in the barrel is given elsewhere [19]. It can be seen that the majority of the material flowed into the circular channel, suggesting that the overall flow rate (and thus output rate) of the material in the circular channel was much greater than that in the slit channel. According to eqs (11) and (12), the melt flow rates in the circular channel were approximately 24 and 11 times greater than those in the slit channel, respectively. Besides, the striation flow layers (near the die entrance) on the circular channel side were much thinner than those on the slit channel side, which substantiated the higher flow rate of the material in the circular channel. It was thought that the differences in flow rates of the material between the circular and slit channels were associated with the design of the flow channels used, and these were possibly attributed to the differences in the extrudate swell ratio of these two channels as observed earlier on. It was felt that, with this specific die design, the flows in both circular and slit channels had an effect on one another. Considering the nature of a slit die (being defined as height at least 10 times smaller than width), it was difficult for the melt to flow into such a small die height (1.2 mm). To be able to flow into the slit channel, the molecular chains of the melt had to be more disentangled (uncoiled), whereas, in the circular flow channel (4 mm in diameter) with an easier material flow, the polymer chains were less disentangled on entering the channel. It is reasonable to suggest that the flow with a higher degree of molecular disentanglement on entering the die would give rise to higher swelling of the extrudate at the die exit due to molecular re-coiling. This was one of the reasons why the extrudate swell ratio of the melt in the slit channel was greater than that in the circular one. It could be said that the design of the flow channels influenced the flow-ability and thus the swelling ratio of the melt. This speculation was verified by considering the flow patterns of the rubber compound using another dual die, which was specially designed and manufactured in this work, having circular and rectangular channels. The slit channel was replaced by use of a rectangular channel whose width and height were equal (being $3.7 \times 3.7\,mm^2$). The results are visualized in Fig. 6. It can be seen that the flow patterns of the melt in the circular/rectangular die were different from those in the circular/slit die, especially the central flow and the striation flows near the die entrance. It was clearly

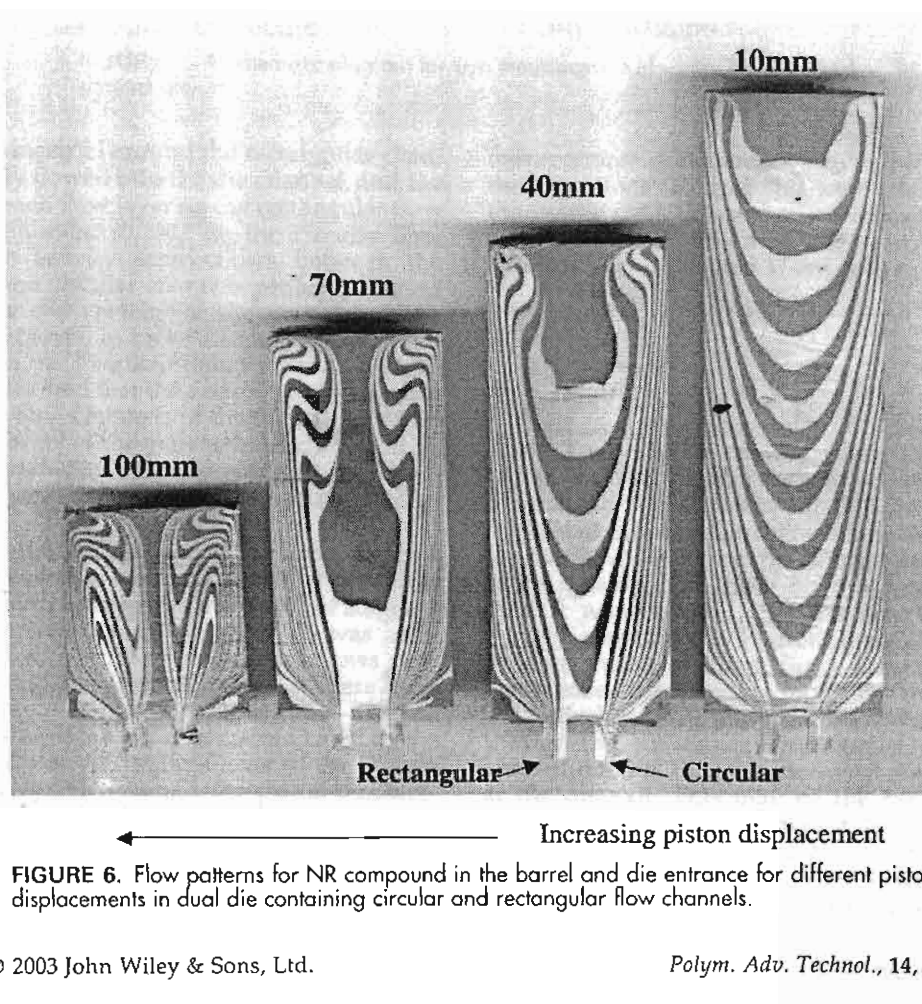


FIGURE 6. Flow patterns for NR compound in the barrel and die entrance for different piston displacements in dual die containing circular and rectangular flow channels.

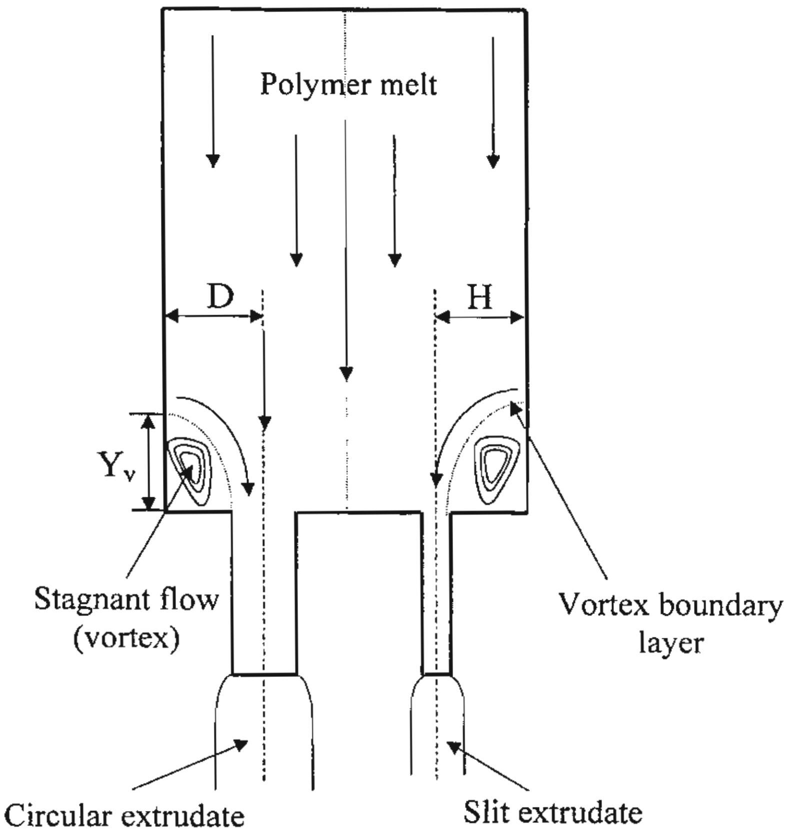


FIGURE 7. A schematic diagram for size measurement of vortex flows in dual channel die.

seen that the material around the rectangular channel now easily flowed into the die channel, and the thickness of each flow layer on the rectangular flow channel was similar to that on the circular flow channel. The equally easily flows between the rectangular and circular channels probably caused a decrease in the velocity of the material at the centre of the barrel, in order to maintain the total output of the flow. The flow pattern results in Fig. 6 evidently confirmed that the design (actual size and shape) of the flow channels of the die had an effect on the flow-ability of the melt near the die entrance, which in turn affected the molecular entanglements (orientations) and the amount of extrudate swell at the die exit.

Another synergetic effect of the flow patterns explaining the differences in extrudate swell ratio in slit and circular channels was the vortex or stagnant flow size near the die entrance, which was associated with the occurrence of stagnant flows near the die entrance. As observed in Fig. 5, there were some stagnant flows near the die entrance for both circular and slit channels. These are indicated by the unchanged size of the 1st flow layer at the bottom corner of the barrel although the piston displacement was increased, while the thickness and positions of other flow layers (from 2nd to nth layers) seemed to change with increasing piston displacement. Figure 7 shows a schematic diagram for measurement of the vortex size in the dual channel die. Equations (15) and (16) were used to calculate the vortex sizes (X) of the flow in the circular and slit channels in the dual die, respectively. The definitions of Y_v, D and H are seen in Figure 7.

$$X = \frac{Y_V}{D} \tag{15}$$

$$X = \frac{Y_V}{H} \tag{16}$$

Table 2 shows the dimensionless sizes of some vortex boundary layers (layers 2nd–4th), measured from the flow patterns in Fig. 5. Considering layer nos 2–4, it is noticeable that the sizes of the vortices for the flow layers on the slit channel side were twice greater than those on the circular channel side. It was believed that the flow with greater vortices was likely to exhibit higher elastic character and greater swelling. Since this dimensionless vortex size was related to molecular orientations of the extrudates [13], the greater the vortex size the greater the molecular orientations as the melt entered the die, thus the increased extrudate swell at the die exit. This may be the reason why the differences in extrudate swell ratio in the circular and slit channels were more pronounced in the case of PS melt, as mentioned earlier. It was also noted that the vortex size of the flows seemed to decrease

TABLE 2. The Dimensionless Vortex Size of a Few Vortex Boundary Layers (Layers 2nd–4th) for NR Compounds at Die Entrance of Circular and Slit Flow Channels of Dual Die for Different Piston Displacements

Piston or extrusion displacement (mm)	Dimensionless size of vortices					
	Circular channel			Slit channel		
	Layer#2	Layer#3	Layer#4	Layer#2	Layer#3	Layer#4
0	0.7	1.0	1.2	1.2	1.7	2.2
40	0.7	0.9	1.1	1.2	1.7	2.1
70	0.6	0.6	0.7	1.0	1.3	1.4
100	0.5	0.6	0.6	0.9	1.0	1.2

with increasing piston displacement, and this would have resulted in a decrease in extrudate swell. The extrudate swell results as a function of piston displacement are not given in this article, but previous work by Sombatsompop and Dangtangee [5] clearly indicated that, for a constant extrusion speed, the extrudate swell decreased as a function of piston displacement. The reasons for this were discussed in connection with the melt temperature rise and the complexity of the flows developed in the rheometer.

CONCLUSION

Studies on extrudate swell and flow properties of PS and LLDPE melts in single and dual channel dies in a constant shear rate rheometer were carried out. The results suggested that the extrudate swell ratio for PS melt was greater than that for LLDPE melt for any given shear rate due to the differences in power law indexes and secondary flows at the die entrance. In the single channel die, the extrudate swell of both PS and LLDPE melts in the circular flow channel die were greater than that in the slit flow channel; whereas, in the dual channel die the slit flow channel exhibited a higher extrudate swell ratio than the circular channel. The higher extrudate swell in the slit channel in the dual die was associated with two possible factors, one being the higher degree of disentanglements of the polymer molecules flowing into the slit channel, which was influenced by the actual size and geometry of the slit, and the other being the dimensionless size of the vortex flows observed near the die entrance, which was related to the molecular orientation of the extrudate.

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PAT

Die swell ratio of polystyrene melt from an electro-magnetized capillary die in an extrusion rheometer: effects of barrel diameter, shear rate and die temperature

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A constant shear-rate extrusion rheometer with an electro-magnetized capillary die was utilized to investigate die swell behavior and flow properties of a polystyrene melt as the application of an electro-magnetic field to the capillary die was relatively novel in polymer processing. The test conditions such as magnetic flux density, barrel diameter, extrusion rate and die temperature were studied. The results suggest that the maximum swelling of the polystyrene melt with application of the electro-magnetic field could be enhanced up to 2.6 times (260%) whereas that without the electro-magnetic field was 1.9 times (190%). The barrel diameter of 30 mm was found to be a critical value in the case of the die swell ratio and flow properties of the polystyrene melt were significantly affected by the magnetic flux density. This involved the number and angle of magnetic flux lines around the barrel part. Under the electro-magnetic field, there were two mechanical forces influencing the die swell ratio and the flow properties; magnetic torque and shearing force. The die swell at wall shear rates less than 11.2 sec^{-1} was caused by the magnetic torque, whereas at higher wall shear rates it was dependent on the shearing force. For a given magnetic flux density, the maximum increase in the die swell ratio as a result of the magnetic torque was calculated to be approximately 20%. Increasing the die temperature from 180 to 200°C reduced the overall die swell ratio and suppressed the effect of the magnetic flux density.

KEYWORDS: swelling; extrusion; processing; rheology; melt

INTRODUCTION

It is widely accepted that the design and processing of polymeric products depend heavily on the properties of the molten material flowing under strictly controlled conditions. Such a polymer has the combined properties of an elastic solid and a viscous fluid. The viscous nature is dominant during the processing, while the elastic character is still present to cause product phenomena such as die swell, sharkskin, and melt fracture.[1] In an extrusion process, die swell, or Barus effect, is an important parameter for determining the size and the quality of the extruded-polymer product, and it is also useful for assessing the polymer elasticity in extrusion.

Effects of test variables on the changes in die swell of polymer melts have been widely examined; for example, shear rate, die temperature, type and concentration of additives, material molecular weight, die geometry (size, shape and entry angles), and shearing and thermal histories.[2–13] The die swell has been explained in terms of molecular mechanisms of entanglement, orientation and recoil, even though no direct evidence is available for these in the molecular scale. Work in this field has been extensively developed comparing experimental data with different numerical models. Swan *et al.*[2] investigated the effects of die temperature, flow rate, molecular weight distribution on the thickness and diameter swells as a function of melting time using an annular die, and found that the swell was slightly affected by changes in melt temperature and flow rate, and strongly dependent on molecular weight distribution. Lee and Ho[3] determined the die swell behavior of a polymer melt by designing a die for profile extrusion applications, and found that the melt temperature, die length, and residence time affected the die swell. The die swell ratio became smaller as the melt temperature, melt residence time and die length increased. Kar and Otaigbe[4] experimentally measured the die swell ratio of various thermoplastic melts in an extruder over a wide range of shear rates and melt temperatures, and the results were then compared with various existing numerical models. They

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suggested that the die swell was closely related to the first normal stresses and varied strongly with shear stress and shear rate, but was independent of melt temperature, although the experimental data were not confirmed by the models. Dufrancatel-Veiller *et al.*[5] observed that the die swell of thermoplastic polyurethane was greatly influenced by material residence time and the changes in molecular structure evolution during processing.

Additives loaded into polymer melt also influence the degree of swelling of polymer melts. Shin *et al.*[6] determined the extrudate swell of the SBR[Q1] using different carbon black grades and die length/diameter[Q2] ratios. They found that the extrudate swell reduced as the carbon black loading and the length/diameter ratio of the die increased. Abraham *et al.*[7] observed significant changes in die swell and viscosity of a low-density polyethylene when added with a small amount of dicumyl peroxide. Sombatsompop *et al.*[8] measured the die swell of poly(vinyl chloride) (PVC) compound added to natural wood fibers and suggested that the die swell decreased with wood content due to a decrease in elasticity of the PVC. The die swell could also be indicative of interaction between the wood fibers and the PVC molecules.

It has been observed that the die swell of a polymer melt also depends on the design of the die used. Kiriakidis and Mitsoulis[9] performed studies on extrudate swell of a high-density polyethylene melt using slit and capillary dies with respect to the effect of die-length/diameter using the finite element method. They found that for any given shear rate, the swelling was greater with the capillary die than the slit one, and the extrudate swell decreased drastically as the die-length/diameter ratio increased. Sombatsompop and Dantangee[10] experimentally measured the elastic swell of natural rubber (NR) extrudate in a capillary rheometer using two dies located along the barrel, and found that the die swell of the rubber depended on the residence time of flowing in the barrel. The work was extended to study the effect of actual barrel/die diameter ratios on the die swell of polymer melts in the rheometer,[11] and it was found that the change in extrudate swell was linearly influenced by the entrance pressure drop at low actual barrel/die diameters (D_B/D_D from 20/4 to 30/7 mm/mm), but was associated with the change in material viscosity at high actual barrel/die diameters (D_B/D_D from 35/7 to 40/8 mm/mm). At a constant barrel diameter, the smaller the die diameter the greater the extrudate swell due to increases in the extensional deformation, and shear rate, and a reduction in the melt residence time. They also designed and manufactured two flow channel dies with different geometries, one being circular and the other being slit channels in order to use in the rheometer, and the extrudate swell behavior of thermoplastic melts was investigated.[12] The results indicated that at a given shear rate, the slit channel exhibited higher die swell, this being opposite to the results reported by Kiriakidis and Mitsoulis.[9] Gullet and Seriai[13] stated that the swelling phenomenon is mainly due to the recoverable elongational strain induced by the converging flow at the die entrance, as well as by recoverable shear strain originating within the die. They also offered an equation for the quantitative prediction of extrudate swell of polystyrene (PS) as well as linear polyethylene melts from the elastic material properties such as the entrance pressure drop as a result of different contraction ratios, the relaxation modulus and the recoverable shear strain.[14]

Apart from the earlier published papers, Cao and Li[15] offered a different parameter to alter the swelling ratio of a polypropylene melt by applying ultrasonic irradiation during extrusion. The results revealed that the use of appropriate irradiation intensity could reduce the die entrance pressure and swelling ratio of the melt at the die exit as well as improve the apparent quality of the extrudate. Q1
More recent work by Sombatsompop[16] studied the effect of a Q2
magnetic field on the die swell ratio of PS melt in a capillary die of a constant shear-rate rheometer, and the unexpected results suggested that application of a magnetic field to the die could result in a significant increase in the swelling ratio of the PS extrudate of up to 25% as compared with the results of other polymer melts measured without the magnetic field.

There appear to be no other researchers than those of references 15 and 16 who have applied an electro-magnetic field to the polymer testing and processing machinery and studied the flow behavior of the melts. The present work is aimed to extend previous work[16] by applying an electro-magnetic field, whose magnetic flux density could be varied, on the changes in the die swell of PS melt in a capillary rheometer. The flow properties of the PS melt in the rheometer were also determined in terms of the relationship between shear stress and shear rate, where the tests were performed under the test conditions at which the die swell ratios were carried out. The effects of barrel diameter, wall shear rate and die temperature on the die swell were mainly considered in this work. The Finite Element Method Magnetics (FEMM) software (V3.3) was used to simulate the flow of the magnetic flux densities around the rheometer (barrel and die components) under various test conditions.

EXPERIMENTAL

Raw material

All tests in this work used PS (Styron 656D 267), in granular form supplied by Siam Polystyrene Co., Ltd (Bangkok, Thailand). It was characterized by a melt flow index of 7 and a density of 1.350 g/cm^3 (BS2782 Method 720A; 1979).

Rheometer and an electro-magnetized die

A constant shear rate rheometer was employed in this work as used in previous works.[10–12] The barrels with different diameters (ranging from 25 to 40 mm) and 150 mm long were used. The capillary die used in this work was made of mild steel (Grade 1020), 60 mm long and 6 mm in diameter. To make an electro-magnetized die, the die body was wrapped with copper wires in order to generate or apply the electro-magnetic field to the die. To prevent an electrical short-circuit, Teflon film was used between the outer die wall and the copper solenoid coil. The experimental apparatus is shown in Fig. 1. The electro-magnetic field was generated to the die by applying electricity to the copper solenoid coil, the electricity varying from 2 to 10 A. The dimension and arrangement of the electro-magnetized die can be seen in Fig. 2. It was of some concern that the application of the electro-magnetic field would result in additional heat due to the

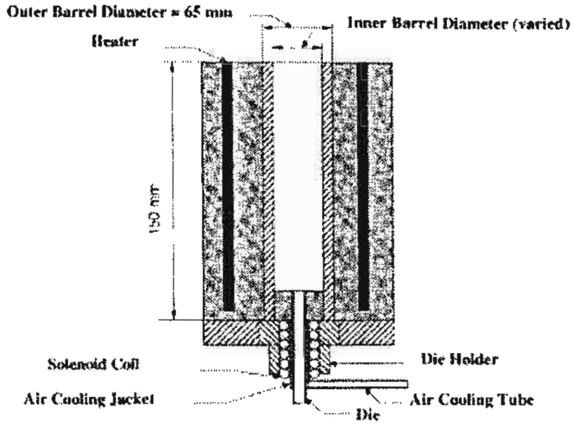

Figure 1. Experimental rheometer arrangement for die swell measurement of PS melt using an electro-magnetized die system.

induction effect across the solenoid coil. This additional heat would then affect the die and melt temperatures during the tests. Therefore, an air-cooling jacket unit was constructed and placed around the electro-magnetized die (see Fig. 1), the cooling unit being located between the solenoid coil and the die body. A flow-rate of the cooling air was adjusted and calibrated to reduce the additional heat so that the required temperature was achieved—there will be some experimental results in the discussion section confirming that no additional heat occurred during the test. A small pressure hole was located between the two die locations to detect die entrance pressure drop by using a photo-conductive light pressure sensor,[17] which was designed and manufactured in our research group. The apparatus temperature was controlled using a DD6 temperature controller.

