

Comments on Some Assumptions Made for the Determination of Polymer Melt Flow Properties

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Some assumptions generally made for the derivation of the rheological equations (apparent shear stress, apparent shear rate and apparent melt viscosity) were investigated using recently published experimental evidence. These assumptions include: no slip at the duct wall, steady and constant flow patterns of the melt along the duct, isothermal flow, and incompressibility of polymer melts. A number of implications were found regarding these assumptions.

Key words: Rheological properties, polymer melt flow, shear heating, non-Newtonian fluids, extrusion.

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ข้อเสนอแนะสำหรับสมมุติฐานที่ใช้ในการวัดคุณสมบัติทางการไหล ของพอลิเมอร์หลอมเหลว

ณรงค์ฤทธิ์ สมบัติสมภพ และ อาลัน วูด (2541)
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การตั้งสมมุติฐานบางส่วนเพื่อใช้ในการศึกษาและแจกแจงคุณสมบัติทางการไหลของของไหลพอลิเมอร์ (เช่น ความเค้นแรงเฉือน อัตราความเฉือน และ ค่าความหนืดของของไหล) ได้ถูกวิเคราะห์และตรวจสอบ โดยอาศัยหลักฐานการวิจัยค้นคว้าในทางปฏิบัติที่ถูกดำเนินการโดยสมมุติฐานดังกล่าวคือ ไม่มีการไหลอย่างฉับพลันของของไหลที่ผนังท่อ รูปแบบการไหลคงที่และแน่นอนตลอดความยาวของท่อ อุณหภูมิของของไหลขณะที่กำลังไหลคงที่ และของไหลไม่สามารถถูกอัดด้วยความดันได้ จากการวิเคราะห์พบว่า บางสมมุติฐานไม่สอดคล้องกับพฤติกรรมที่เกิดขึ้นจริงในทางปฏิบัติ

คำสำคัญ คุณสมบัติทางการไหล การไหลของพอลิเมอร์ ความร้อนอันเนื่องมาจากการเสียดสีของพอลิเมอร์ ของไหล non-Newtonian การอัดรีดพลาสติก

INTRODUCTION

The most common method used for the measurement of the flow characteristics of polymer melts is that of the capillary rheometer. This technique is widely accepted to give accurate and reliable results. The capillary rheometer consists of a heated barrel at the

bottom of which is fitted a small capillary die. These devices are of two general designs, these being constant shear stress devices and constant shear rate devices.^(1,2) The rheological properties of a polymer melt are important in processing, but they are complicated by a number of factors such as flow rate, pressure,

and temperature. The rheological characteristics of polymer melts are usually expressed in the form of shear stress and shear rate, these two parameters being used for the determination of fluid viscosity. Polymer melts in general exhibit pseudoplastic non-Newtonian flow behaviour, in which the shear stress is not proportional to the shear rate.^(2,3) In practice, when determining the flow characteristics of a polymer melt using a rheometer (or viscometer), the measurements taken are the pressure drop along the capillary (P), die length (L), die radius (R), and volumetric flow rate (Q). The apparent shear stress (τ_w) and apparent shear rate (γ_w) at the wall are usually used and they can be calculated as follows: ^(1,2)

$$\tau_w = \frac{RP}{2L} \quad (1)$$

$$\gamma_w = \frac{(3n+1)4Q}{4n\pi R^3} \quad (2)$$

where n is the power law index.

The viscosity (η) of polymer melts is practically expressed in the form: ^(4,5)

$$\eta = \frac{\tau_w}{\gamma_w} \quad (3)$$

There are a number of assumptions generally made in the derivation of the above equations, including the following: ^(4,6)

- a) There is no slip at the duct wall.
- b) The flow patterns of the melt are steady and constant along the duct.
- c) The flow is isothermal.
- d) The melt is incompressible.

From some of the published results and research work by the authors ^(1-3, 7-13), it is felt that these assumptions are not true in practice and they should be discussed and clarified with some plausible reasons. Therefore, in this paper, we discuss and make some relevant comments on the above assumptions in detail.

ASSUMPTION 1 : THERE IS NO SLIP AT THE DUCT WALL

This assumption means that the velocity of the fluid particles at the duct wall is zero. If a sudden change in melt velocity (a slip) occurred at the wall, it would be expected that the calculated values of shear rate would be in error and the flow curves plotted would not superimpose as they should. However, this is not found in practice. Generally, a no-slip condition can be assumed for many theoretical derivations. This assumption can be discussed using evidence^(3,5,7,8,14,15) regarding the cause of melt fracture. Many authors have stated that melt fracture is caused by slippage of the melt fracture at the die wall, whereas recent evidence by Sombatsompop and Wood found that the fracture of the extrudate was not caused by such melt slippage at the wall, but resulted from a sudden change in melt velocity profiles across the flow channel.^(7,8) They used many lines of evidence to support their statement, such as the lack of a shift in flow curves at the onset of melt fracture, no plug flow occurrence, and the observation of the layered rubber compound extruded in a capillary rheometer before and after the onset of melt fracture.⁽³⁾ The evidence indicated clearly that no slip occurred either before and after the onset of melt fracture. As a consequence, this assumption was found to be true.

