# Studies of Converging Flows of Viscoelastic Polymeric Melts. II. Velocity Measurements in the Entrance Region of a Sharp-Edged Slit Die

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## **Synopsis**

An experimental study has been carried out to measure velocities of viscoelastic polymeric melts flowing into a sharp-edged slit die and in the fully developed region of the slit die. For the velocity measurement, the technique of streak photography was used, in which photographs are made of the movement of tracer particles suspended in a molten polymer. Materials investigated were polypropylene, high-density polyethylene, and polystyrene. From the photographs, local velocities in the entrance region were determined by measuring the streak length, a reference length, and the exposure time of the camera. A comparison was also made of the experimentally determined velocity profiles in the fully developed region with the theoretically predicted ones, showing a reasonable agreement between the two.

## INTRODUCTION

Since Reynolds first injected dye into a Newtonian fluid and discovered that three different flow regimes exist (laminar, transition, and turbulent), investigators have tried to develop accurate methods of obtaining point velocities and flow directions.

In their review paper on velocity-measuring probes, Mandersloot et al.<sup>1</sup> discuss the various types of pitot tube probes that have been evolved. The probe works on the principle that the velocity along a streamline can be related to the static pressure and the impact pressure. Another technique that has been utilized is that of the hot-wire anemometer. This type of velocity probe measures the rate of heat transfer. This measurement is then converted to point velocity.

However, the hot-wire anemometer and the pitot tube suffer from the fact that the measuring device obstructs the flow path and thus does not accurately portray the velocity profiles. This phenomenon becomes most severe as more complicated fluids are investigated (i.e., viscoelastic materials). Consequently, investigators tried optical techniques to measure the point velocities. The one that has had the most success is that of observing very small particles with a microscope as they flow with the fluid. While this technique does introduce a foreign body into the fluid,

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it is felt that no detectable disturbance will occur. To ensure this condition, the particle size must be very small compared to the immediate geometry and the particle concentration must be very low in the fluid. This method also assumes that the particle is flowing at the same velocity as the suspending fluid itself.

Some modifications of the above-mentioned method for point velocity determinations have appeared in the recent literature. Berman and Santos<sup>2</sup> describe a method to obtain velocity measurements by the detection of the Doppler shift of laser light that is scattered from a moving object. Popovich and Hummel<sup>3</sup> and Goldish et al.<sup>4</sup> have developed a method called flash photolysis. In this technique, photochromic dye is dissolved in the test fluid and irradiated with ultraviolet light to produce a colored trace. This trace has been related quantitatively, according to Frantisak et al.,<sup>5</sup> to the velocity profile in both laminar and turbulent flow. While this last method has been successful, it is limited to systems in which the dye will dissolve in the fluid. In general, the above-mentioned modifications of particle observation have very interesting possibilities. However, to date they have only been tested with Newtonian fluids. The possibility of extending these methods to polymer melts is not known at this time.

The technique of using particle observation for point velocity measurements is not new. However, it has only been recently applied to studies involving non-Newtonian viscoelastic fluids and two-dimensional flow. Atkinson et al.<sup>6</sup> have used the technique of streak photography to obtain point velocities during profile development. In streak photography, the particles are illuminated from the side, viewed against a dark background, and photographed. The tracer particles to be used must have a high reflectance coefficient, a density only slightly greater than the test fluid, and negligible settling properties. The illumination is provided by a discontinuous source of light. Thus, the path of each moving particle observed is a discontinuous streak. From a knowledge of the exposure time and the measured streak distance, the velocities can be obtained. Uebler<sup>7</sup> has used a very similar technique; however, he was measuring velocity profiles in the entrance region for a viscoelastic polymeric solution. More recently, Allen and Schowalter<sup>3</sup> employed the technique of streak photography to measure fully developed velocity profiles of a viscoelastic polymer solution flowing through a circular tube, and Murthy and Boger,<sup>9</sup> to measure developing velocity profiles of inelastic polymeric solutions.