Measurements of die swell ratio and flow properties

Die swell ratio

The elastic swell ratio (B) of the polymer extrudate was determined by calculating the ratio of diameters of the extrudate to the die. The extrudate diameter was dependent on its size

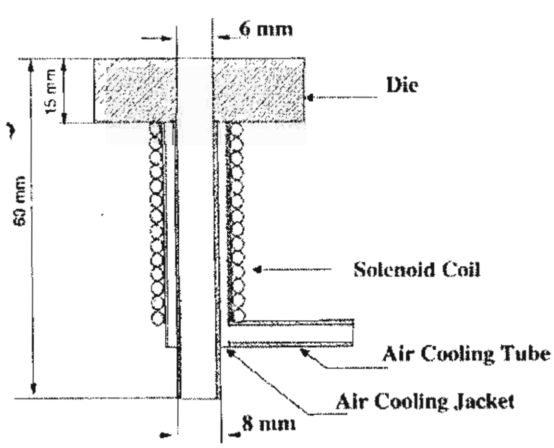

Figure 2. The dimensions and configuration of the electro-magnetized die.

when fully swollen, this being about 1–2 inch away from the die exit. A video-camera (SSC-DC398P with 752Hx582V pixels and 480TV lines) and a high resolution macro zoom lens (3.3× magnification) were used to visualize the extrudate leaving the die exit whose results were recorded and displayed in real time using a personal computer. The size of the extrudate was then measured by replaying the recorded flow. With this procedure, any error related to the cooling and gravity effects could be minimized.

Flow curves

Flow curves (relationship of wall shear stress to wall shear rate) of the PS melt were simultaneously determined under the conditions at which the extrudate swell was measured, whose calculations can be obtained elsewhere.[12] It should be noted that Bagley's corrections were not applied to the shear stress data generated in this work, as the shear stress data is solely being used for comparative reasons to illustrate the magnitude of the changes observed in the flow characteristics of the materials as a function of the electro-magnetic intensities applied.

Analysis of the flow of magnetic intensities

FEMM software (V3.3; 2003; Germany) was used to computer-simulate distribution patterns of the magnetic flux density at/around the barrel and die sections. The input data included the number of magnetic flux lines, relative permeability of all materials used, and source current density. For comparison, the number of magnetic flux lines was fixed at 19. The relative permeabilities of air, mild steel and copper materials used were 1, 760, and 0.99 H/m, respectively. The source current densities at 2, 4, 6, 8 and 10 A were 2.2, 4.4, 6.6, 8.8, 11 MA/m^2, respectively.

Experimental variables

In this work, the die swell ratios and the flow curves of PS melt through use of the electro-magnetized die were determined with different test parameters as listed later, compared with those produced without application of the electro-magnetic field:

- *Magnetic flux density*: It has been described earlier that the electro-magnetized die was generated by supplying electricity to the copper coil wrapped around the die body and air-cooling unit. The magnetic flux density of the electro-magnetic field was altered by varying the amount of electric current from 2, 4, 6, 8 to 10 A, which corresponded to the magnetic flux densities of 1.51, 1.74, 1.92, 2.04 and 2.11 T, respectively. These values were measured at the centre position of the copper coil which appeared to give the maximum value for a given electric current. It was understood that the magnetic flux density generated varied from location to location on the experimental rig (barrel and die bodies). Therefore, the die swell results obtained in this work were reported as a function of the maximum magnetic flux density in the capillary die (at the centre position of the copper coil). However, the density distribution of the magnetic flux was also computer-simulated in order to explain the changes in the die swell ratio of the melt when altering the magnetic flux densities.

- *Barrel diameter or melt contraction ratio*: This was referred to as a ratio of barrel diameter to die diameter. The ratio was determined at various barrel diameters ranging from 25, 30, 35 to 40 mm, whereas the die diameter was fixed at 6 mm throughout this work. The barrel diameter was referred to as the inner part of the barrel where the outer diameter of the barrel was fixed at 65 mm (see Fig. 1). In this case, the temperature of the die was fixed at 180°C.
- *Wall shear rate*: The piston speed in the constructed rheometer was varied by trial error experiments, from 5 to 100 mm/min, not to reach the critical shear stresses for the onset of PS melt distortions (sharkskin and melt fracture) under the test temperatures between 180–200°C. Accordingly, the die swell measurements were accurately done. The corresponding shear rates calculated from the piston speeds were dependent on the size of the barrel used.
- *Die temperature*: A barrel diameter was selected to investigate the effect of die temperature on the die swell of PS melt in the rheometer at various temperatures of the electro-magnetized die varying from 180 to 200°C.

RESULTS AND DISCUSSION

Changes in overall die swell and flow properties

Figure 3(a)–3(d) show the die swell ratio of PS melt as a function of wall shear rate in the die, which had various magnetic flux densities from 0 to 2.11 T, with different barrel diameters (D_B) at a fixed die diameter (D_D) of 6 mm and a die temperature of 180°C. In general, it can be observed that the die swell increases with increasing shear rate for all cases as one would expect.[1] Concerning the swelling ratio of the extrudate from the die without magnetic field, the average value of the die swell ratio for PS melt was in the range of 1.5–1.9, regardless of the barrel size used, and the overall die swell appeared to increase with increasing the barrel diameters from 25 to 40 mm. The increase in die swell ratio due to increasing the contraction ratio (increasing D_B/D_D ratio) was associated with the increase in the entrance pressure drop which can be seen by the flow curve results in Fig. 4(a) and 4(b), illustrating the effect of barrel diameter and magnetic flux densities on the flow curve of the PS melt at a die temperature of 180°C. The increase in the pressure drop was due to the increased shear rate and also enhanced extensional deformation and elastic energy stored in the melt as the contraction ratio increased.[1,4,10]

In this work, the effects of barrel diameter, magnetic flux density and wall shear rate were studied. It was very interesting to observe in Fig. 3(a)–3(d) that the die swell ratio for any given wall shear rate did not change very much with magnetic flux density for all barrel diameters except for the barrel diameter of 30 mm (Fig. 3b). Similar behavior was also found for the melt properties shown in Fig. 4(a)–4(d).

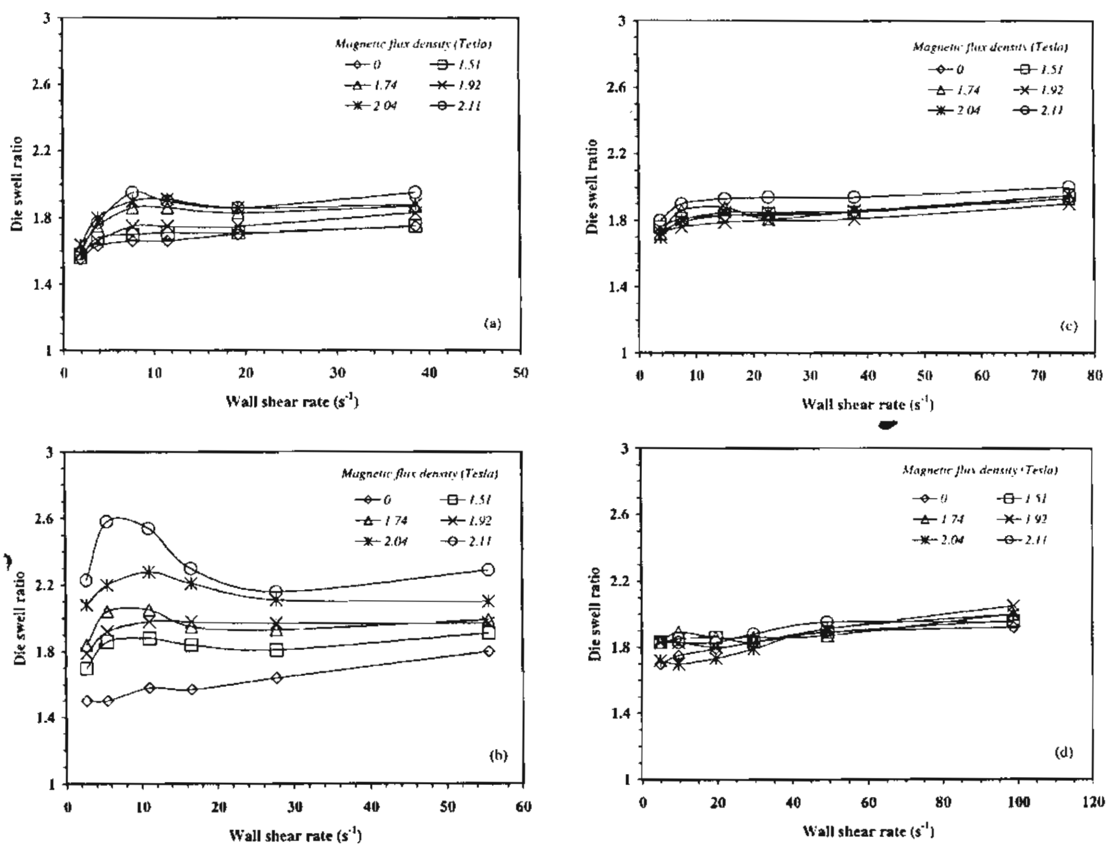

Figure 3. Die swell ratio of PS melt as a function of wall shear rate at various magnetic flux densities using a 6 mm die diameter at 180°C: (a) 25 mm barrel diameter; (b) 30 mm barrel diameter; (c) 35 mm barrel diameter; (d) 40 mm barrel diameter.

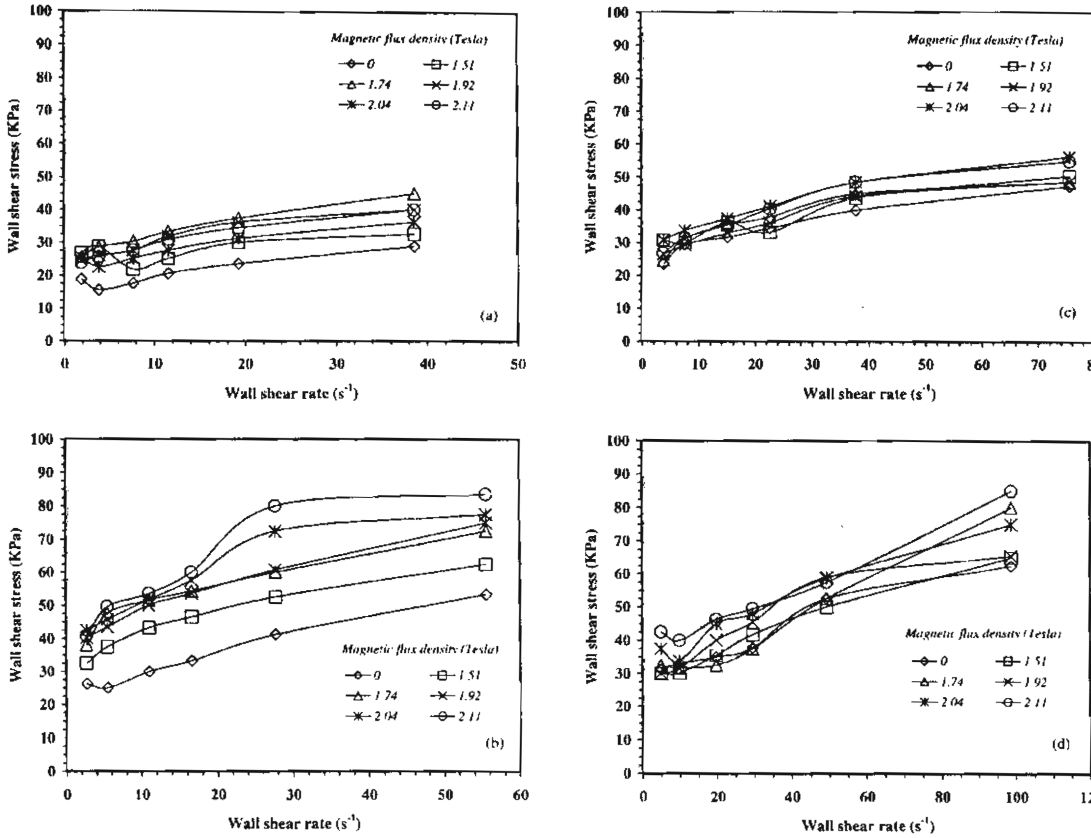

Figure 4. Effect of barrel diameter and magnetic flux densities on the flow properties for PS melt at a die temperature of 180°C: (a) 25 mm barrel diameter; (b) 30 mm barrel diameter; (c) 35 mm barrel diameter; (d) 40 mm barrel diameter.

This result implied that there was a critical barrel size at which the magnetic flux density affected the melt property in the rheometer. The changes in die swell ratio with wall shear rate and magnetic flux density at 30 mm barrel diameter were significant whereas those at the other barrel diameter sizes were very small.

Mechanism of melt swelling by magnetic forces

The results in Figs. 3(b) and 4(b) (barrel diameter of 30 mm) clearly showed that the die swell ratio and flow properties increased with increasing magnetic flux density. The effect was more pronounced at relatively low wall shear rate, where the die swell ratio increased from about 1.6 to 2.6 with increasing the magnetic densities from 0 to 2.11 T. Figure 5(a) and 5(b) show the extrudates leaving from the die exit without application of the electro-magnetic field, and with 2.11 T magnetic flux density, respectively. The extrudate from the electro-magnetized die had a much greater swelling ratio than that from the normal die. The increase in die swell ratio with the electric current to the copper solenoid coil indicates that no additional heat generation occurred from the induction effect of the solenoid coil. If heat was generated, one would expect to observe a decrease in the die swell ratio of the melt, since it is known that an increase in die or melt temperature results in a reduction of extrudate swell.[1,10,13] Increasing the magnetic flux density led to an increase in the magnetic torque acting on the PS melt flowing in the barrel and die. This would suggest that some molecular configuration of PS may change under the magnetic torque due to electron delocalizations and distortions of electron clouds within the benzene rings. This then resulted in the changes in the molecular orientation or/and alignment of the melt during the flow of the melt in the barrel and also in the die. This explanation was supported by Amundson[18] and Chandrasekhar.[19] However, this finding seemed to contradict the results given by Kimura[20] who stated that only polymers that had anisotropic structure would be able to align under the magnetic field. In this work, although the PS used in this work was isotropic when it remains in a non-flowing state it could be anisotropic as it was flowing along the die. This was because when a polymer melt flows from a large reservoir (barrel) to a small capillary die the molecular chains of the melt have to re-arrange themselves in the flow direction. This molecular transition then caused the melt to be more anisotropic. Under the magnetic field, the alignment of the polymer chains was more and thus increased the die swell ratio at the die exit. The changes in the molecular alignment were indicated by the changes in the wall shear stresses and thus the degree of swelling of the melt at the die exit. This behavior becomes more pronounced with higher magnetic torque by increasing the magnetic flux density.

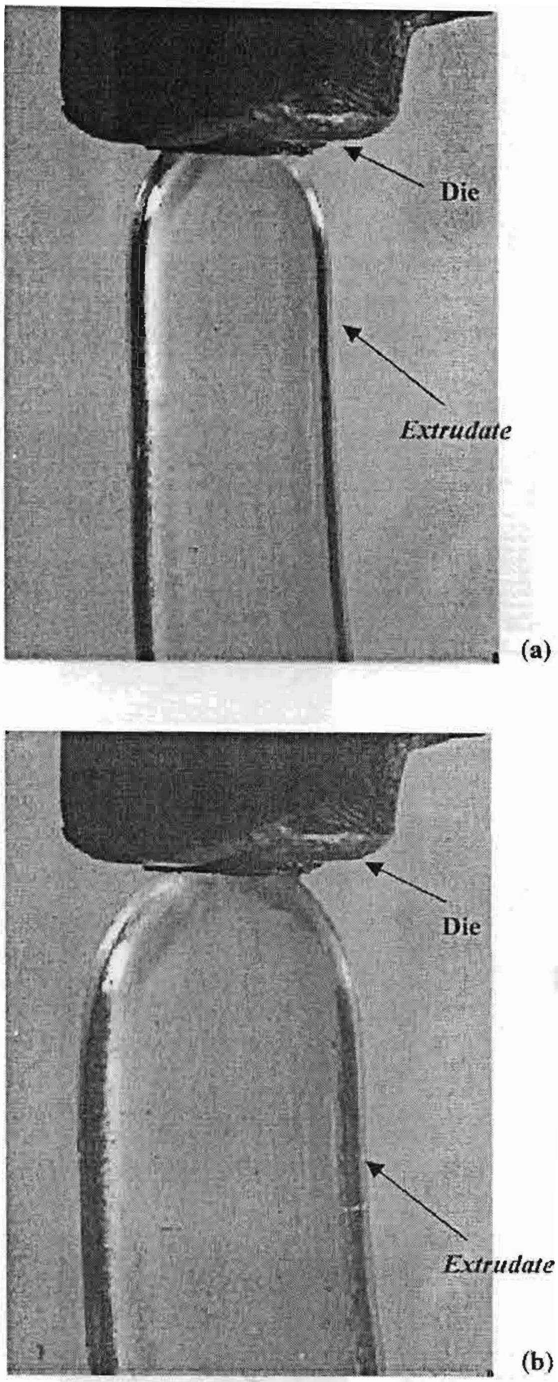

Figure 5. An example of PS extrudates while leaving the dies: (a) without electromagnetic field; (b) with 2.11 T magnetic flux density.

Effect of barrel diameter

At first, the changes in the die swell and melt properties were expected to be caused by the differences in magnetic flux densities in the die, so that the magnetic flux densities at different positions; die entrance, centre of the die length, and die exit along the die length for all barrel diameters used were measured. The results are shown in Fig. 6(a)–6(c). It can be seen that for any given measured position, the magnetic flux density increased with increasing electric current, and it was in a linear fashion at the die entrance. The magnetic flux density was relatively high around the centre of the die length (Fig. 6b); the maximum values being around 2.11 T (at 10 A electric current). For a given electric current and measured position along the die, the magnetic flux densities were indifferent for all barrel diameters used. This was reasonably possible since the die dimension was fixed. Therefore, the results suggest that the changes in the die swell ratio and melt properties due to the changes in barrel diameter are associated with the magnetic flux distributions in the barrel part. Figure 7(a)–7(d) show distributions of the magnetic flux lines

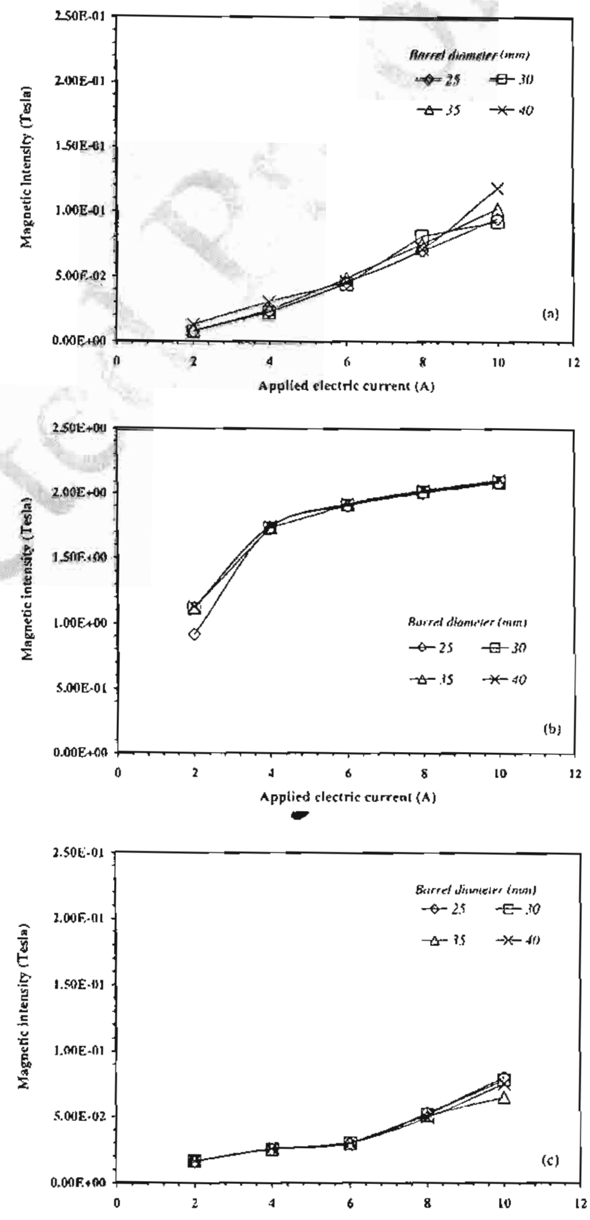

Figure 6. Magnetic flux densities at different positions along the die length for different barrel diameters at 180°C: (a) at die entry region; (b) at centre of die length; (c) at die exit region.