Figure 1. Flow patterns of natural rubber compounds in the barrel of a capillary rheometer at various piston displacements (from right to left: 40 mm, 80 mm, and 120 mm down the barrel).

ASSUMPTION 2: FLOW PATTERNS ARE STEADY AND CONSTANT ALONG THE FLOW CHANNEL

The general theory assumes that a fully developed flow pattern exists at the entrance of the tube and that this still persists at the exit of the tube. Due to the inter-relationship between the viscosity of a fluid and the formation of

boundary layers,⁽¹⁶⁾ it seems likely that the boundary layers formed on opposing sides of the die will join a short distance down the die. This assumption has been found to be untrue, this statement being based on the results of the determinations of the flow patterns or the velocity profiles occurring in the barrels of a capillary rheometer and an injection moulding machine.^(1-3, 9-11) Figure 1 shows flow patterns of

natural rubber compounds occurring in the barrel of a capillary rheometer using the coloured layer method^(1-3, 9) at different piston displacements. It can be seen that the flow patterns of the polymer melts at a fixed point in

the barrels changed continuously with piston displacement. In addition, for a given time or piston position the flow patterns at various points along the barrel length were different.



Figure 2. Velocity profiles of a low-density polyethylene melt in the barrel of an injection moulding machine at different screw displacements as indicated.
(9-11)

In the case of velocity profiles in the barrel of an injection moulding machine carried out using a novel technique by Sombatsompop and Wood,⁽⁹⁻¹¹⁾ an example of velocity profiles at a fixed point along the barrel as a function of screw displacement is given in Figure 2. Similar behaviour is seen, with velocity profiles

changing continuously with screw displacement.

ASSUMPTION 3: THE FLOW IS ISOTHERMAL

When measuring the rheological properties of polymer melts using a rheometer,

this normally having a relatively small diameter die (less than 1.5 mm diameter), the melt temperature in the die is assumed to be

uniform within the limits of experimental error, with the melt temperature changing on the order of 4-5°C.⁽¹²⁾ The small change in the melt

Figure 3. Relationship of Melt Temperature and Pressure upon compression and Decompression in the Barrel (near the nozzle) of Injection Moulding using Polyethylene Melt (LDPE, LD100BW) at 190 °C

temperature can be neglected and therefore this assumption, in this case, is confirmed to be true. However, the results of the melt temperature profiles in the barrel of the injection moulding having the diameter of 35 mm and using a die of 10 mm diameter were not observed to be uniform across the duct.⁽¹³⁾ In this case, the melt temperature increased by around 10-15°C. This change in melt temperature is relatively large and cannot be neglected. The changing melt temperature in the dies of the rheometer and the injection moulding machine was due to shear heating and conduction effects. In the case of the capillary rheometer, which uses a small die, the effect of shear heating during the flow of the

polymer melts appeared to be less significant than that of the heat conduction through the die wall. Therefore, a smaller change in the melt temperature was obtained. On the other hand, the effect of shear heating became the dominant factor in the case of the injection moulding, thus leading to the larger increase in melt temperature. As a consequence of this, the isothermal assumption is not always true in polymer processing.

ASSUMPTION 4: THE FLUID IS INCOMPRESSIBLE

At high pressure, the compressibility of the polymer can be very important, as in the case of an injection moulding process. The

effect of melt pressure on the melt flow properties was discussed using the present results shown in Figure 3. The work was carried out by measuring the average radial melt temperature near the nozzle in the barrel of an injection moulding machine (Negri Bossi 60NB Italy) as a function of screw position and pressure applied. All the measurements were recorded using a data logger and a computer. Figure 3 shows the values of melt temperature and pressure occurring during the compression and decompression, and the positions of the screw. This result indicates that the sudden retraction of the screw was due to the decompression of the melt. Therefore, it should be noted that polymer melts are *compressible in practice*. In terms of melt temperature, it can be seen that the melt temperature increased with increasing melt pressure with a corresponding decrease as the pressure was removed. This variation in melt temperature would be an additional reason for the assumption of isothermal flow to be error with the melt temperature being non-uniform. In addition, from work⁽¹⁷⁾ concerning the relationship between temperature, pressure, and density of polymer melts, complicated phenomena were observed when determining properties of polymer melts (such as the density). It has been found that pressure and temperature affected density of the polymer melts, the density increasing with pressure and decreasing with temperature. In conclusion, from experimental evidence it has been shown that in practice:

- a) There was no slip at the duct wall.
- b) The flow patterns of the melt were not steady and constant along the duct.
- c) The flow was isothermal in the case where the size of the flow channel was less than 1.5 mm.
- d) The melt was compressible.

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