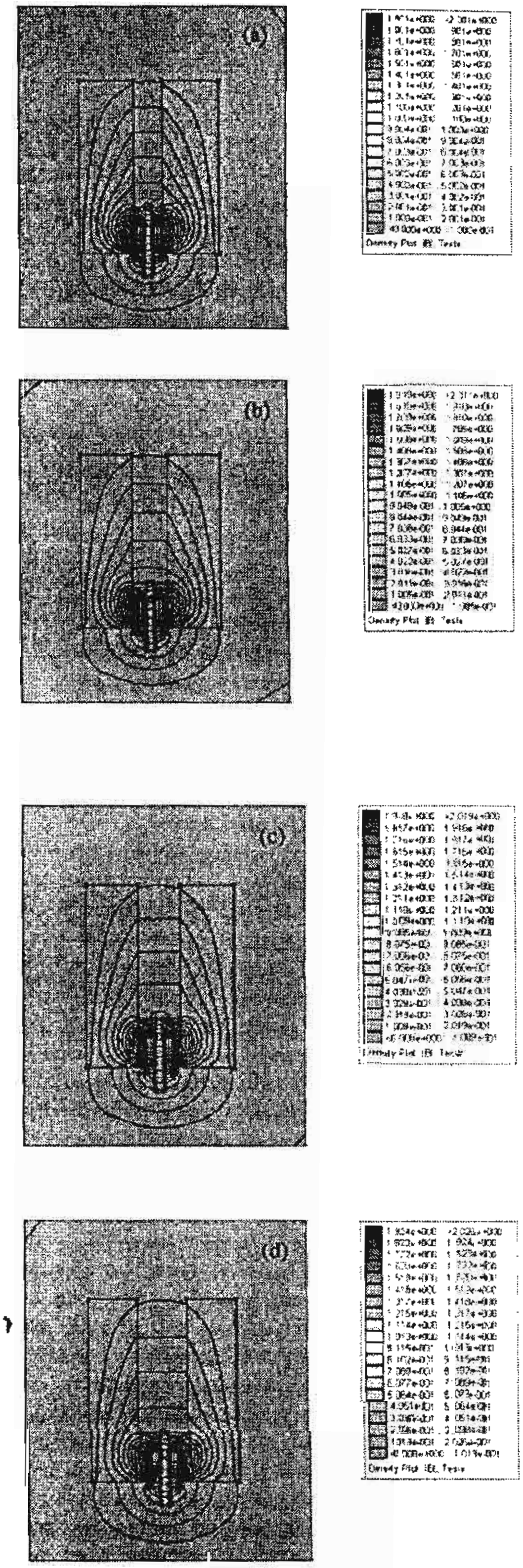

Figure 7. Q3Distributions of the magnetic flux lines for different barrel diameters using FEMM software and magnetic flux density of 2.04 T: (a) 25 mm barrel diameter; (b) 30 mm barrel diameter; (c) 35 mm barrel diameter; (d) 40 mm barrel diameter.

for all barrel diameters with a selected electric current of 8 A (at 2.04 T magnetic flux density) using the FEMM software. The magnetic flux density patterns for all barrel diameters were very similar, and the flux density in the barrel component was less than 0.2 T. However, the number of magnetic flux lines for each barrel diameter was different depending on the barrel part. The numbers of magnetic flux lines decreased when increasing the barrel diameter from 30 to 35 mm. The number of magnetic flux lines for barrel diameters of 25 and 30 mm was five whereas those for barrel diameters of 35 and 40 mm were four. This was expected since the barrel diameters of 25 and 30 mm had more steel parts than those of 35 and 40 mm; the outer diameter of the barrel part was fixed at 65 mm and the inner diameter of the barrel was varied as mentioned in the Experimental section. The results in Fig. 7(a)–7(d) were consistent with those in Fig. 3(a)–3(d) and Fig. 4(a)–4(d). It can be said from these results that the greater the number of magnetic flux lines, the stronger the magnetic torque that could interact with the contained material (PS melt) in the barrel, as the molecular configuration of the melt then changes while flowing from the barrel to the capillary die. This was why the barrel diameter of 30 mm exhibited a greater swelling ratio than those of 35 and 40 mm. It was however found that the 25 mm barrel diameter gave a lower die swell ratio than the 35 and 40 mm barrel diameters, because of the less barrel-to-die contraction ratio.[10] Comparing the die swell ratios of the PS melt with the barrel diameters of 25 and 30 mm, it was found that the die swell ratio of PS melt for 30 mm barrel diameter was much greater than that for 25 mm. A possible cause for this would be the degree of magnetic torque generated in the barrel part which is empirically associated with the direction and angle of the occurring magnetic flux line, determined by eq. (1). It should be noted that eq. (1) can be used solely for qualitatively explaining the changes in the die swell ratio for different barrel sizes, absolute values of magnetic forces (then magnetic torque) cannot be obtained by eq. (1).[21]

$$F = Qv \times B \qquad (1)$$

where F and Q represent magnetic force on a charged particle (N) and charged particle (C), respectively, v is charged velocity (m/s) and B is magnetic flux density (T).

Equation (1) indicates that a magnetic force occurs in a perpendicular direction to its magnetic flux density. Figure 8 shows schematic diagrams of the magnetic flux line (B) and magnetic force (F) acting on the polymer melt flowing in the barrels having diameters of 25 mm (Fig. 8a) and 30 mm (Fig. 8b). The proposed diagram is part of the magnetic flux lines in Fig. 7. It can be seen that the slope of the magnetic flux line for the 30 mm barrel diameter is greater than that for the 25 mm barrel diameter. This implies that the magnetic force acting on the melt was greater in the 30 mm barrel diameter than in the 25 mm barrel diameter, and thus increased the die swell ratio of the melt due to the effect of the magnetic torque on the die swell and flow properties.

Q3

Wall shear rate effect

A relationship between the wall shear rate and the die swell ratio was found to be non-linear. It was observed at the barrel

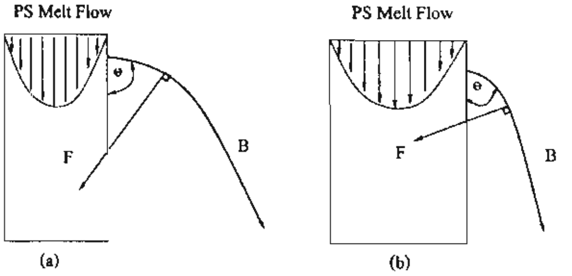

Figure 8. Schematic diagrams of magnetic flux line (B) and magnetic force (F) acting on the polymer melt flowing in the barrels: (a) 25 mm barrel diameter; (b) 30 mm barrel diameter.

diameter of 30 mm when the electro-magnetic field had a pronounced effect on the die swell ratio of the PS melt as shown in Fig. 3(b). It was also found that the die swell ratio increased at shear rates between 5.5 to 11.2 sec^{-1} and then decreased to level off at shear rates greater than 11.2 sec^{-1}. This behavior was observed for all magnetic flux densities; the higher the magnetic flux density, the more pronounced the change of the die swell ratio. The die swell ratio changes are considered to be caused by two applied mechanical forces; one being a shearing force due to the extrusion rate and the other being a magnetic torque due to the applied electric current. At low wall shear rates (lower than 11.2 sec^{-1}), the considerably increased die swell ratio was obtained by the magnetic torque. This was because the slow extrusion rates allowed the magnetic torque to act for a longer time on the melt during the flow in the barrel and the die. When the melt was forced to flow at higher extrusion rates (greater than 11.2 sec^{-1}), it would have less time to be induced by the magnetic torque, so that the increase in the die swell ratio was solely caused by the shearing force as a normal effect. Therefore, the overall die swell ratio decreased at shear rates greater than 11.2 sec^{-1}. For a given magnetic flux density, the maximum increase in the die swell ratio by the magnetic torque was calculated to be about 20% (for the barrel diameter of 30 mm).

Effect of die temperature

Figure 9(a)–9(d) show the effect of die temperature on the die swell ratio for PS melt using the barrel diameter of 30 mm. It can be seen that for any given wall shear rate and magnetic flux density, the die swell ratio decreased with increasing die temperatures from 180 to 200°C. Increasing the die temperature automatically increases the melt temperature to result in an increase in the viscous characteristic of the melt and thus a decrease in the elastic recovery and swelling ratio at the die exit. For the other barrel diameters, a similar effect of die temperature on the die swell ratio was obtained. Another interesting point to consider is that the swelling peak at wall shear rates around 11.2 sec^{-1} became less pronounced when increasing the die temperature. This suggests that the temperature suppresses the magnetic torque acting on the molecular structure of the polymer melt. It is actually known that the magnetization of a material is decreased by temperature.[22]

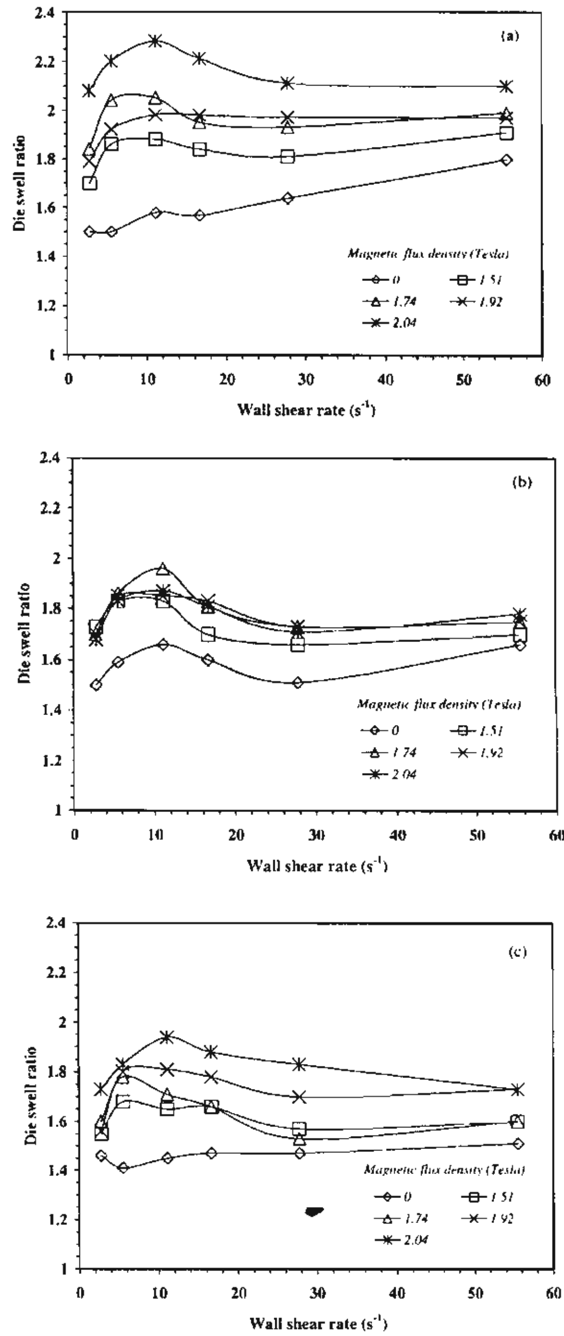

Figure 9. Effect of die temperature on die swell ratio for PS melt using the barrel diameter of 30 mm: (a) 180°C; (b) 190°C; (c) 200°C.

SUMMARY

The die swell behavior of PS melt in a constant shear rate extrusion rheometer with application of electro-magnetic field to the capillary die was studied to investigate the effects of barrel diameter, wall shear rate and die temperature. The following findings were made in this study.

(1) The maximum swelling of the PS melt was up to 260% with application of electro-magnetic field whereas without the electro-magnetic field, it was 190%.
(2) In normal capillary die, the die swell ratio of PS melt increased with increasing barrel diameter (or contraction ratio), wall shear rate, but decreased with increasing die temperature.
(3) The barrel diameter of 30 mm was found to be a critical size at which the die swell ratio and flow properties of the PS melt considerably changed by varying the magnetic flux density, the changes in the die swell and flow properties being associated with the number and angles of magnetic flux lines in the barrel part.
(4) An increase in the die swell at wall shear rates less than $11.2\ \mathrm{sec}^{-1}$ was caused by the magnetic torque, whereas at higher wall shear rates the die swell ratio was very much dependent on the shearing force. This phenomenon was postulated to be associated with the residence time for the melt in the barrel, under applied magnetic torque. For a given magnetic flux density, the maximum increase in the die swell ratio was calculated to be about 20%.

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A Parallel Co-extrusion Technique for Simultaneous Measurements of Radial Die Swell and Velocity Profiles of a Polymer Melt in a Capillary Rheometer

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ABSTRACT

This article proposes a new experimental technique to *simultaneously* measure radial die swell and velocity profiles of polystyrene melt flowing in the capillary die of a constant shear rate rheometer. The proposed technique was based on parallel co-extrusion of colored melt-layers into uncolored melt-stream from the barrel into and out of the capillary die. The size (thickness) ratio of the generated melt layers flowing in and out of the die was monitored to produce the extrudate swell ratio for any given radial position across the die diameter. The radial velocity profiles of the melt were measured by introducing relatively light and small particles into the melt layers, and the times taken for the particles to travel for a given distance were measured. The proposed experimental technique was found to be both very simple and useful for the simultaneous and accurate measurement of radial die swell and velocity profiles of highly viscous fluids in an extrusion process. The variations in radial die swell profiles were explained in terms of changes in melt velocity, shear rate, and residence time at radial positions across the die. The radial die swell and velocity profiles for PS melt determined experimentally in this work were accurate to 92.2% and 90.8%, respectively. The overall die swell ratio of the melt ranged from 1.25 to 1.38. The overall die swell ratio was found to increase with increasing piston speed (shear rate). The radial extrudate swell profiles could not be reasoned by the shear rate change, but were closely linked with the development of the velocity profiles of the melt in the die. The die swell ratio was high at the center (~1.9) and low (~0.9) near the die wall. The die swell ratio at the center of the die reduced slightly as the piston speed was increased.

Key words: Die swell, Velocity profiles, Co-extrusion, Capillary rheometer, Polymer melt.

INTRODUCTION

The properties of a polymer melt are a combination of the properties of an elastic solid and a viscous fluid. During processing the viscous nature is the dominant characteristic. However, the elastic character is still present. When polymer melts are subject to a constant stress they can store strain energy. On removal of the stress, the elastic component of the strain can be recovered whereas the viscous component cannot. This behavior is well known as the Maxwell spring behavior of linear viscoelasticity (1). When a polymer melt is extruded from a reservoir through a die the cross-sectional area of the extrudate is greater than that of the die, this being referred to as *"die swell"*. The general explanation for die swell is related to the recoverable elastic deformation developed during flow through the die. Die swell is considered one of the main factors to determine quality and dimension of the polymer product. Understanding these phenomena enables engineers to design polymer processing components and equipment such as screws, dies and sizing units, and optimize the processing conditions. The die swell theories are not always accurate, although many researchers (2-6) have claimed the validity of their theoretical approaches for precisely predicting the swelling behavior of the melts, but are only in qualitative agreement with experimental data. Usually, die swell increases as: the die length (residence time) decreases, the shear rate or shear stress increase, and the molar mass of the polymer increases. On the contrary, die swell can be minimized by increasing die temperature and die land length or reducing the shear rate or shear stress.

Experimental methods for measuring the die swell ratio of polymer melts have involved many difficulties such as draw-down effects, molecular relaxation at the die exit, contraction due to cooling, and crystallization (1). Previous studies in the field have been directed towards both experimental and theoretical investigations, but mostly by computer simulations. For experimental studies, various techniques have been used, including a direction measurement, extrudate weighing method, video camera equipment, and laser techniques (7-9). Some modifications, to minimize the effects of crystallization, draw-down and contraction to cooling, have been applied, including the use of an oil bath having the same density as the fluid, and the change of extrusion direction. Song *et al* (7) conducted a flow marker technique to study the flow patterns of various rubber compounds, including NR, SBR and EPDM in the barrel of both slit and capillary rheometers. They found that the flow simply moved radially inward to the capillary die as the ram moved down the barrel without secondary flows occurring. They also used laser equipment attached to a Monsanto Processability Tester to measure the swelling ratio of the rubber extrudates. Sawn *et al* (8) developed an experimental technique to simultaneously measure the thickness and diameter swell ratios of a melt parison by using moving video cameras, and investigated the effects of die temperature, flow-rate, and polymer molecular weight. Gullet and Seriai (2) stated that the swelling phenomenon is mainly due to the recoverable elongational strain induced by the converging flow at the die entrance, as well as by recoverable shear strain originating within the die. They also offered an equation for the quantitative prediction of extrudate swell of a polystyrene melt from the elastic material properties such as the entrance pressure drop, the relaxation modulus and the recoverable shear strain. Later work by Serisi *et al*, (3) offered another simple analytical expression to predict extrudate swell of polystyrene at a wide range of residence times. Lee and Ho (4) determined the die swell behavior of a polymer melt for designing a die for profile extrusion applications. They found that the melt temperature, die length, and residence time affected die swell. The die swell ratio became smaller as the melt temperature, melt residence time and die length were increased. Similar behavior of die swell affected by wall shear rate, melt temperature and L/D ratio was also observed by Kar and Otaigbe (5) who examined such behavior in various thermoplastic melts in a Randcastle microtruder for a wide range of shear rates, L/D ratios, and melt temperature. They also noted a linear relationship between die swell and maximum recoverable deformation, and a non-linear relationship between die swell, storage and loss moduli. Wilczynski *et al* (6) presented a method for modeling the free multi-layer forming problem in the co-extrusion process. They also suggested that differences in the viscosity of flow material resulted in displacement of the polymer stream towards the more viscous phase and displacement of the maximum of the velocity profile to the opposite direction. Instead the differences in flow rate led to transferring of the interface towards the phase of the lower flow rate, referred to as swelling behavior. Sombatsompop *et al* (9-11) carried out detailed investigations on the die swell behavior of polystyrene (PS) and linear low-density polyethylene (LLDPE) melts influenced by different die designs such as actual die size, contraction ratio and the effect of a magnetic field on the swell ratio. Their latest work (12) involves use of a dual channel die, having mixed circular/slit flow channels, to observe the die swell behavior of PS melt in a capillary rheometer. In the single channel die, the extrudate swell of PS melt in a circular flow channel die was greater than that in the slit flow channel, whereas, in the dual channel die the slit channel exhibited a higher extrudate swell ratio.

Knowledge of flow patterns and velocity profiles in the die is necessary in analyzing the flow behavior of polymer melts. Many researchers (13-18) have utilized a number of methods or techniques to measure the velocity profiles and flow patterns of flowing polymer melts, the accuracy of the measurements varying with the ways the velocity profiles are measured. Most investigations have been carried out in the die entry region of both slit and capillary rheometers (19-20). The existing methods and techniques used include pigmented banding (14), volume change (15), dye and/or particle injections (16), and Laser Doppler Velocimetry (LDV) (17-18, 20). This latest technique is widely used as being relatively accurate, but the apparatus is expensive and complicated. Sombatsompop *et al* (14-15) measured the velocity profiles of natural rubber compounds flowing in the barrel and the die of a capillary rheometer. They found that the velocity profiles of the melt in the barrel changed continuously with piston displacement while those in the die were not parabolic in nature. In addition, non-zero melt velocity at the die wall was observed. Meissner (17) and Yarusso (18) utilised laser Doppler velocimetry to study changes in the velocity profiles near the entrance and exit regions of a slit die. They found that the velocity profiles for shear thinning fluids changed from a parabolic form in the capillary to a plug-like form at the die exit, and the degree of extrudate swell was influenced by this velocity rearrangement. Recently, Sombatsompop and Wood (21) developed a novel technique, the so-called Cooled-Stainless Tube (CST), for the determination of velocity profiles in flowing polymer melts in real processing machines such as the injection molding machine. The CST technique has also been used by Wood and Rasid (22) for the velocity profile measurement of the melts in a single-screw extruder.

To date, data on die swell behavior and velocity profiles of more complex processes, such as co-extrusion or multi-layer processes are still limited and required for solving many complex flow problems in various types of processing equipment. For example, in co-extrusion processes, the dimension of each material layer in the co-extruded products has to be determined accurately. This can be achieved by knowledge of radial die swell ratio profiles and their dependence on processing conditions. Recently, most experimental data are only available for the overall die swell, which is defined as the ratio of the extrudate size to the die size, and the explanation for the change in die swell is usually associated with the recoverable elastic deformation developed during flow through the die as discussed earlier (1). Scientific evidence (23-24) has clearly suggested that velocity profiles and die swell are closely related, but most work in literature has been carried out with these two parameters separately and theoretically. In this present work, we offer a new and simple experimental technique for simultaneous measurements of radial die swell and velocity profiles of a PS melt, the swell and velocity results being reported in each position across the die diameter. The changes in radial die swell results of the melt are explained in terms of their relation to the velocity profiles, calculations of shear rate profiles, and residence time. In addition, the effect of piston speed (wall shear rate) on the development of both radial die swell and velocity profiles of the melt has been discussed.

EXPERIMENTAL

Raw material

All tests in this work used a polystyrene (Styron 656D 267), supplied in granular form by Siam Polystyrene Co., Ltd (Bangkok, Thailand). It was characterized by a melt flow index of 7 and a density of 1.350 g/cm^3 (BS2782 Method 720A; 1979).

Design and manufacture of the experimental apparatus

General descriptions

In the present work, all measurements were carried out in a specially designed and manufactured capillary rheometer, whose barrel and die systems were connected to an AGS-500D (SHIMADZU) tensile testing machine. This design aimed to offer a new and simple technique, for the first time, to simultaneously measure radial die swell and velocity profiles of a polymer melt flowing in the capillary die of a rheometer. The proposed technique was based on a parallel co-extrusion of colored melt-layers into an uncolored melt-stream from the barrel into and out of a capillary die. The size (thickness) ratio of the generated melt layers flowing in and out of the die was followed, the extrudate swell ratio for any given radial positions across the die diameter being then calculated. For velocity profile measurements, the times taken for relatively fine particles, inserted into the melt at different radial position across the duct, to travel along with the flowing melt for a given distance in the die were monitored.

Experimental design and arrangement

A new experimental technique, the so-called the Parallel Co-extrusion Technique (PCT), was developed in this work to *simultaneously* measure the radial die swell and velocity profiles of a flowing polymer melt in an extrusion rheometer. An experimental rig was specially designed and manufactured, and consisted of a barrel, two piston systems, a die and die holder and operational control systems. All the experimental components were assembled and connected to a tensile testing machine with utmost care to ensure the alignment of the apparatus. The experimental arrangement of the PCT apparatus is shown in **Figure 1**.

Barrel: The barrel was made of SKD61 tooling steel that can be used at elevated temperatures, and was 40×10^{-3}m in diameter and 160×10^{-3}m in length. A small pressure hole (1×10^{-3}m in diameter) was located near the bottom of the barrel (near the die entrance) for pressure drop measurement. The inner surface of the barrel was hardened by hard chromium plating giving the hardness of approximately 55HRC. The barrel had four holes located along its length for installing heaters and a thermocouple (Type K).

Co-extruding pistons: The piston system in this work was specially designed in two separate parts (Piston#1 and Piston#2) so that a parallel co-extrusion process was possible. Piston#1 was used to extrude the bulk melt from the barrel into the die, as usually performed in a normal commercial rheometer. The diameter and length of Piston#1 were 40×10^{-3}m and 150×10^{-3}m, respectively. The piston body was made of stainless steel while its tip was made of copper. Piston#1 was hollow and was heated using 4 heater bars. Inside Piston#1, there were a series of small pistons (called Piston#2) located to extrude a number of colored melts loaded and heated in Piston#1 into the uncolored melt stream in the barrel. All the small pistons (Piston#2) containing colored PS melt, moved upward in the opposite direction to the Piston#1, which was moving down the barrel. This action would co-extrude the colored melt layers into the uncolored melt stream in Piston#1 at different radial positions across the barrel (and then die) diameter.

Die and die holder: The die used in this work was $64x10^{-3}$m in length and $5x10^{-3}$m in diameter, and could be divided into two parts, one being the outer part made of ST37 mild steel and the other being the inner part made of Borosilicate glass, that can withstand high temperature and pressure. The outer part was square-drilled over half the diameter of the die to allow the flow of the melt to be viewed. A separate die plate (holder), designed for holding the die, was bolted to the bottom of the barrel. The material used for the die holder was the same as for the die.

Visualization equipment: This aimed to visualize the flow of the co-extruded melt inside and outside the capillary die. A color video-camera (SSC-DC398P with 752Hx582V pixels and 480TV lines) and a high resolution macro zoom lens (3.3X magnification) were used to record the flows for further calculations of radial die swell and velocity profiles. All the results were recorded and displayed in real time using a personal computer.

Operation control systems: These can be divided into three separate systems: temperature, pressure, and test speed control systems.

1. The temperature control system: This consists of four heater bars, a type K thermocouple and a DD6 temperature controller, these being used for monitoring and controlling the temperature of the apparatus components (barrel, Piston#1 and Piston #2, die and die holder).

2. The pressure control system: This work uses a novel pressure sensor designed and developed by Sombatsompop *et al*, the so-called Photo-conductive Light Dependent (PLT) sensor whose details can be sought elsewhere (25). The sensor was situated at the base of the barrel just above the die face and connected using an adapter.
3. The test speed control system: Since the experimental rig used in this work was connected to the cross-head of the tensile testing machine, the test speed was controlled and monitored by use of a transducer that was supplied with the testing machine, the accuracy of the measurements being in the order of ±0.1%. The speed of the piston was used to calculate the wall shear rate.

Measurements of radial die swell and velocity profiles

The principle of the PCT technique involved the extruding of colored melt-layers contained in Piston#2 into the bulk uncolored (transparent) polymer melt in the barrel. At the same time, the melt in the barrel continuously flowed into the die by the movement of Piston#1. The colored melt layers in the barrel flowed from the barrel into the die at the same velocity as the uncolored melt – this can be confirmed by the flow property results of the colored and uncolored melt-layers which will be shown later. The colored melt layers then appeared inside and outside the die at various radial positions, which corresponded to the positions of the melt layers which were determined in the barrel. **Figure 2** shows a schematic diagram of thickness measurements of melt layers in the die region and in the extrudate outside the die for radial die swell measurements. During the experiment, we observed that the flow of the colored PS melt-layers across the barrel and capillary diameters was very symmetric along the barrel and capillary axis. Therefore, the measurements of die swell and velocity profiles of the melt-layers were made only on one side of the die diameter, the data on the other side of the die being then generated based on the symmetric nature of the flow.

Radial die swell profiles

Through trial and error experiments, we found that the swelling of the PS melt under the test conditions used in this work attained equilibrium around 3mm from the die exit. Therefore, the degree of die swell was measured at 5mm from the die exit. The radial die swell ratio values (B_r) were obtained by comparing the thickness of the colored layer of the extrudate outside the die, for a given reduced radius (r/R) position in the die, to that inside the die as illustrated in **Figure 2** and **Equation 1**. The r/R range of interest used in this work was from 0.0 to 0.86. The accuracy of the radial die swell profile can be verified by averaging the values of the radial die swell (B_r) obtained across die diameter, and comparing with the overall die swell ratio ($B_{overall}$) directly measured at the die exit. This statement was based on the fact that the overall die swell was a result of swelling and de-swelling (contracting) of the melt layers across die diameter. The $B_{overall}$ value was referred to as the ratio of the extrudate radius (R_{ext}) to the die radius (R_d) (as shown in **Equation 2**).

$$B_r = \frac{dr_{ext}}{dr_d} \tag{1}$$

$$B_{overall} = \frac{R_{ext}}{R_d} \tag{2}$$

Previous work (12-13) has shown that flow patterns in the barrel changed continuously with piston displacement, and a fully developed laminar flow seemed to be exhibited when the piston displacement reached approximately ½ the barrel length. Therefore, die swell ratio values at any radial positions across the die were averaged and obtained at the piston displacement range of 85-92mm down the barrel. In this work, the speed of Piston#1 was varied from 0.050×10^{-3}, 0.083×10^{-3} and 0.166×10^{-3} m/s (corresponding to wall shear rates of 5.1, 8.5 and 17.1 s^{-1}, respectively) in order to study the effect of the extrusion rate on the change of the die swell ratio. The test temperature used was 200°C.

Velocity profile measurement

In this work, foreign particles were introduced into the melt system for measuring velocity profiles of the PS melt. The foreign particles used were natural corn fibers, supplied by V.P. Plastics Products (1993) Co., Ltd (Bangkok Thailand). The particles were relatively small (having an average particle size of 250 μm) and light so that they would not settle out in the melt stream. Only 0.2% of corn particles were added into the extrudate and this particle concentration did not affect the rheological properties of the melt. The measurements were carried out by recording the times taken for the small particles loaded into the melt layers to travel for a given distance in the die. The experimental procedure for the determination of velocity profiles of melt commenced by preparing a colored polymer composite using a twin screw extruder (Haake Polylab-Rheomex CTW100P, Germany) at 40rpm rotating screw speed and a die temperature of 200°C. The composites were pigmented using CI Solvent Red-24, supplied by Orient Chemical Industries, Co., Ltd (Osaka, Japan). The experimental apparatus used for die swell measurement was also employed for velocity profile determinations. The melt velocity for any given radial position was obtained by the measurement of the time taken for the particles to travel along the flow path for a given distance (10mm before the die exit). It should be noted that the melt velocity was *simultaneously* measured with the die swell ratio, melt velocity values at any radial positions across the die being averaged and obtained at the piston displacement range of 85-92mm down the barrel. The accuracy of the velocity profile measurement can be verified by integrating the radial melt velocity (V_r) for any given die radius (r) as expressed in **Equation 3**, to give the output rate (Q).

$$Q = 2\pi \int_0^R r V_r dr \tag{3}$$

It should also be noted that all the measurements in this work were restricted to the isothermal state, defined as a condition in which the temperature of the melt and the duct wall are the same (22), and the melt velocities at the duct wall were assumed to be zero (1).

Flow property measurement

The relationship between wall shear stress and wall shear rate for the PS melt, using the same rheometer, die and test conditions as used for radial die swell and velocity profile measurements, were established, the calculations being obtained elsewhere (14). In this work, Bagley's corrections were not applied to the generated shear stress data, as the shear stress data were solely used for comparative reasons to illustrate the magnitude of the changes observed in the flow characteristics of the materials as a function of the piston speeds used.

RESULTS AND DISCUSSION

Effect of particles and pigment on the change in melt properties

Since this work used the color technique to reveal the flow visualization of PS melt in order to evaluate the radial die swell profile, and also utilized light corn particles to facilitate the measurement of radial velocity profiles of the melt, it is reasonably required to study the effect of the pigment and corn particles on the rheological properties of the PS melt. This was carried out using the same rheometer as utilized for the measurements of die swell and velocity profiles. **Figure 3** shows the results of wall shear stress and wall shear rate for the PS melt with and without pigment, and the PS melt sample with corn particles. It can be seen that all the curves were very similar, the differences being within the experimental error of ±2.5%. This confirmed that the introduction of the pigment and the corn particles did not affect the overall flow properties of the melt as the amounts of these foreign objects were relatively small.

Flow patterns and general observation on overall die swell ratio

Figure 4 shows an example of the flow patterns of colored and uncolored PS melt layers flowing in the rheometer. **Figure 4a** reveals the flows in the barrel and die entry regions while **Figure 4b** shows the flows near the die exit and outside the die. It should be noted that the flow-specimen rod in **Figure 4a** was produced by cooling the flow system down to room temperature using an air cooling system, after a specified piston displacement. The cooled specimen rod was then removed from the Rheometer before being photographed. It can be seen that the flow patterns in the barrel and near the die entrance of PS melt were very simple, the material in the barrel moving radially towards the die. For a given melt layer, the thickness of the melt layer decreased as it flowed from the barrel to the die. On exiting the die, the thickness of the melt layer became greater, this being referred to as swelling of the extrudate. **Figure 5** shows the overall die swell ($B_{overall}$) of PS melt as a function of piston displacement for three different piston speeds (0.050×10^{-3}, 0.083×10^{-3} and 0.166×10^{-3} m/s) at a die temperature of 200°C. It can be seen that, for a given piston speed, the overall die swell ratio was in the equilibrium condition at the piston displacements from 85 to 92 mm, the differences in the swelling ratios for various piston displacements being within the experimental error of ±2.5%. The overall die swell ratio ranged from 1.25 to 1.38, this swelling value for PS melt being similar to that found in previous work (12). The swelling ratio appeared to increase with increasing piston speed as was expected because the extrusion speed of the piston is a direct function of shearing stress and wall shear rate generated during the flow, and die swell is generally associated with the stored elastic stress (shearing stress), which is released on exit from the die (9).

Development of radial die swell and velocity profiles

Figure 6 illustrates the die swell ratio as a function of the reduced radial (r/R) position in the die for three different piston speeds. It can be seen that, for a given piston speed, the radial swell ratio decreased with increasing r/R position. The highest die swell ratio occurring at the duct center, was in the range of 1.6-1.9. The lowest value of die swell ratio was 0.9, which occurred around r/R positions of 0.6-0.86. In most literature, die swell is reported as being related to the stored elastic stress which is released on exit from the die. This stress is predominantly due to the shape of the velocity profile in the die which is believed to be parabolic in nature; the shearing stress (also shear rate) will be high at the die wall and low in the center of the flow (1,13). Therefore, one would expect the die swell across the die to be high at the wall and low at the center. However, this does not seem to be the case as evidenced by the results in **Figure 6**. The variations of die swell ratio of the melt across the die diameter in this work can be explained using the velocity profiles of PS melt which were simultaneously measured in experiments. **Figure 7** shows the velocities of the melt as a function of reduced radial position for different piston speeds - the effect of piston speed on the die swell profiles will be discussed later. It can be noticed that the general trend of the radial velocity profiles was very similar to that for the die swell profiles. The highest melt velocity was found at the center of the die, the melt velocity decreasing with increasing r/R value due to the drag effect (20-21). It was thought in this work that the shape of the radial die swell profiles (in **Figure 6**) was associated with the development of velocity profiles in the die (in **Figure 7**) and the equalization of the velocity profiles at the die exit. That is, when the melt was flowing in the die it had a relatively high velocity at the center and relatively low near the die wall as can be expected in shear flow. On exiting the die, the velocities of the melt across the die diameter had to equalize to form a plug flow. During this flow pattern transition, the melt velocity at the center had to slow down while that near the wall accelerated. A sudden reduction in melt velocity at the center causes the melt to flow radially and led to an expansion of the melt in radial direction. This extrudate expansion suppressed the dimension change of the melt near the die wall - the die swell ratio near the die wall decreased in order to maintain the overall swell ratio of the extrudate for a given piston speed. This explains why the extrudate swell ratio was relatively high at the center of the die.

The velocity profile results were then utilized to calculate radial shear rate profiles (γ_r) of the melt flowing in the die, whose results are shown in **Figure 8**. In this work, the maximum shear rate was found around r/R of 0.6. In general, one would expect the maximum shear rate to be at the die wall, provided that the velocity profile of the melt was parabolic. However, the shear rate at the die wall was not considered in this particular case because the melt velocity at the die wall is quantitatively impossible to experimentally measure. Another interesting point to consider is that, the shape of the shear rate profiles did not correspond to that of the die swell profiles. Taking account of the results in **Figures 5, 6** and **8**, it can be implied that the wall shear rate was the main factor for controlling the *overall* die swell ($B_{overall}$), but it did not correspond to the shape of the die swell profiles across the duct. This has a practical implication when determining the size of the inner layer of the melt in a co-extrusion process. That is, an ideal shear rate profile (low value at the center and high near the wall) cannot be used to accurately predict the size of the inner layer of a co-extruded product. **Figure 9** shows the calculated residence time profiles of the PS melt and the results can be observed to correspond well with the die swell profiles - the smaller the residence time the higher the swell ratio.

Accuracy of the radial die swell ratios and velocity profiles

As stated earlier, the accuracy of the radial die swell profile measured by PCT technique can be verified by averaging the values of the radial die swell (B_r) obtained across die diameter, and comparing the result with the overall die swell ratio ($B_{overall}$) determined at the die exit. Similarly, the accuracy of the radial velocity profile measurement can be verified by integrating the profile across the duct, and comparing the result with the total output rate. **Table 1** shows the comparison of $B_{overall}$ and average B_r values for PS melt for three different piston speeds. It can be seen that the degrees of extrudate swell ratio obtained from both methods were not the same, the differences being relatively large especially at 0.166×10^{-3} m/s piston speed. This gives the average value of %difference of 7.82%. The errors could be further improved, but beyond of the scope of this paper, by increasing the number of measuring points across the duct diameter, and also attempting to measure the die swell ratio near the die wall as closely as possible. Considering this difference, the accuracy of the radial die swell ratios determined by PCT technique was 92.2%.

Table 2 shows a comparison of actual and experimental output rates for PS melt at different piston speeds. It can be seen that the values of experimental output rates for three piston speeds were similar to those of the actual output rates, the average differences being calculated to be 9.13%. The errors in the calculation of the experimental output rates may arise due to a number of flow related factors, such as assumption of zero-melt-velocity at the die wall (21), compressible and de-compressible nature of the melt (26), and melt leakage between the piston and barrel wall (14). The first effect is usually assumed since the melt velocity at the die wall is quantitatively impossible to experimentally measure, and this could result in a decrease of the measured output rate while the second effect causes a fluctuation in the measured output rate. Finally, the value of the actual output rate may possibly be an overestimate due to possible leakage of the melt through the clearance of the barrel-piston, this effect being most possibly at high extrusion rate. Similar finding was also found in previous work on measuring the velocity profiles of a low-density polyethylene melt flowing in an injection molding machine which stated that some differences in the output rates obtained from integration of an experimental velocity profile and the piston speed in the barrel were expected (27). Consideration of the errors suggested that the velocities determined experimentally in this work were accurate to within 90.8%.

Effect of piston speed on radial die swell ratio and velocity profiles

Figures 6 and 7 were also used to study the effect of piston speed on radial die swell and velocity profiles of PS melt, respectively. The piston speeds of 0.050×10^{-3}, 0.083×10^{-3}

and 0.166×10^{-3} m/s corresponded to the wall shear rates of 5.1, 8.5 and 17.1 s^{-1}, respectively. It was found that the die swell profiles of the melt changed with piston speed, especially around the center of the die and at a r/R of 0.60-0.86. As the piston speed increased from 0.050×10^{-3} to 0.166×10^{-3} m/s, the swell ratio of the melt at the center decreased, accompanied by an increase in the swell ratio near the die wall (around r/R of 0.86). The change in die swell ratio for piston speeds of 0.050×10^{-3} and 0.083×10^{-3} was not significant, which corresponded well with the shear rate profiles **(Figure 8)** for these two piston speeds. The decrease in the die swell at the centre and the increase in the die swell near the wall when considering the die swell profiles at piston speeds of 0.050×10^{-3} and 0.166×10^{-3} m/s can be explained in association with the changes in the velocity profiles **(Figure 7)** or shear rate profiles **(Figure 8)** as follows.

Decrease in the die swell at the center: The radial velocity profiles in **Figure 7** were observed to change with piston speed, the velocity profiles at lower piston speed being relatively flatter. It can be expected that the viscosity of the melt would decrease with increasing shear rate (or piston speed), the decrease in the melt viscosity then influencing the velocity profile. Work by Sombatsompop and Chaiwattapipat (28) investigated the effect of the injection speed on melt temperature profiles of polypropylene melt in a circular duct of an injection molder, and clearly indicated that the increase in melt temperature appeared to be greater at the center of the duct for higher injection speeds due to greater shear heating. In the case of a lower injection speed, the melt temperature was relatively high near the duct wall. In relation to this work, the increase in melt velocity at the duct center for high piston speed resulted from an increase in melt temperature and then a decrease in melt viscosity. A similar view was evidenced by

Wilczynski et al (6). Hence, the decrease in the die swell of the melt at the center of the die with the higher piston speed was caused by the viscosity reduction of the melt.

Increase in die swell near the die wall: This can be explained using the shear rate profile results as shown in **Figure 8**. It can be seen that the shear rate of the melt near the die wall at a piston speed of 0.166×10^{-3} m/s was much greater than that at piston speed of 0.050×10^{-3}, this increased shear rate resulting in the increase in die swell. It should also be noted that the increased die swell suppressed the expansion of the melt around the center of the die.

In summary, it was found that the PCT technique was simple and versatile for the simultaneous measurement of the radial die swell and velocity profiles of a polymer melt inside a small capillary die. This technique can be improved to obtain more accurate experimental results by increasing the number of colored melt layers across the die diameter. This work will be extended, beyond the scope of this paper, to investigate the effect of processing conditions such as test temperature and die geometry, on the die swell and velocity profiles of the melt, as well as carrying out such measurements in some other types of processing equipment such as single screw extruders.

CONCLUSION

The parallel co-extrusion technique (PCT) proposed in this paper was very simple, versatile and useful for simultaneous measurements of radial die swell and velocity profiles of a flowing polystyrene melt in the capillary die of a constant shear rate rheometer. The proposed technique was based on a parallel co-extrusion of colored melt-layers into uncolored melt-stream from a barrel into and out of the capillary die. The radial die swell profiles were formed by measuring the thickness ratio of the generated melt layers flowing in and out of the die while the radial velocity profiles were generated by introducing relatively light and small particles into the melt layers. The radial die swell and velocity profiles for PS melt determined experimentally in this work were accurate to 92.2% and 90.8%, respectively. The overall die swell ratio of the PS melt increased with increasing piston speed, the overall die swell values ranging from 1.25 to 1.38. It was found that the general trend of the radial velocity profiles was very similar to that for the die swell profiles. The die swell ratio was found to be high at the center (~ 1.9) and low (~ 0.9) near the die wall. The swell ratio of the melt at the center decreased with increasing piston speed. Finally, the distance from the die exit to reach the equilibrium swelling was shorter as the piston speed was increased.

ACKNOWLEDGMENTS

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List of Figures

List of Tables

Table 1: Comparison of $B_{overall}$ and average value of B_r across the die diameter for PS melt at different piston speeds

Piston speed (10^{-3} m/s)	$B_{overall}$	Average B_r	% differences
0.050	1.26	1.19	5.55
0.083	1.31	1.38	5.34
0.166	1.35	1.18	12.59

% Average difference = 7.82%

Table 2: Comparison of actual and experimental output rates for PS melt at different piston speeds

Piston speed (10^{-3} m/s)	Output rate (Q, 10^{-7} m^3/s)		% differences
	Actual value	Experimental value	
0.050	0.62	0.68	9.67
0.083	1.04	1.15	9.61
0.166	2.09	1.92	8.13

% *Average difference = 9.13%*

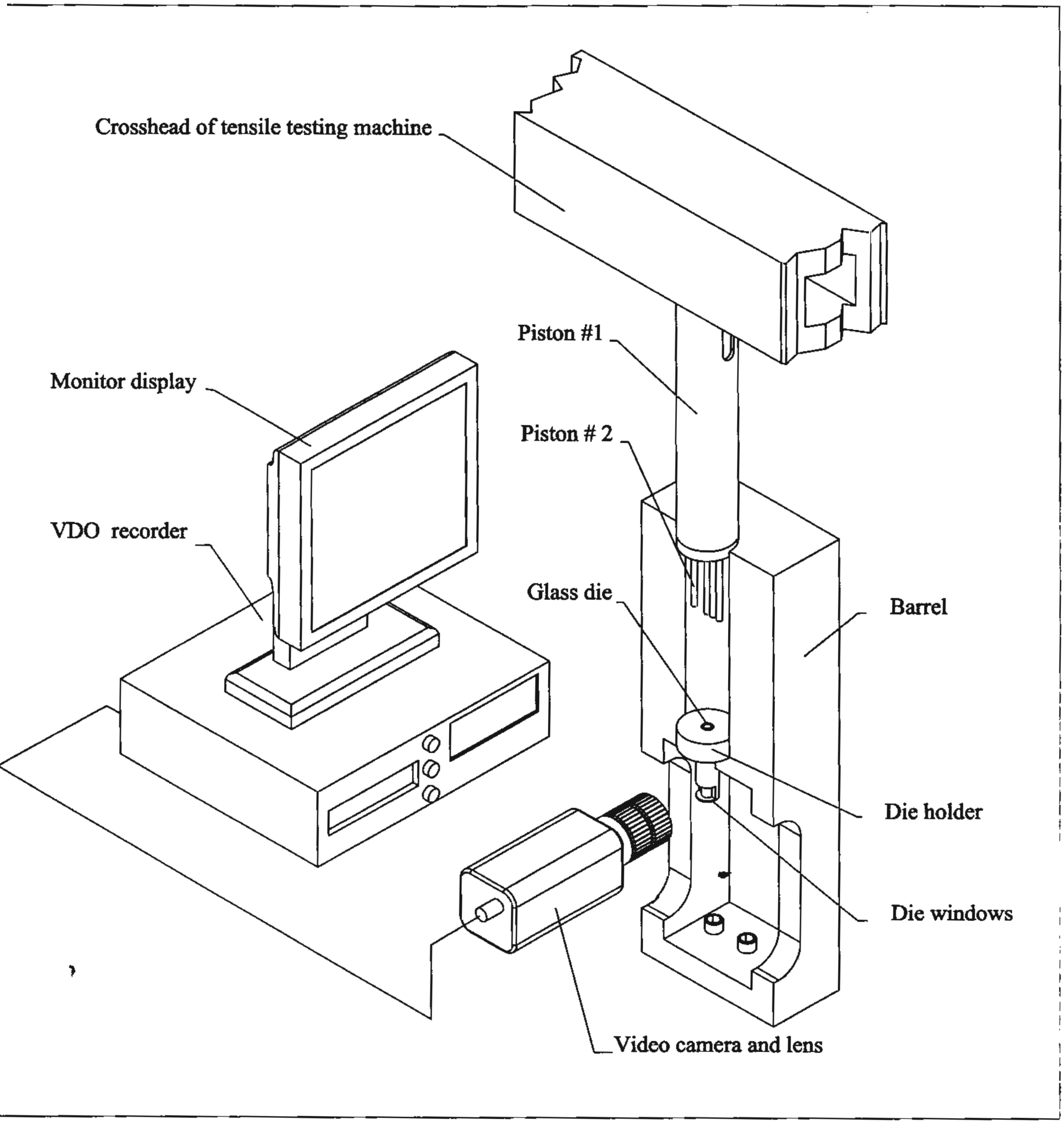
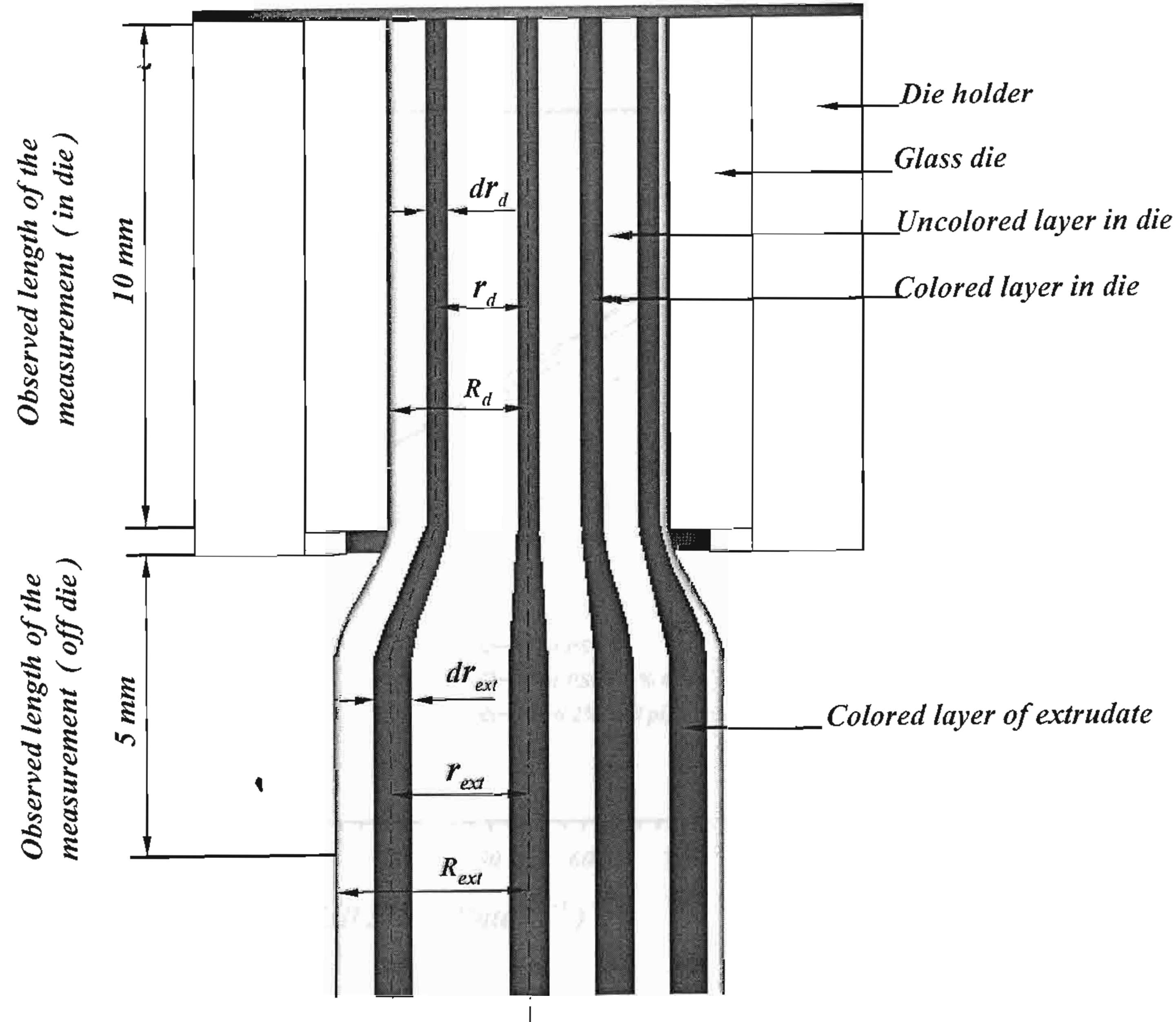
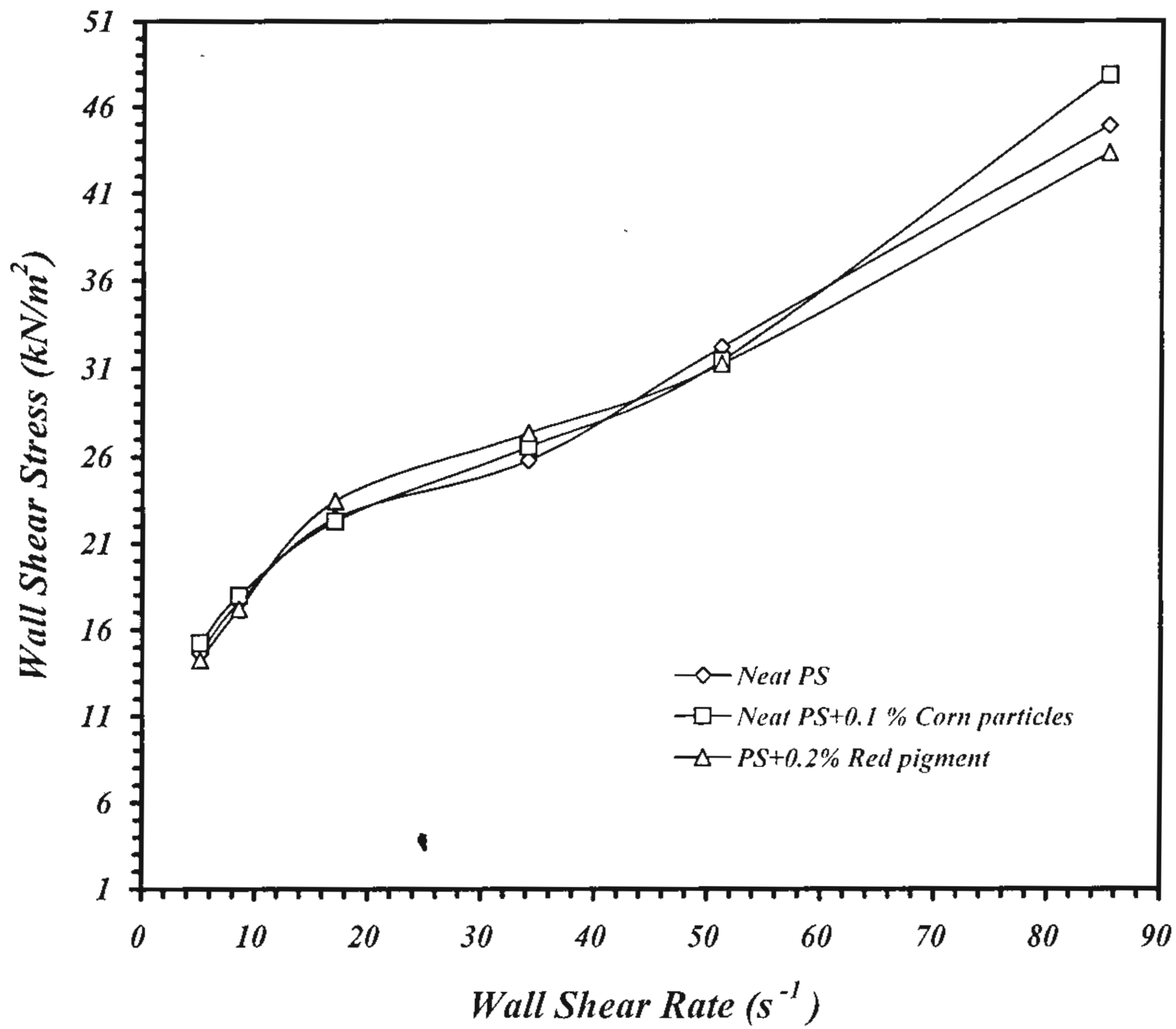

Crosshead of tensile testing machine
Piston #1
Monitor display
Piston # 2
VDO recorder
Glass die
Barrel
Die holder
Die windows
Video camera and lens

Figure 1



Observed length of the measurement (in die)
10 mm
Observed length of the measurement (off die)
5 mm
dr_d
r_d
R_d
dr_{ext}
r_{ext}
R_{ext}
Die holder
Glass die
Uncolored layer in die
Colored layer in die
Colored layer of extrudate

Figure 2



51
46
41
36
31
26
21
16
11
6
1
0
10
20
30
40
50
60
70
80
90
Wall Shear Stress (kN/m²)
Wall Shear Rate (s⁻¹)
Neat PS
Neat PS+0.1 % Corn particles
PS+0.2% Red pigment

Figure 3

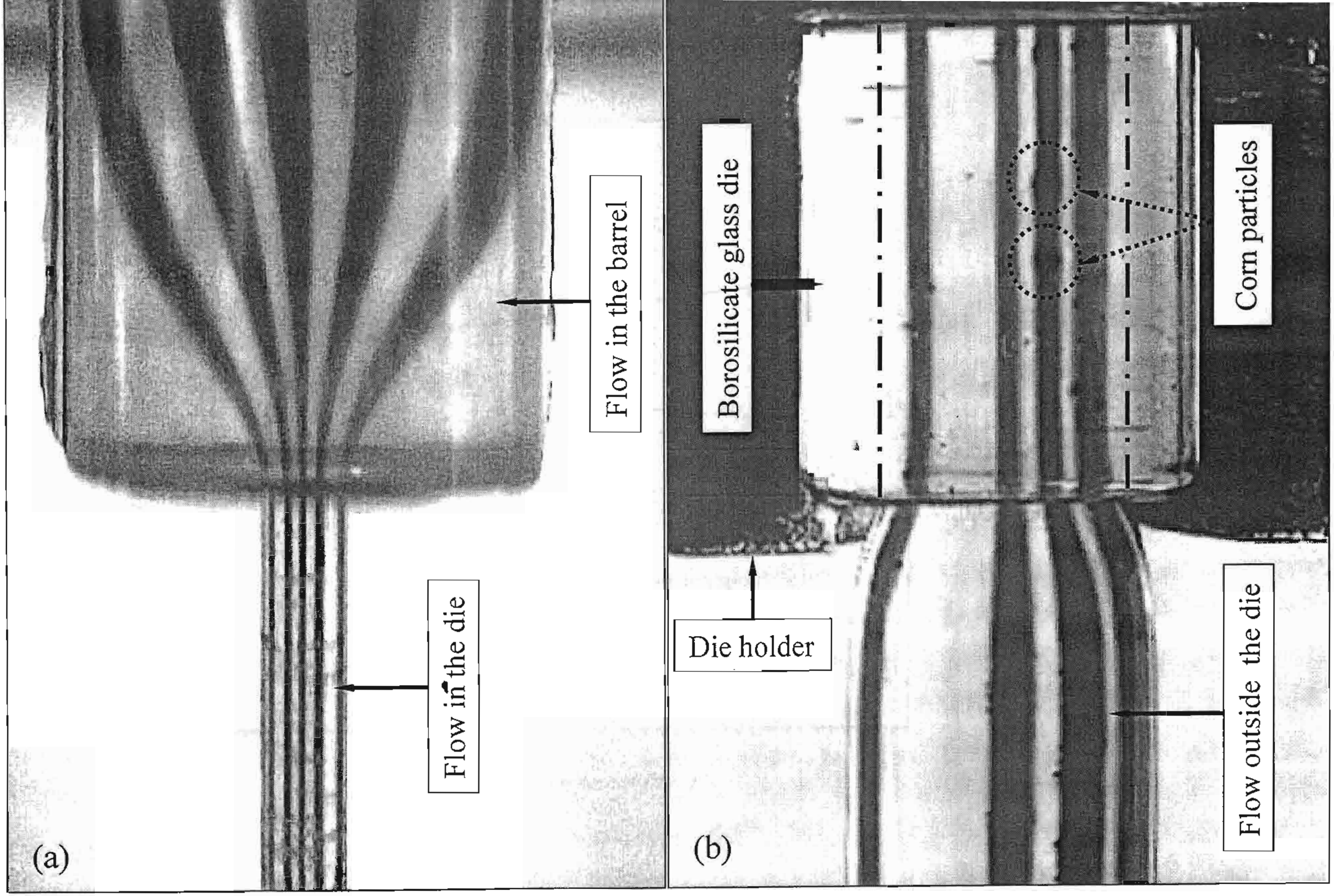

Flow in the barrel
Flow in the die
(a)
Borosilicate glass die
Corn particles
Die holder
Flow outside the die
(b)

Figure 4

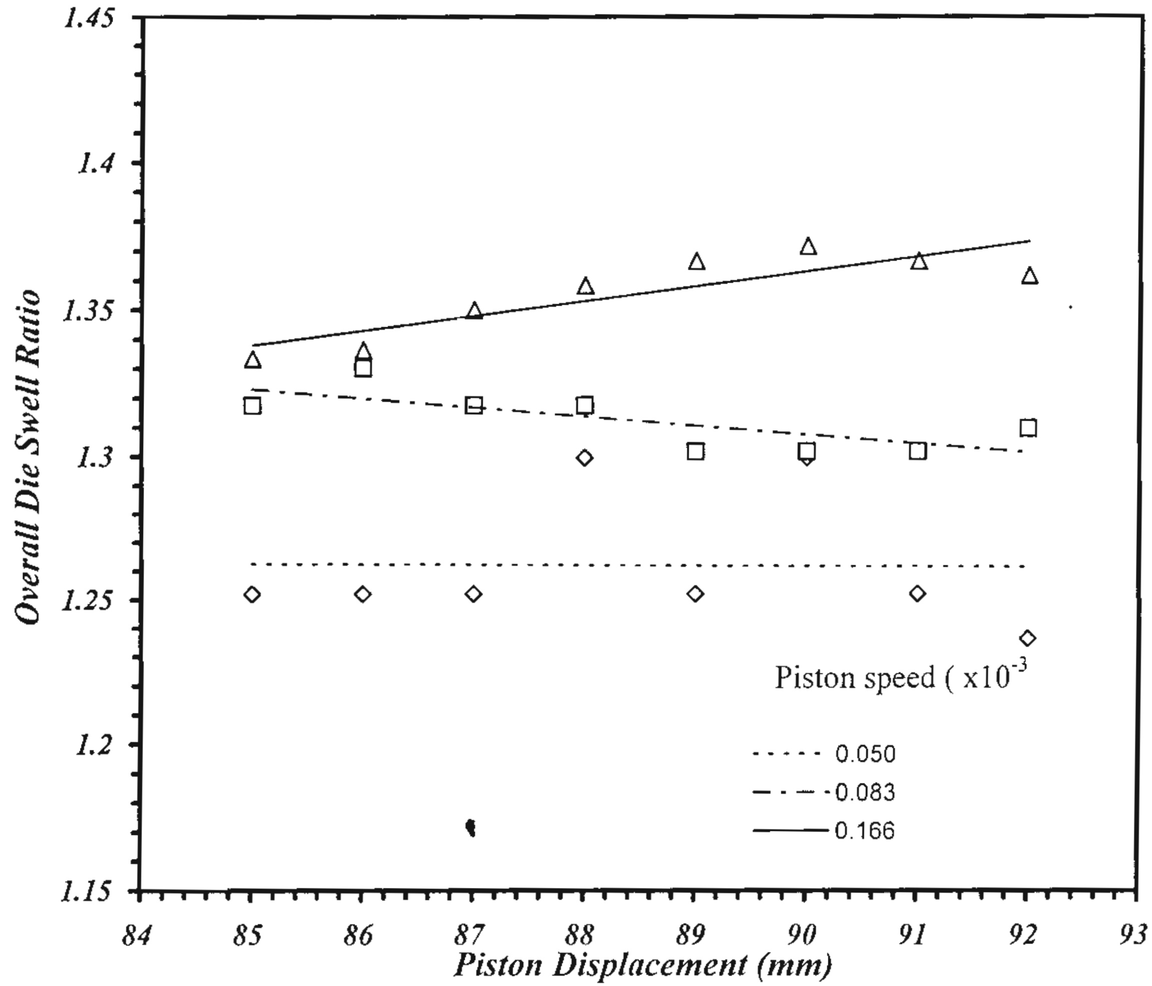

1.45
1.4
1.35
1.3
1.25
1.2
1.15
Overall Die Swell Ratio
84
85
86
87
88
89
90
91
92
93
Piston Displacement (mm)
Piston speed (x10-3
0.050
0.083
0.166

Figure 5

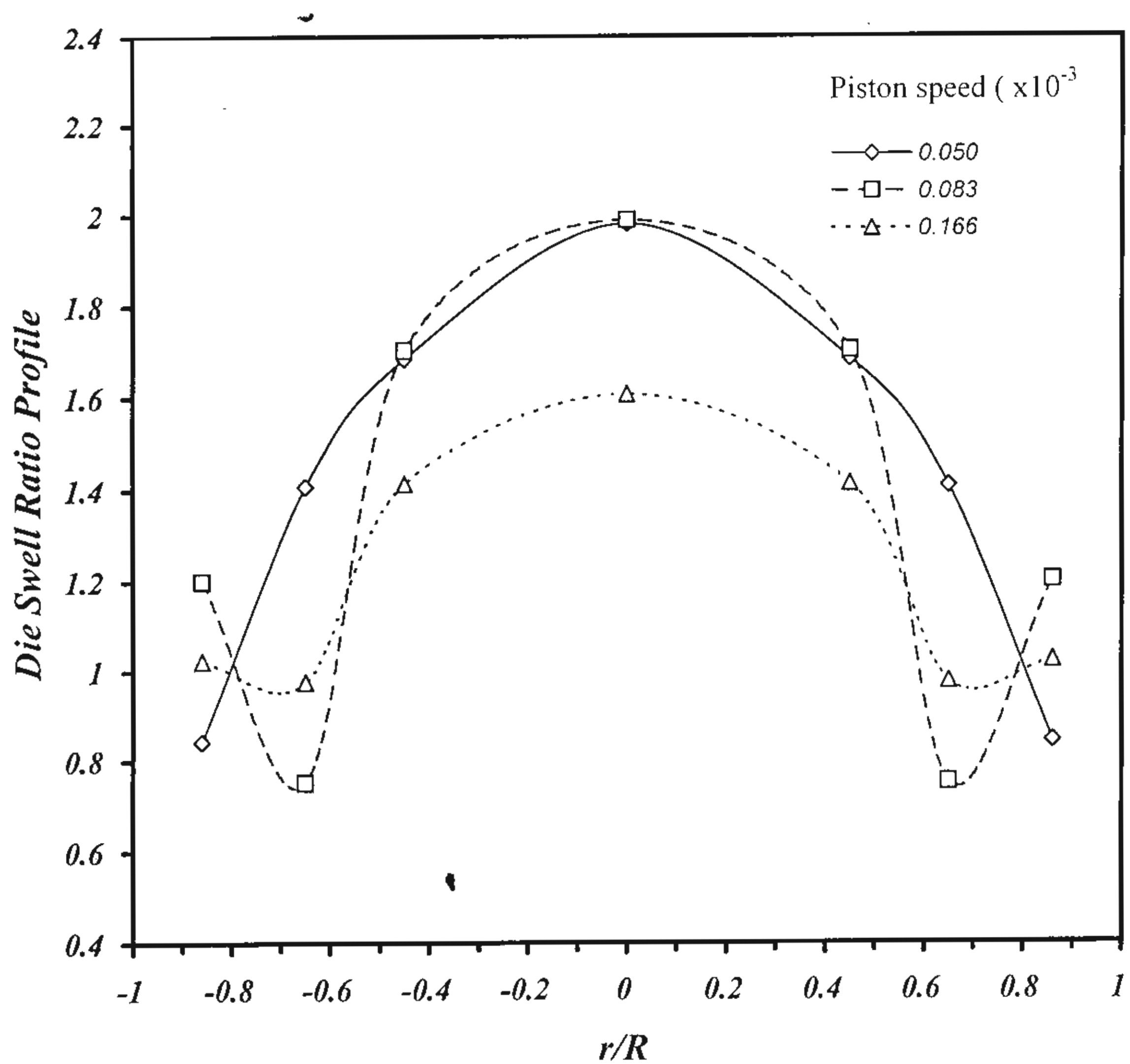
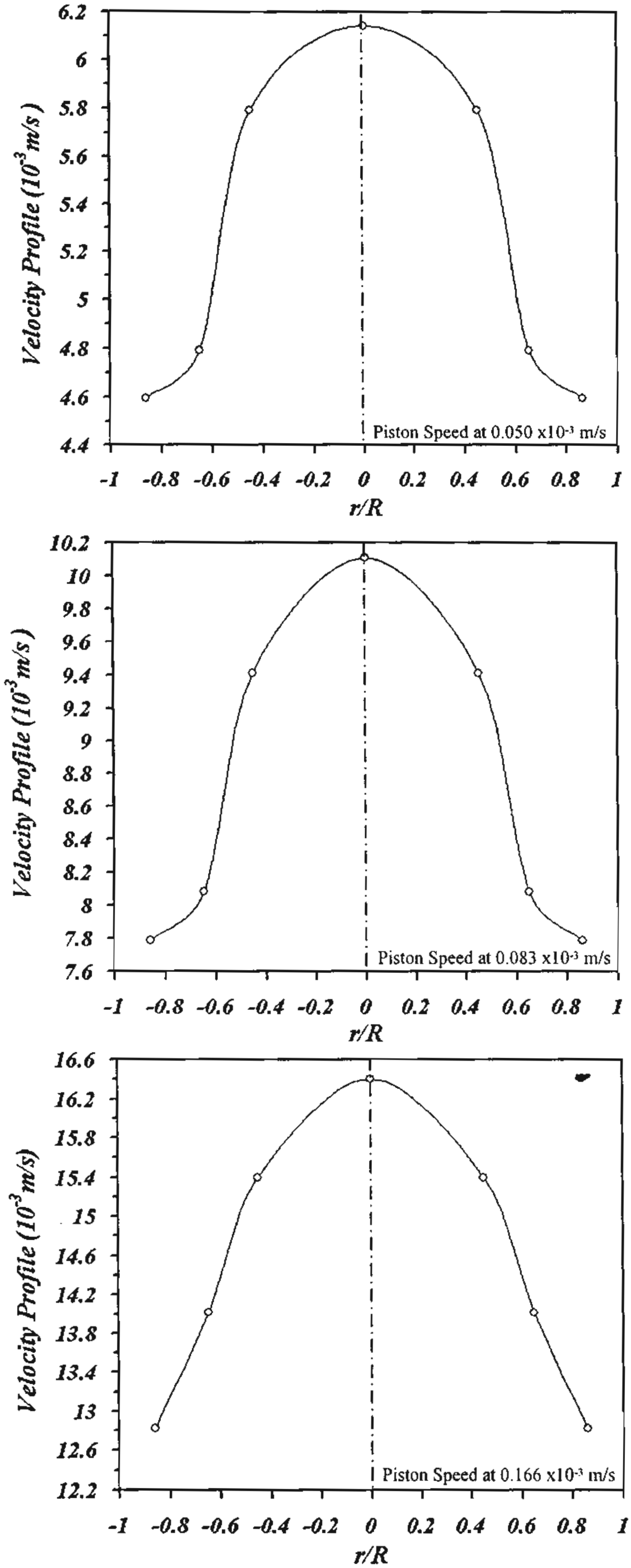

Piston speed (x10^-3
0.050
0.083
0.166
Die Swell Ratio Profile
2.4
2.2
2
1.8
1.6
1.4
1.2
1
0.8
0.6
0.4
-1
-0.8
-0.6
-0.4
-0.2
0
0.2
0.4
0.6
0.8
1
r/R

Figure 6



Velocity Profile (10^{-3} m/s)
r/R
Piston Speed at 0.050 x10-3 m/s
Piston Speed at 0.083 x10-3 m/s
Piston Speed at 0.166 x10-3 m/s

Figure 7

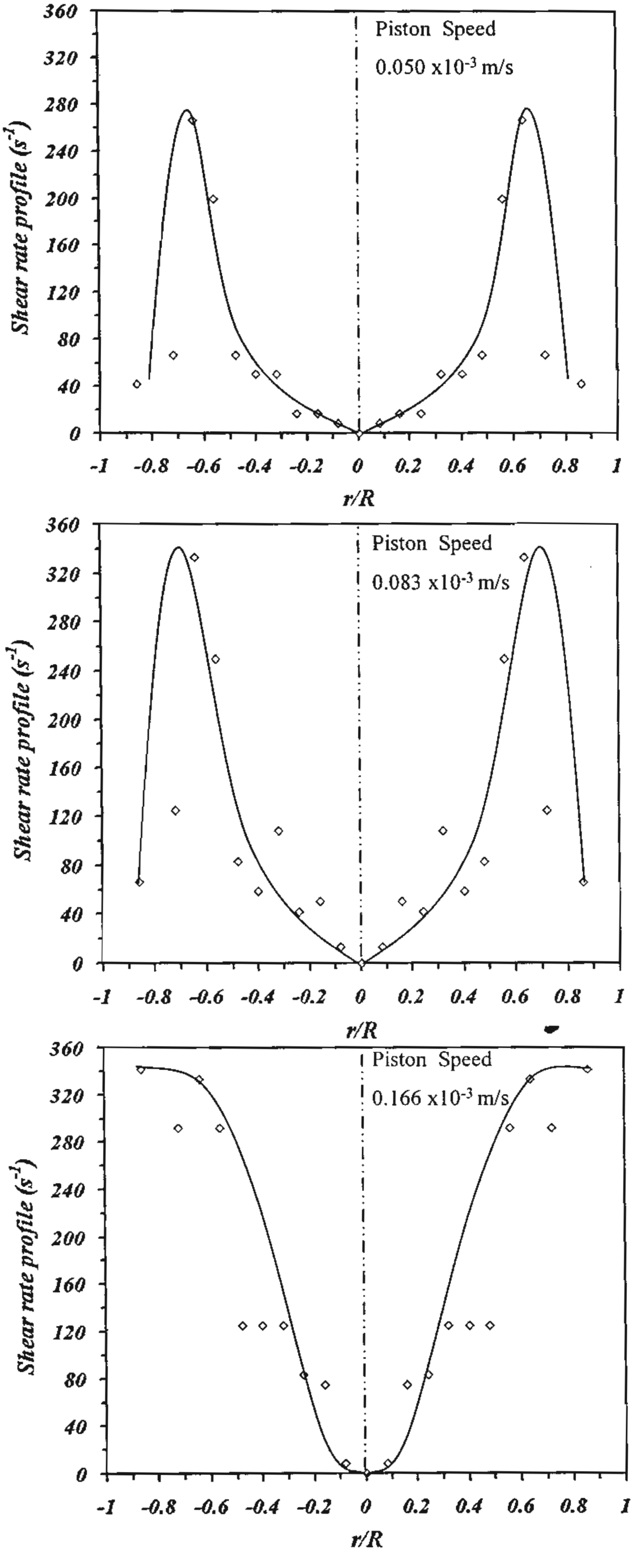

Piston Speed
0.050 x10-3 m/s
Piston Speed
0.083 x10-3 m/s
Piston Speed
0.166 x10-3 m/s
Shear rate profile (s-1)
r/R
360
320
280
240
200
160
120
80
40
0
-1
-0.8
-0.6
-0.4
-0.2
0
0.2
0.4
0.6
0.8
1

Figure 8

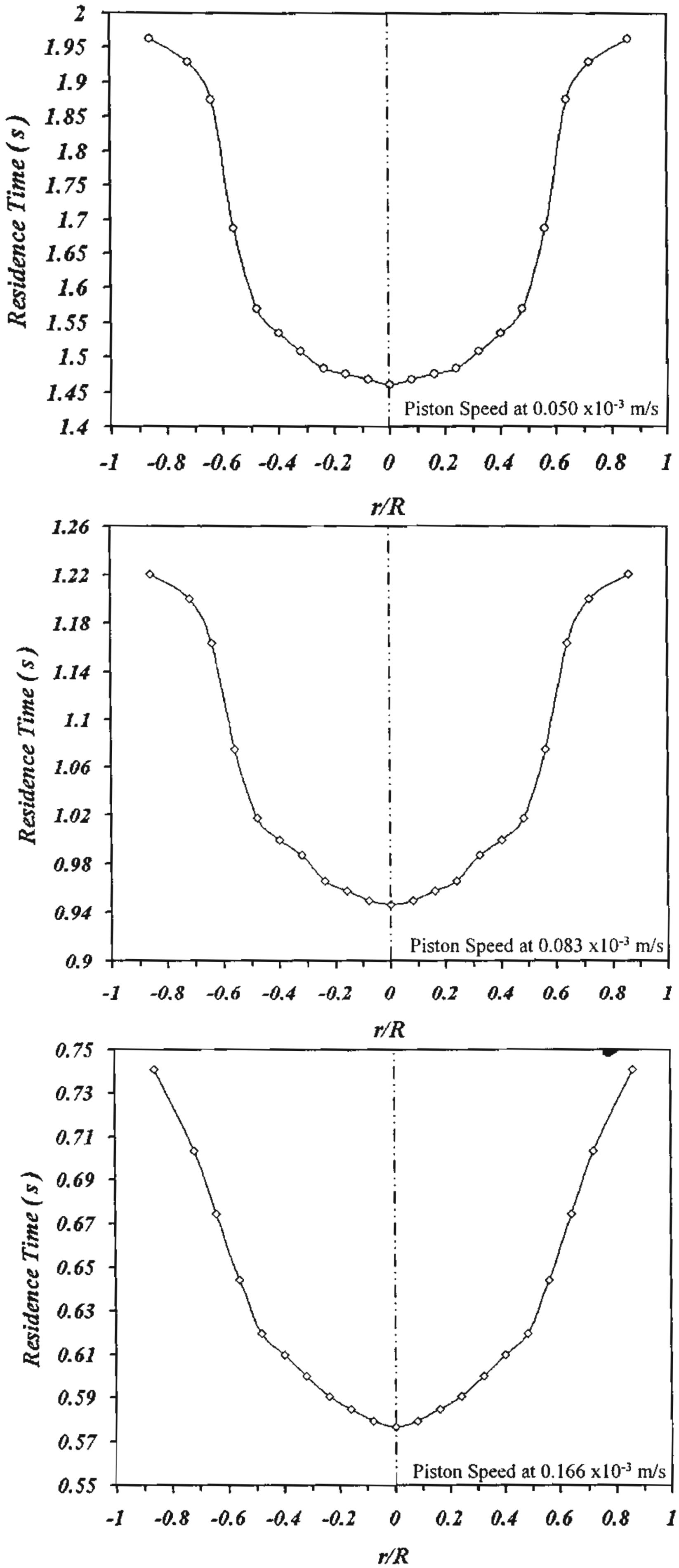

Residence Time (s)
r/R
Piston Speed at 0.050 x10^-3 m/s
Piston Speed at 0.083 x10^-3 m/s
Piston Speed at 0.166 x10^-3 m/s

Figure 9

Experimental Studies on Radial Die Swell and Velocity Profiles of Flowing PS Melt in an Electro-magnetized Die of an Extrusion Rheometer

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Abstract

This article continues from previous work (*Intawong & Sombatsompop; 2004, accepted for publication by Polym. Eng. Sci.)* investigating the radial die swell and velocity profiles of polystyrene melt in a capillary die of a constant shear-rate extrusion rheometer, using a parallel co-extrusion technique. In this article, an electro-magnetized capillary die was used to monitor the changes in the radial die swell profiles of the melt, this being relatively novel in polymer processing. The magnetic flux density applied to the capillary die was varied in a parallel direction to the melt flow, and all tests were performed under the critical condition at which sharkskin and melt fracture did not occur in the normal die. The experimental results suggest that the overall die swell for all shear rates increased with increasing magnetic flux density to a maximum value and then decreased at higher densities. The maximum swelling peak of the melt appeared to shift to higher magnetic flux density, and the value of the maximum swell decreased with increasing wall shear rate and die temperature. The effect of magnetic torque on the die swell ratio of PS melt was more pronounced when extruding the melt at low shear rates and low die temperatures. For radial die swell and velocity profiles, the radial swell ratio for a given shear rate decreased with increasing r/R position. There were two regions where the changes in the die swell ratio across the die diameter were obvious with changing magnetic torque and shear rate, one around the duct centre and the other around r/R of 0.65-0.85. The changes in the die swell profiles across the die diameter were associated with, and can be explained using, the melt velocity profiles generated during the flow. In summary, the changes in the overall die swell ratio of PS melt in a capillary die were influenced more by the swelling of the melt around the centre of the die.

Key words: Die swell, velocity profiles, extrusion, polymer melt

Introduction

The co-extrusion process of polymer products involves two or more melt streams flowing together along a die. Knowledge of the flow properties of each melt stream is very essential in terms of controlling the size and the quality of the melt streams, and the final products. Studies of such flow properties and flow patterns have been carried out both experimentally and theoretically. In the experimental investigations, the flow patterns of the melt layers flowing in the die have been found to be very complex as a result of differences in melt viscosity of the extruded layers across the flow channel (1-2). The complexity of the flows affects the properties of the melt stream and its instability during the process. In a conventional way, the stability of the melt can be characterized by a number of terms such as extrudate swell, sharkskin and melt fracture (3). The last two phenomena usually occur when polymer melts are extruded through a die at rates exceeding a critical shear stress (4-5). Extrudate swell is considered one of the main factors to determine quality and dimension of the polymer product and is usually explained in two different mechanisms, one being due to the recoverable elastic deformation developed during flow (6-8), and the other in association with the re-organization of melt velocity profiles developed in the die and the die exit (9-13). Christodoulou *et al* (11) used a direct measurement to investigate the swelling ratio of ethylene-vinyl acetate (EVA) copolymer through a newly developed co-extrusion technique coupled with a color-layer technique in a capillary rheometer. The experimental results suggested that the extrudate swell was mainly associated with the development of velocity profiles along the die. The velocity profiles of the melt could also have been affected by molecular structure which has an influence on the swelling ratio of the melt at the die exit, this also being supported by Munstedt *et al* (12).

It is widely known that both extrudate swell and melt velocity profiles of polymer melts are affected by the design of the dies. The effects become very complex when multi-layer flows are involved. Most published papers studied the dependency of extrudate swell and velocity profiles of polymer melts on the die size, die shapes (capillary, slit, annular and profiled dies), and number of flow channels in a die (14-21). Apart from these papers, there have been some recent publications dealing with relatively new die design related parameters such as ultrasonic irradiation and magnetic field application during extrusion (22-24). Cao and Li (22) offered a different parameter to alter the swelling ratio of a polypropylene melt by applying ultrasonic irradiation during extrusion and they found that the use of appropriate irradiation intensity reduced the extrudate swell ratio, as well as improve the apparent quality of the extrudate. Kimura (23) made an extensive review on the effect of a magnetic field on various polymer systems, semi-crystallize and amorphous polymers, during melting and crystallizing states. He concluded that the effect of a magnetic field on non-magnetic polymers was small, but occurred in anisotropic polymers. A change in the alignment of the polymer chains under the magnetic field was proposed to be a result of magnetic torque, which was caused by the induced motion of electrons under the applied magnetic field. In the case of atactic polymers (amorphous polymers) which lack anisotropic structures, the alignment of the molecules under the magnetic field became very difficult and direction of the magnetic alignment of these atactic polymers is so far still unknown. Recent work by Sombatsompop (24) studied the effect of magnetic field at 2.16T flux density on the extrudate swell ratio of polystyrene (PS) melt in a capillary die of a capillary rheometer, and the results suggested that the application of a magnetic field to the die could result in a significant increase in the swelling ratio of the PS melt of up to 25%. The author proposed that the increased swelling of the PS could have been caused by the change in molecular orientation or alignment. The application of a magnetic field to the die and the monitoring of the swell ratio of polymer melts in real testing/processing machineries are relatively new in polymer processing applications.

In this article, we aimed to simultaneously measure the radial extrudate swell and velocity profiles of polystyrene melt using the parallel co-extrusion technique as detailed in our previous work (25), and to extend this work by applying an electro-magnetic field to the die used. Unlike previous work by Sombatsompop (24) using a permanent magnetized die having a constant magnetic flux density (2.16 Tesla) to monitor the changes in the overall swelling of the polystyrene extrudate, this present work investigated the extrudate swell ratio and melt velocity of the PS melt for different positions across the die diameter, and the magnetic flux density could also be varied, as a result of use of non-permanent magnetized (electro-magnetized) die. The results were then compared with those without applying the magnetic field to the die. The effect of the magnetic flux density in the die on the changes in the extrudate swell ratio and velocity profiles of the melt across the die diameter would be of great interest and become a new and unique parameter in altering the extrudate properties (thus final product properties) in real extrusion processes. In addition, the effects of extrusion rate and die temperature on the extrudate swell and velocity profiles were also investigated. The flow properties of the PS melt in the rheometer were also determined in terms of the relationship between shear stress and shear rate, the tests being carried out under the same test conditions at which the extrudate swell ratios were tested.

Experimental

Raw material and experimental apparatus

Since this work continues from our previous work (25), all raw materials (Polystyrene, Styron 656D 267 having a MFR of 7), and the experimental apparatus (constant rate capillary rheometer) remained the same except for the capillary die. This present work was aimed to study the effects of an electro-magnetic field on the radial extrudate swell and velocity profiles of PS melt flowing in a capillary rheometer. Therefore, the capillary die had to be re-designed to accommodate an electro-magnetized die.

Design and manufacture of an electro-magnetized die

The capillary die used in this work was made of mild steel (Grade 1020) and was 64mm long and 5mm in diameter. To make an electro-magnetized die, the die body was wrapped with copper wires in order to apply the electromagnetic field to the die. To prevent an electrical short-circuit, Teflon film was used between the outer die wall and the copper solenoid coil. The dimensions and arrangement of the electro-magnetized die, including the magnetic field profile for this particular die system, are shown in **Figure 1.** According to the relationship between magnetic force (F), charged particle (Q), charged velocity (v) and flux density (B) evidenced by Ulaby (26), the magnetic flux direction was parallel to the flow of the polymer melt in the capillary die. The electro-magnetic field was generated to the die by applying electricity to the copper solenoid coil, the electricity varying from 0A to 10A. It was of some concern that the application of the electro-magnetic field would cause additional heat due to the induction effect across the solenoid coil. This additional heat would then affect the die and melt temperatures during the tests. Therefore, an air-cooling jacket unit was constructed and placed around the electro-magnetized die, the cooling unit being located between the solenoid coil and the die body. The flow-rate of the cooling air was adjusted and calibrated to reduce the additional heat so that the desired temperature was achieved - some experimental results in the discussion section will confirm that no additional heat occurred during the test. A small pressure hole was located between the two die locations to detect the occurrence of entrance pressure drop, the pressure being taken using a photo-conductive light pressure sensor which was originally proposed by Sombatsompop *et al* (27), which was designed and manufactured in our research group. The apparatus temperature was controlled using a DD6 temperature controller.

Measurements of radial extrudate swell and velocity profiles, and flow curves

The radial extrudate swell and velocity profiles of the PS melt were simultaneously measured using the Parallel Co-extrusion Technique (PCT) proposed by the authors (25). The technique was based on a parallel co-extrusion of colored melt-layers into an uncolored melt-stream from the barrel into and out of a capillary die. It should be noted that the extrudate swell ratio and melt velocity profiles at any radial positions across the die interested in this work, were averaged and obtained at the piston displacement range of 85-92mm down the barrel.

The radial extrudate swell ratio values (B_r) were obtained by comparing the thickness of the colored layer of the extrudate outside the die as shown in **Equation 1**, for a given reduced radius (r/R) position in the die. The r/R range of interest used in this work was from 0.0 to 0.86. The accuracy of the radial extrudate swell profile can be verified by averaging the values of the radial extrudate swell (B_r) obtained across die diameter, and comparing with the overall extrudate swell ratio ($B_{overall}$) directly measured at the die exit, the overall extrudate swell ratio being referred to as the ratio of the extrudate radius to the die radius - the detailed description of the technique, including its accuracy, can be found in previous work (25).

$$B_r = \frac{dr_{ext}}{dr_d} \tag{1}$$

The velocity profile measurement in this work was based on monitoring a relatively small and light foreign object flowing along the melt stream. The foreign particles used were natural corn fibers, supplied by V.P. Plastics Products (1993) Co., Ltd (Bangkok Thailand). The particles were relatively small (having an average particle size of 250 μm) and light so that they would not settle out in the melt stream. Only 0.2% of corn particles were added into the extrudate, and this particle concentration did not affect the rheological properties of the melt (25). The preparation procedure for the foreign particles can be found elsewhere (25). The measurements were carried out by recording the times taken for the small particles loaded into the melt layers to travel for a given distance in the die (10mm before the die exit). The experimental apparatus used for die swell measurement was also employed for velocity profile determinations. The accuracy of the velocity profile measurement verified by integrating the radial melt velocity for any given die radius to give the output rate, this being calculated to be approximately 91%.

In this paper, the radial extrudate swell and velocity profiles were measured with and without the application of an electro-magnetic field to the die. Flow properties (apparent viscosity VS wall shear rate) of the PS melt were also simultaneously determined in the capillary rheometer under the same conditions at which the extrudate swell and velocity profiles were measured, whose calculations can be obtained by Sombatsompop and O-Charoen (20).

Experimental variables

All measurements were performed with and without the application of an electro-magnetic field to the capillary die. Different test parameters were studied as listed below.

- Magnetic flux density: In this work, the magnetic flux lines generated in the die had a parallel direction to the flow of the polymer melt. The magnetic flux density of the electro-magnetic field was altered by varying the amount of the electric current from 0, 2, 4, 6, 8 to 10A, these values corresponding to the magnetic flux densities of 0, 0.75, 1.23, 1.56, 1.69 and 1.85 Tesla, respectively. These values were measured using a Tesla-meter at the centre position of the copper coil length which appeared to give the maximum value for any given electric current, this being re-checked using a computer simulation through use of the Finite Element Method Magnetics (FEMM) software (V3.3; 2003; Germany) (the result not shown). It was accepted that the generated magnetic flux density varied from location to location on the experimental rig (barrel and die bodies). Therefore, the extrudate swell results obtained in this work were reported as a function of the maximum magnetic flux density which occurred in the capillary die (at the centre position of the copper coil).

- Wall shear rate: In order to study the effect of the extrusion rate, the piston speed was varied from 0.050×10^{-3}, 0.083×10^{-3} and 0.166×10^{-3} m/s (corresponding to shear rates of 5.1, 8.5 and 17.1 s^{-1}, respectively). Through trial and error experiments, these shear rates were below the critical shear stresses for the onset of PS melt distortions (sharkskin and melt fracture) under the test temperatures, this allowing the extrudate swell measurements to be more accurate.
- Die temperature: To evaluate the effect of temperature on the extrudate swell and velocity profiles and flow properties with and without the application of electro-magnetic field to the die, the temperature of the electro-magnetized die was varied from 200 to 220°C.

Results and Discussion

Overall die swell ratio and a proposed mechanism

Figure 2 shows the captured PS extrudate flowing from the die exit at a wall shear rate of $5.1 s^{-1}$ and a die temperature of 220°C with and without the application of the magnetic field to the die. It can be clearly seen that the swelling level of the extrudate with the magnetic field was much greater than that without the magnetic field. **Figures 3a-3c** show the overall die swell ratio of PS melt as a function of magnetic flux density in the die for three different shear rates at die temperatures of 200, 210 and 220°C, respectively. The general results suggest that the overall die swell for all shear rates increased with increasing magnetic flux density to a maximum value and then started to gradually decrease at higher magnetic flux densities. The magnetic flux density to give the maximum overall die swell varied depending on the shear rate used. This behavior is observed for all die temperatures used **(Figures 3a-3c)**. The changes in the die swell with magnetic flux density can be explained in twofold:

- At 0-1.56T magnetic flux density: The overall die swell ratio was observed to increase with magnetic flux density. The increase in the die swell ratio as a result of the use of the electro-magnetized die was thought to involve the magnetic torque under the electro-magnetic field, and then the change in molecular alignment of the melt during the flow. In this article, we propose that the change in molecular configuration and alignment of the PS resulted from the magnetic torque occurring as a result of the motion of the existing electrons and distortions of the electron clouds within the benzene rings present in the PS molecules. However, this proposed explanation seems to contradict a review by Kimura (23) who stated that the alignment of a polymer under a magnetic field could occur only if the polymer exhibits an anisotropic characteristic, which was the case for crystallizable polymers. For amorphous polymers, the alignment of the molecules under the magnetic field was very difficult. In this work, we propose that although the polystyrene used is an amorphous material, and does not have an anisotropic structure, it was induced by a magnetic field. This was because the die swell ratio of the PS melt in this work was measured *during the flow*. As the melt was flowing from the barrel into the capillary die, the polymer molecules would have to align and extend themselves in order to achieve the flow transition. The alignment and extensibility of the molecular chains allowed the melt to be more anisotropic. As a result, it could be induced by the magnetic torque as it flowed along the electro-magnetized die. According to Kimura (23), the diamagnetic susceptibility of amorphous PS was positive (due to the presence of benzene groups) and the alignment of the molecules tended to be parallel to the magnetic field direction (die length direction, see **Figure 1**). If so, the alignment of the PS melt during the flow in the capillary die would, to some extent, cause an increase in the swell ratio of the extrudate on exiting the die. This view was also supported by Amundson (28) and Chandrasekhar (29). The changes in the molecular orientation and alignment would result in variations of the wall shear stresses and thus the degree of swelling of the melt at the die exit. It should also be noted that the increase in die swell ratio when using the electro-magnetized die confirmed that no additional heat had occurred from the induction effect from the solenoid coil. If heat had occurred, one would expect to observe a decrease in the die swell ratio of the melt (3,30).

- At magnetic flux density greater than ~1.56 Tesla: The overall die swell ratio was observed to decrease with magnetic flux density. The decrease in die swell as a result of the use of the electro-magnetized die after ~1.56T magnetic flux density was probably caused by a change in melt temperature during the flow. As discussed earlier, the magnetic torque acting on the PS melt caused the alignment change of the polymer molecules in a parallel direction of the magnetic field (flow direction in this case). At high magnetic flux densities greater than 1.56T, greater alignment may be expected and this would cause high friction of the flowing melt, and thus additional generated heat. Two possible phenomena would then be expected to cause the reduction of the die swell ratio at this magnetic flux density region: one being a reduction of the magnetic flux density, and the other being an increase in the viscous characteristic. The effect of temperature on the die swell ratio will be shown and discussed later.

It can also be noted that as one would expect, without the magnetic field (at 0 Tesla), the higher the shear rate the greater the overall die swell ratio (3). When applying the electro-magnetic field to the die, the increasing rate of the die swell ratio was greatest at the shear rate of $5.1s^{-1}$ and lowest at the shear rate of $17.1s^{-1}$. This probably involved the residence time of the melt flowing in the die. At low shear rates, the melt would have had more time to be induced by the magnetic torque, and thus caused a higher swelling as it exited the die. When the melt was forced to flow at the higher extrusion rates, it would have had less time to be induced by the magnetic torque, the increase in the die swell ratio being solely caused by the shearing force as normal.

The maximum swelling peak of the melt appeared to shift to higher magnetic flux density with increasing wall shear rates, and the value of the maximum swell ratio decreased with increasing wall shear rates. Both effects were linked to the residence time of the melt as discussed earlier. If the melt had more time (slow extrusion) to absorb the magnetic forces produced from the magnetic field in the die it was likely to swell more. This implies that there was some balancing effect between the shearing force and magnetic torque that acted on the molecules of the melt. From the results reported in this work, the magnetic effect was dominant at low shear rates while the shearing force was more pronounced at high extrusion rates.

When considering the effect of die temperature **(Figures 3a-3c)**, it was found that the changing trend of the swelling curves as a function of magnetic flux density was very similar, but that all the values decreased with increasing die temperature. **Figures 3a-3c** were re-plotted to obtain more understanding of the die temperature effect on the extrudate swell for each wall shear rate, the results being shown in **Figures 4a-4c**. The decrease in the extrudate swell with increasing die temperature was associated with reductions of the melt elasticity and melt viscosity (19,30) and also the magnetic flux density (in turn decreasing the magnetic torque acting on the melt) (31). The reduction in melt viscosity with increasing die temperature is evidenced by **Figure 5.** It can also be observed from **Figures 3 and 4** that the effect of the die temperature on the swell was more significant at low wall shear rates. For a given shear rate, the magnetic field had a greater effect on the melt at lower die temperatures.

Radial die swell and velocity profiles

In this section, a die temperature of 220°C was selected to study the effect of the magnetic field on the changes in radial die swell and velocity profiles of flowing PS melt in the rheometer. **Figures 6a-6c** illustrate the die swell ratio as a function of the reduced radial (r/R) position in the die for different magnetic flux densities at wall shear rates of 5.1, 8.5 and $17.1 s^{-1}$, respectively. It can be seen that, for a given wall shear rate, the radial swell ratio decreased with increasing r/R position, the highest die swell ratio occurring at the duct center and the lowest die swell ratio being near the die wall. The change in the die swell profiles across the die diameter had already been discussed, and also in previous work (25), in connection with the change in melt velocity profiles. It was noticeable that the die swell profiles changed continuously with changing magnetic flux density. The die swell changes appeared more pronounced at a low shear rate ($5.1 s^{-1}$), which corresponded with the overall die swell as discussed earlier in **Figures 3 and 4**. There were two regions exhibiting obvious changes in die swell ratio across the die diameter, one being around the centre of the duct and the other being around r/R of 0.65-0.85. At the centre position, the die swell ratio appeared to increase with increasing magnetic flux density up to ~1.56T, and to decrease for higher magnetic flux densities. At r/R positions of 0.65-0.85, the die swell ratio decreased, indicating that the melt layers flowing around these positions had contracted on exiting the die. This implies that the changes in the overall die swell ratio discussed in **Figures 3a-3c** were more affected by the swelling of the melt around the centre of the die.

The changes in the die swell profiles of the melt due to the application of the electro-magnetic field to the die are best explained using the velocity profiles as a function of the reduced radial positions that were simultaneously measured, for different shear rates and magnetic flux densities at a die temperature of 220°C. The results are shown in **Figures 7a-7c**. It can be noticed that the general trend of the radial velocity profiles was very similar to that for the die swell profiles. The highest melt velocity was found at the center of the die, the melt velocity decreasing with increasing r/R value. Previous work (25) suggested that the shape of the radial die swell profiles was associated with the development of velocity profiles of the melt flowing in the die and the equalization of the velocity profiles at the die exit. A sudden reduction in melt velocity at the center flowing in the die to the die exit caused the melt to flow radially and led to an expansion of the melt in radial direction, this suppressing the dimension change of the melt near the die wall. Similar to the die swell profiles, the melt velocity at the centre increased with increasing magnetic flux density up to a maximum and then decreased at higher magnetic flux densities, whereas that near the die wall progressively decreased with increasing magnetic flux density. The reasons for the changes are similar to those mentioned in the overall die swell measurement. Again, similar to the die swell, the effect of the magnetic field on the velocity profile was more pronounced when using a low wall shear rate ($5.1 s^{-1}$).

It is clear at this point that the application of the electro-magnetic field to the die caused the changes in the radial velocity profiles of PS melt. It is proposed that there were two mechanical forces acting on the polymer molecules during the flow in the electro-magnetized capillary die, one being magnetic torque and the other being shearing force generated un-evenly across the flow channel. To gain more in-depth understanding, the shear rate profiles of the melt across the die diameter were utilized, as illustrated in **Figures 8a-8c**, shear rate data being calculated from the melt velocity profiles in **Figures 7a-7c**. Since the actual velocity profile of the melt in this work was not parabolic in nature, the maximum shear rate was not found near the die wall, but instead, it appeared around r/R of 0.5-0.7. It seems that the magnetic torque would have had a considerable effect on the melt velocity at the centre of the duct since the shearing force in this region was relatively low (theoretically assumed zero). Near the die wall, the shear rate became high and this resulted in a depression of the magnetic torque effect. In **Figure 7**, the melt at the centre of the duct could be more aligned and oriented under the magnetic field, and this alignment possibly facilitated the melt to flow at higher velocity at the centre of the duct. This may be the reason why the swelling ratio of the melt at the centre of the duct increased with the magnetic flux density. As a consequence, in order to maintain the volumetric flow rate, the velocity of the melt near the die wall had to reduce, and thus the die swell ratio decreased.

Conclusion

The effect of an electro-magnetic field on the die swell and velocity profile behavior of PS melt in a capillary rheometer was investigated. The following points were noted:

- For overall die swell measurements, the die swell ratio of the PS melt obtained with the magnetic field was much greater than that without the magnetic field. The overall die swell for all shear rates increased with increasing magnetic flux density to a maximum value (around 1.56T) and then decreased at higher magnetic flux densities. The maximum swelling peak of the melt appeared to shift to a higher magnetic flux density, and the value of the maximum swell decreased with increasing wall shear rate and die temperature. The change in die swell ratio using the die with magnetic field was explained in terms of the magnetic torque and the shearing force, acting on the melt *during the flow*. The effect of magnetic torque on the die swell ratio of the melt was more pronounced when extruding the melt at low shear rates and low die temperatures.
- For radial die swell and velocity profiles, the radial swell ratio for a given shear rate decreased with increasing r/R position. The changes in die swell profiles across the die diameter was associated with the development of the melt velocity profiles during the flow, both changing continuously by changing the magnetic flux density and shear rate. The die swell and velocity profile changes appeared more pronounced at low shear rate. There were two regions where the changes in the die swell ratio across the die diameter were obvious, one around the duct centre and the other around r/R of 0.65-0.85. The magnetic torque had a considerably greater effect on the melt at the centre of the duct, causing increases in melt velocity and die swell.
- The changes in the overall die swell ratio of PS melt flowing in an electro-magnetized die were affected more by the swell ratio of the melt layers around the centre of the die than that near the die wall.

Acknowledgments

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List of Figures

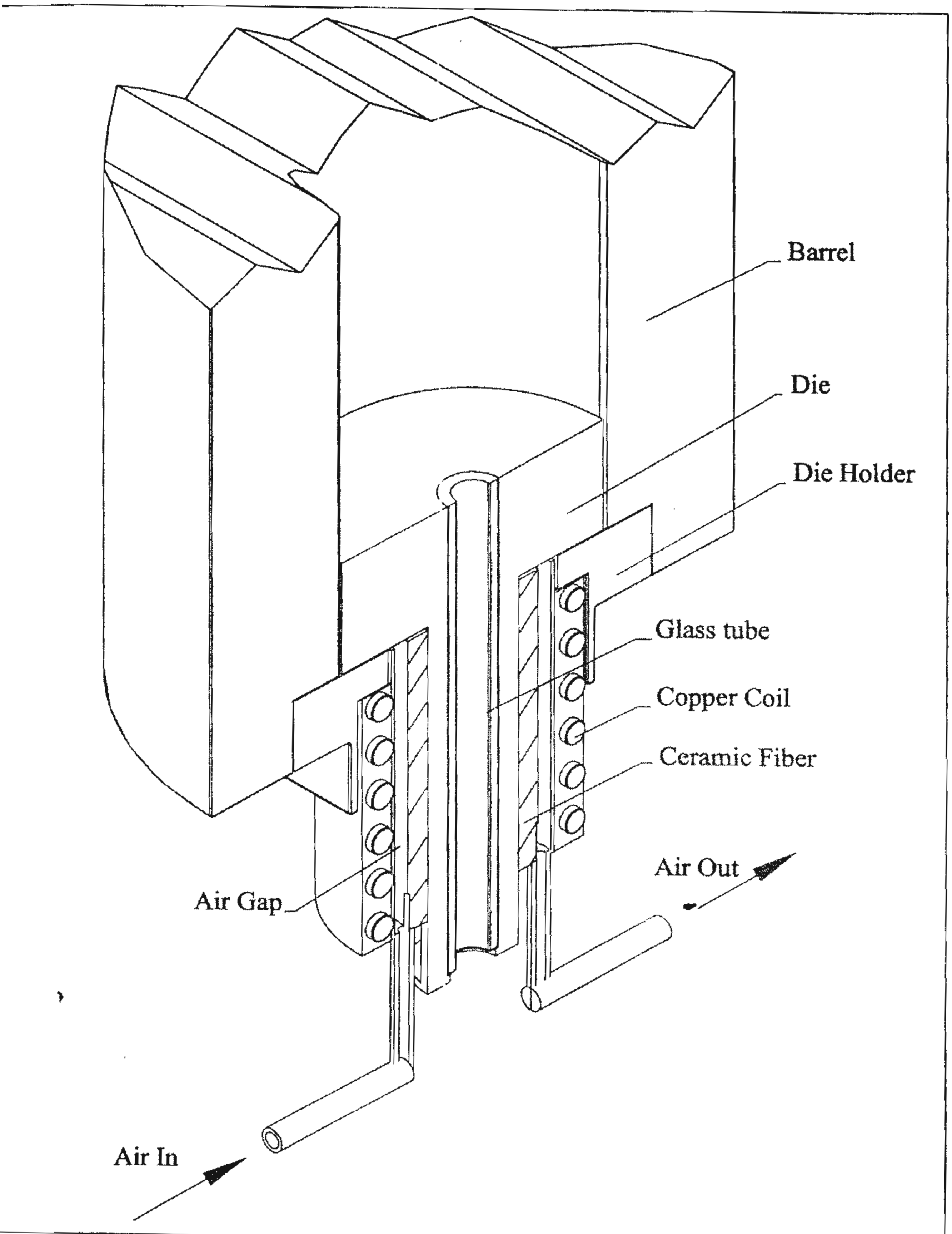

Barrel
Die
Die Holder
Glass tube
Copper Coil
Ceramic Fiber
Air Out
Air Gap
Air In

Figure 1

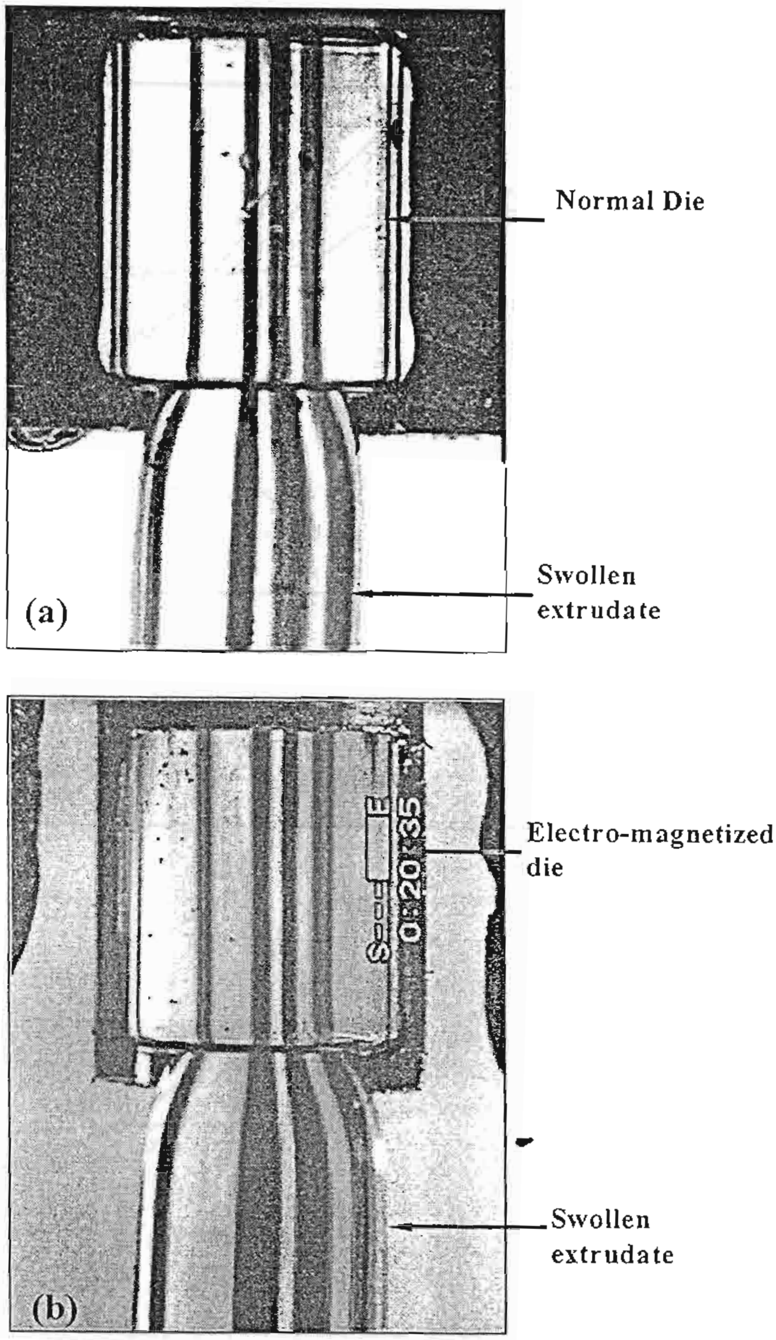

Normal Die
Swollen extrudate
(a)
Electro-magnetized die
0:20:35
Swollen extrudate
(b)

Figure 2

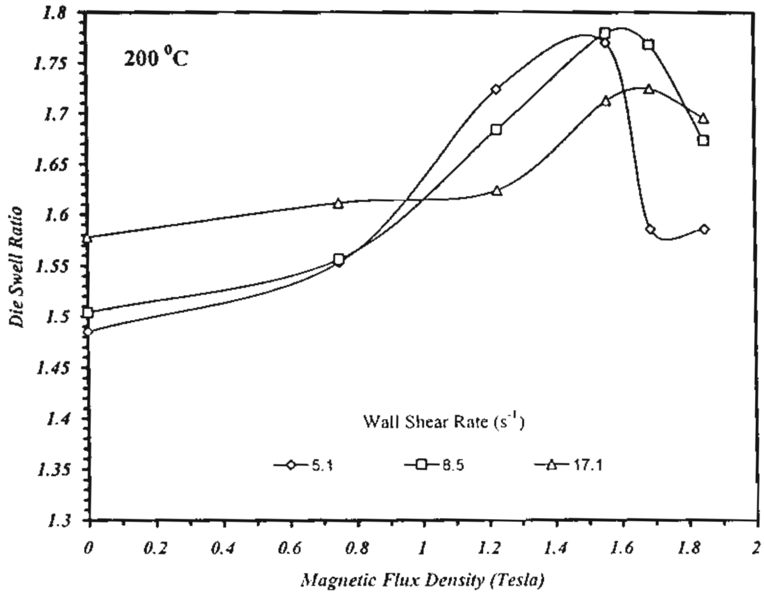

200 °C
Die Swell Ratio
Magnetic Flux Density (Tesla)
Wall Shear Rate (s^{-1})
5.1
8.5
17.1

Fig.3a

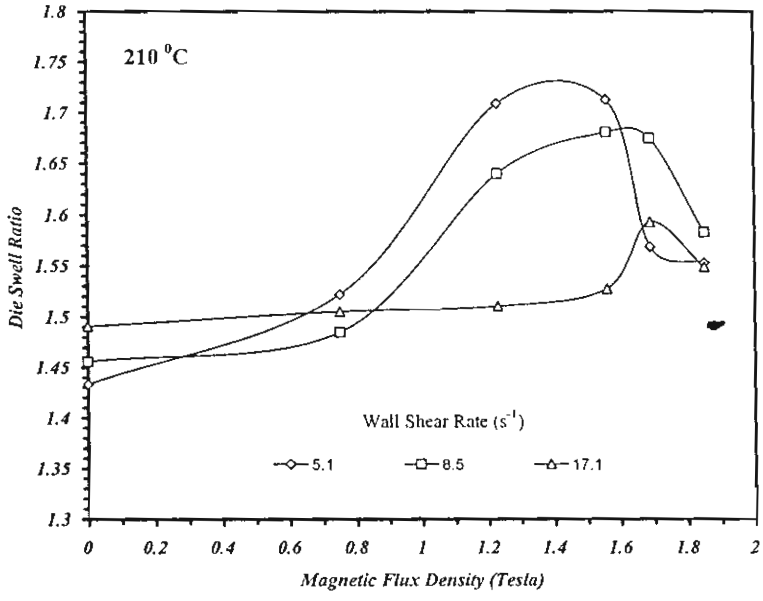

210 °C
Die Swell Ratio
Magnetic Flux Density (Tesla)
Wall Shear Rate (s^{-1})
5.1
8.5
17.1

Fig.3b

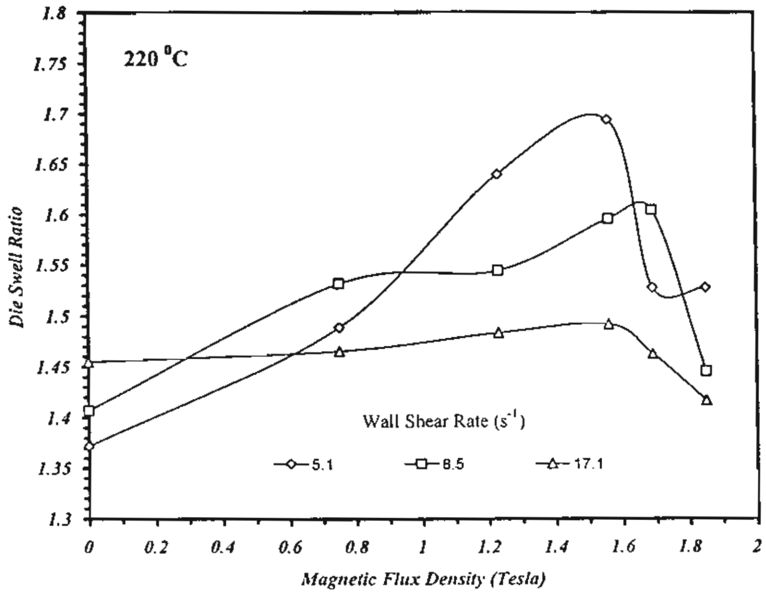
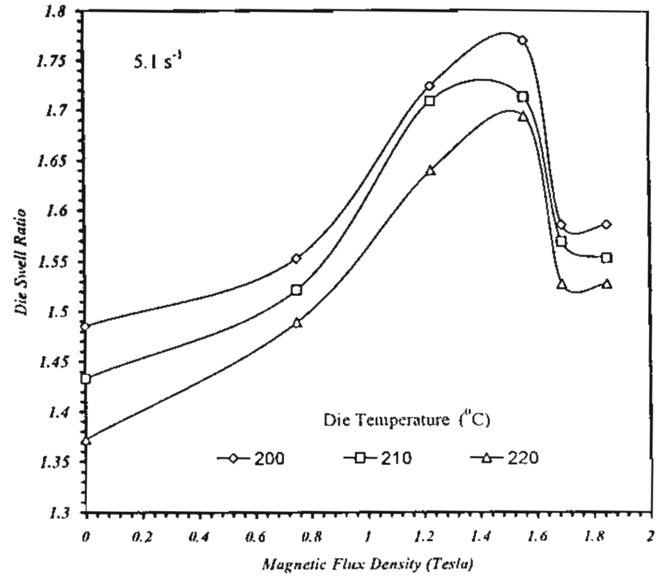


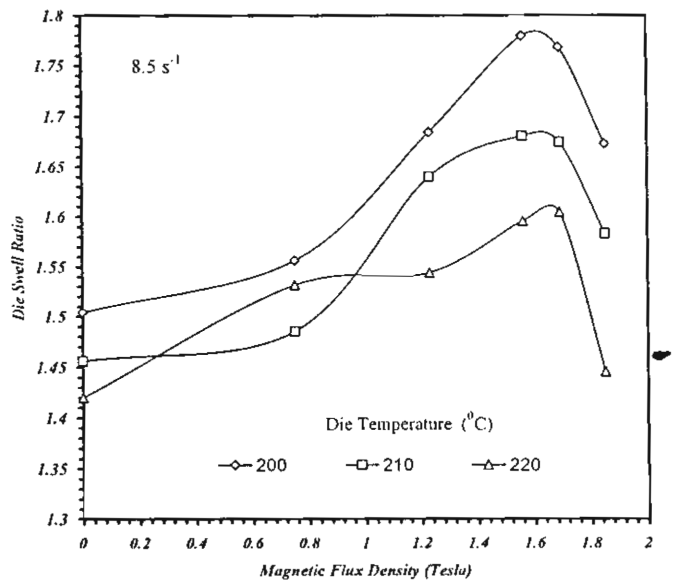
Fig.3c

Figure 3



5.1 s-1
Die Swell Ratio
Magnetic Flux Density (Tesla)
Die Temperature (°C)
200
210
220
1.8
1.75
1.7
1.65
1.6
1.55
1.5
1.45
1.4
1.35
1.3
0
0.2
0.4
0.6
0.8
1
1.2
1.4
1.6
1.8
2

Fig.4a



8.5 s-1
Die Swell Ratio
Magnetic Flux Density (Tesla)
Die Temperature (°C)
200
210
220
1.8
1.75
1.7
1.65
1.6
1.55
1.5
1.45
1.4
1.35
1.3
0
0.2
0.4
0.6
0.8
1
1.2
1.4
1.6
1.8
2

Fig.4b

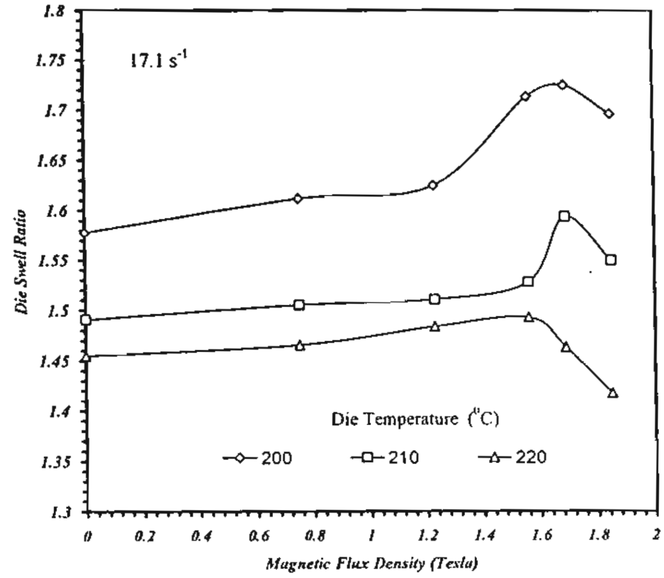
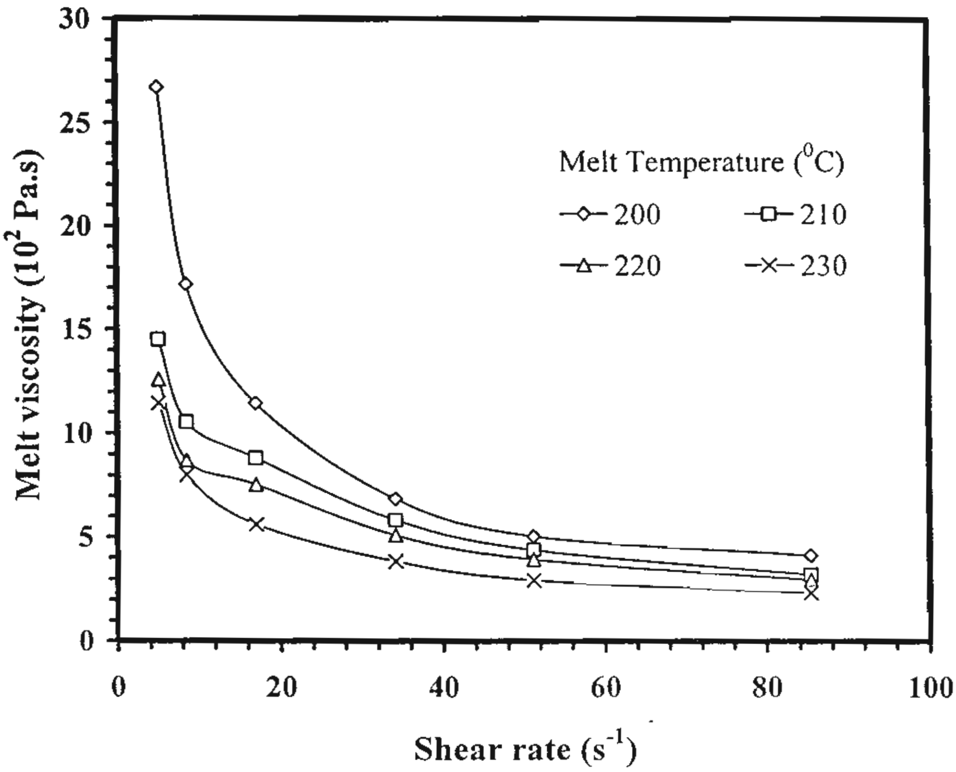


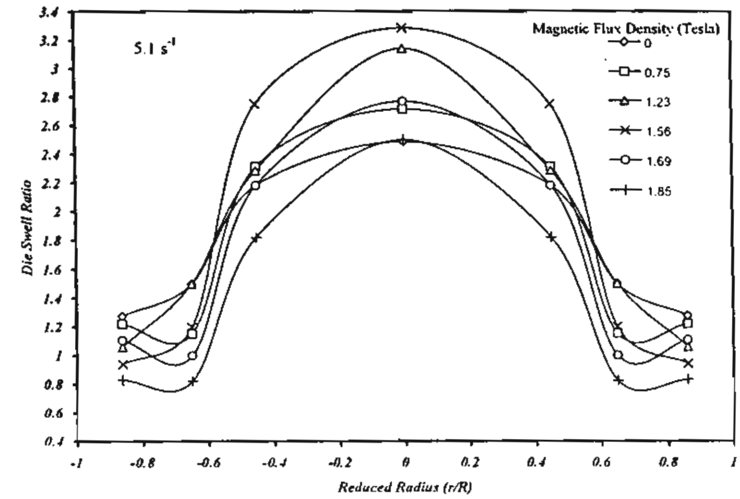
Fig.4c

Figure 4



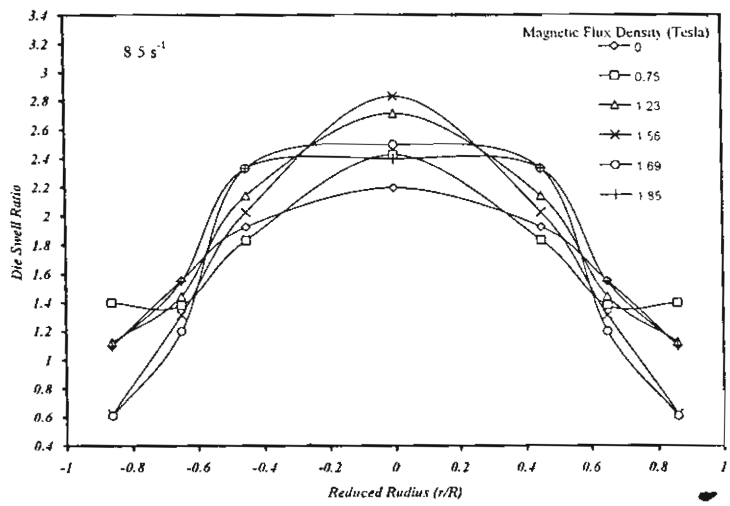
Melt viscosity (10^2 Pa.s)
Shear rate (s^{-1})
Melt Temperature (^{0}C)
200
210
220
230
0
5
10
15
20
25
30
0
20
40
60
80
100

Figure 5



5.1 s^-1
Magnetic Flux Density (Tesla)
0
0.75
1.23
1.56
1.69
1.85
Die Swell Ratio
Reduced Radius (r/R)

Fig.6a



8.5 s^-1
Magnetic Flux Density (Tesla)
0
0.75
1.23
1.56
1.69
1.85
Die Swell Ratio
Reduced Radius (r/R)

Fig.6b

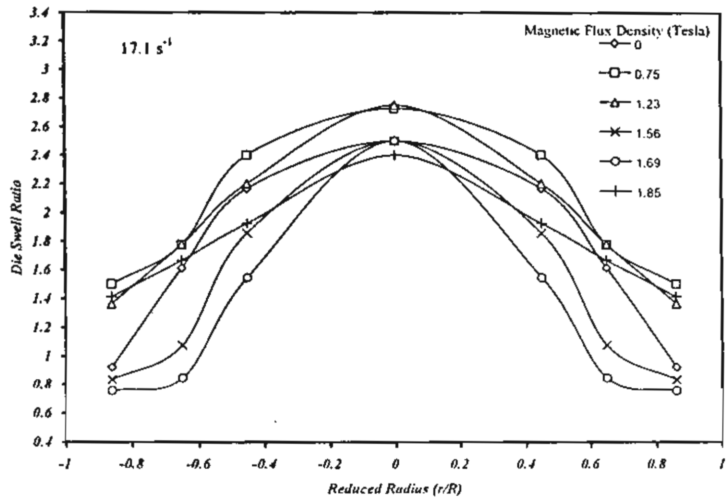


Fig.6c

Figure 6

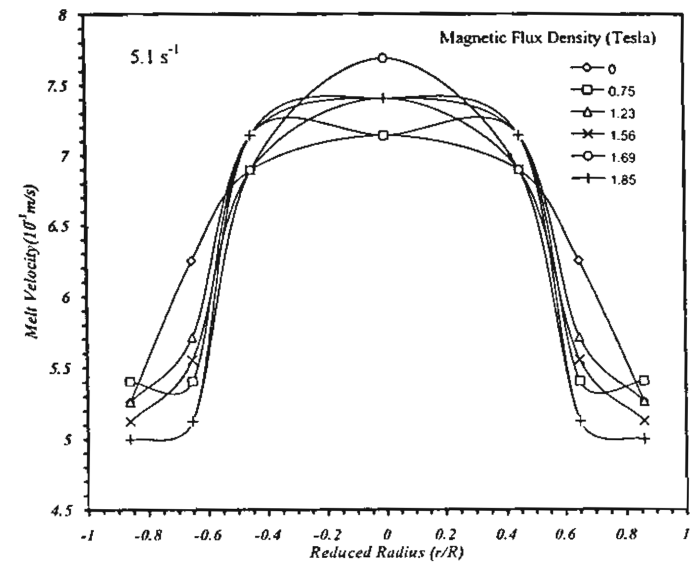

5.1 s^-1
Magnetic Flux Density (Tesla)
0
0.75
1.23
1.56
1.69
1.85
Melt Velocity(10^-3 m/s)
8
7.5
7
6.5
6
5.5
5
4.5
-1
-0.8
-0.6
-0.4
-0.2
0
0.2
0.4
0.6
0.8
1
Reduced Radius (r/R)

Fig.7a

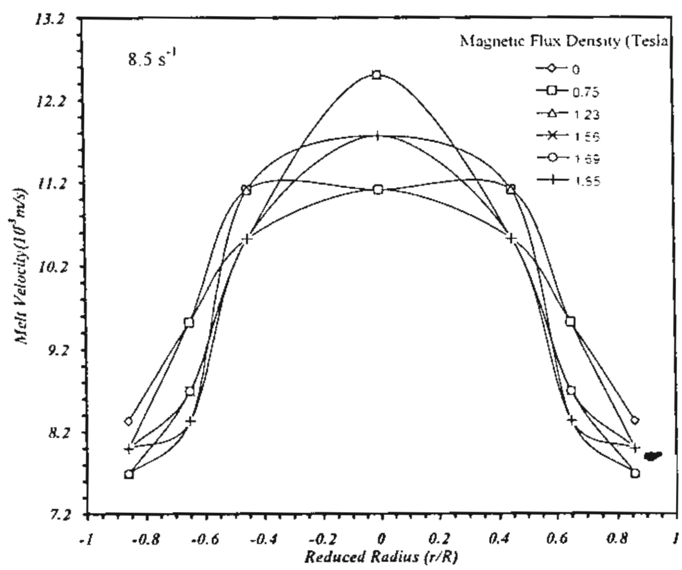

8.5 s^-1
Magnetic Flux Density (Tesla
0
0.75
1.23
1.56
1.69
1.85
Melt Velocity(10^-3 m/s)
13.2
12.2
11.2
10.2
9.2
8.2
7.2
-1
-0.8
-0.6
-0.4
-0.2
0
0.2
0.4
0.6
0.8
1
Reduced Radius (r/R)

Fig.7b

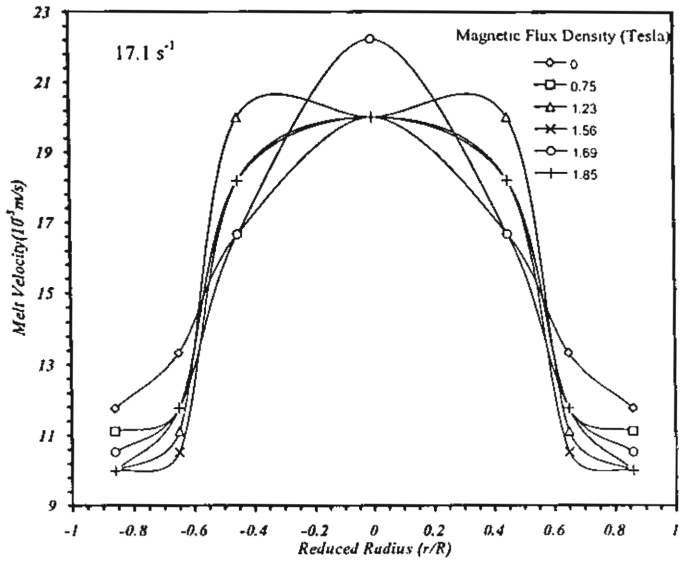
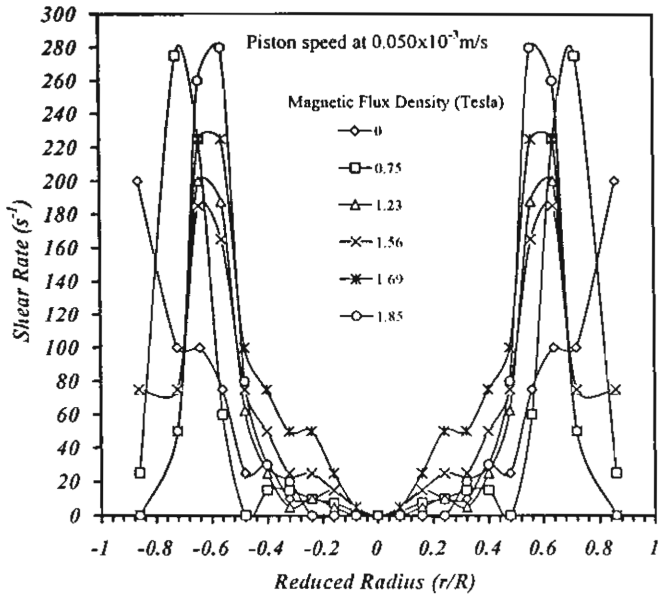


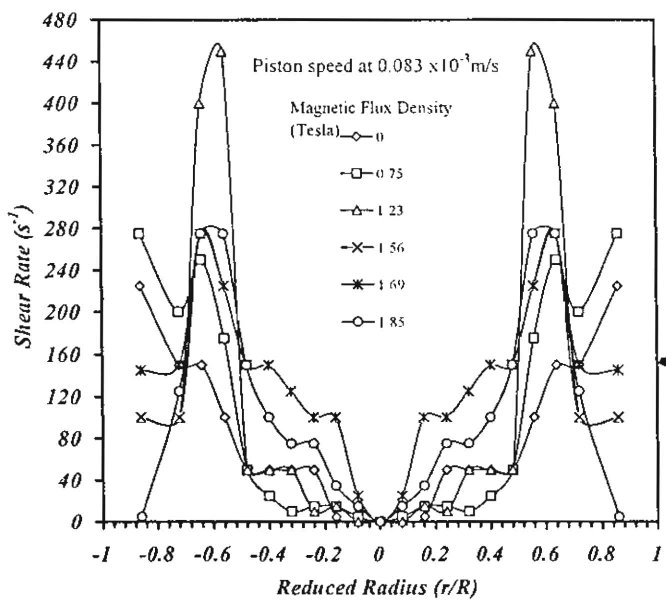
Fig.7c

Figure 7



Piston speed at 0.050x10⁻³m/s
Magnetic Flux Density (Tesla)
0
0.75
1.23
1.56
1.69
1.85
Shear Rate (s⁻¹)
Reduced Radius (r/R)

Fig.8a



Piston speed at 0.083 x10⁻³m/s
Magnetic Flux Density (Tesla)
0
0.75
1.23
1.56
1.69
1.85
Shear Rate (s⁻¹)
Reduced Radius (r/R)

Fig.8b

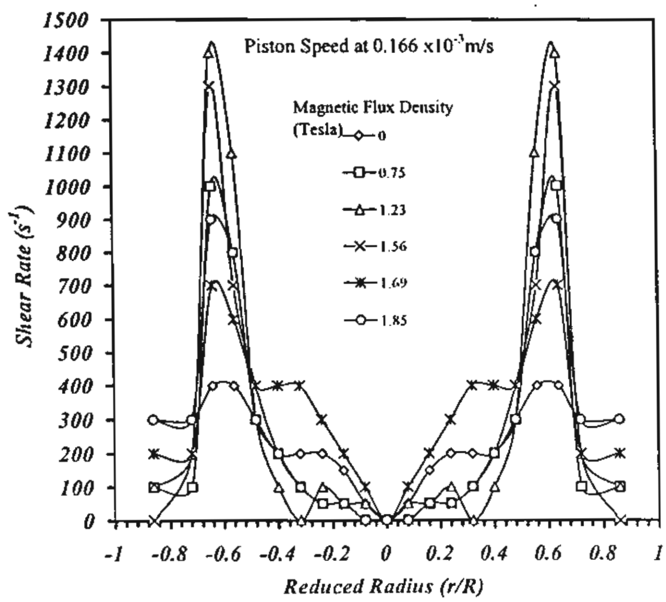


Fig.8c

Figure 8