Galt and Maxwell<sup>10</sup> were the first to obtain velocity information in polymer melts. The technique they employed was very similar to that used by Uebler; however, they only investigated flow through a tube at positions where entrance effects were not important. The main conclusion of their study was that the relative velocity at the wall need not be zero. They claim that in steady laminar flow polymer melts exhibit a high degree of stick-slip behavior at the wall and that this phenomenon is caused by melt elasticity. Using a rectangular slit, den Otter et al.<sup>11</sup> measured the velocity profiles in the fully developed region (i.e., downstream in the slit) of the flow of various polymer melts. The velocities were measured by observing the flow particles in the moving melt. The main experimental difference between this work and that done by Galt and Maxwell was that here no particles were added to the polymer. den Otter observed that every grade of commercially available polymer he examined had enough gel or dust particles of suitable size to be seen with a microscope. He noted that the dust particles were of the order of 0.005 to 0.02 mm in diameter, compared to Galt and Maxwell's added particles of 0.05 mm diameter. The other difference was that Galt and Maxwell obtained photographs of their measurements, while den Otter timed the particles as they traveled a fixed distance.

The results of den Otter's work contradicted those of Galt and Maxwell, in that his measurements indicated a zero velocity at the wall at flow rates just below and just above the onset of melt fracture. den Otter suspects that the slip-stick phenomena Galt and Maxwell postulated might have been due to the size of the particles employed. den Otter noticed that the velocity near the wall was smallest when the smallest particles were observed near the wall. He also pointed out that the large particles moved with varying speed during melt fracture, while the small particles moved at constant speed. den Otter also compared his measurements with the profile calculated by assuming an infinite slit with a power law fluid, and showed that the power law fluid makes the profile too flat at the centerline but gives good agreement at the walls.

More recently, Bartos and Holomek<sup>12</sup> also measured velocity profiles of polymer melts flowing through capillary tubes, using tracer particles. These authors found that velocities at the tube wall are zero under stable flow situations, but they fluctuate once the onset of melt fracture starts to occur. In this context, it seems worth mentioning a recent work by Han and Lamonte,<sup>13</sup> who also investigated the problems associated with melt flow instability by measuring normal stresses at the tube wall. Han and Lamonte found that wall normal stresses, which otherwise give constant values under stable flow situations, start to fluctuate once the onset of melt Since the wall normal stress is directly related to the fracture occurs. fluid velocity at the tube wall, it can be said that the two entirely different experimental techniques, one that employed tracer particles for velocity measurement and another that measured wall normal stresses, give identical results, in that the wall velocities of polymer melts are zero under stable flow situations and fluctuate under unstable flow situations.

## Experimental

The apparatus used for the velocity measurements was essentially the same as that used for the flow birefringence experiment (that is, the same flow system and test cells) described in a previous paper, part I of this series.<sup>14</sup>

In the velocity measurements, however, tracer particles were added to the polymer melt and streak photographs of the particles were taken at various axial positions in the test cell, that is, in the entrance region and also in the fully developed region. From these pictures, the velocities could be determined by knowing the streak length, a reference length (i.e., the slit width), and the exposure time.

A variety of tracer particles and lighting techniques were tried. Initially it was felt that the dust particles present in the commercial polymer would be large enough to be photographed. However, due to limited magnification power of our photographic equipment, observation of dust particles was not possible. Another material tested, carbon black, was either too small to be seen or was too hard to properly disperse in the polymer melt. It was found eventually that a certain copper powder provided the tracer particles that gave the best results. Supplied by the United States Bronze Powder Co. of New Jersey, the material, #C-118, is an atomized, spherical, 99.5% pure copper powder. The particle size distribution was 70% 44 microns and 30% 149 microns. Since the polymer melts were quite viscous and the settling velocities of the particle so small, gravity and inertial effects were ignored.

The experimental procedure was very similar to that used in the birefringence experiments. The camera was mounted at right angles to the test cell. To minimize error due to vibrations, the camera, the test cell, and the feed system were interconnected. The glass windows opposite the camera were blacked out so that no light could pass through the cell. A light source was placed at an angle of approximately 60 degrees to the front glass window and on the camera side of the cell. The particles were blended with the polymer in the hopper of the extruder. When the particles suspended in a flowing molten polymer passed through the entrance region of the test cell, the light illuminated the copper spheres and their image was captured by the camera. In order to ensure that the walls of the cell would not affect the velocity measurements, the camera was focused at the centerline of the cell.

Polymers used for the velocity measurements were the same as those used for the stress birefringence measurements, namely, polypropylene, polystyrene, and high-density polyethylene. For each of these polymers used, measurements were taken at different velocities and different melt temperatures. However, in no cases were extrusion conditions so chosen that flow instabilities could have been observed.

## **RESULTS AND DISCUSSION**

### **Interpretation of Streak Photographs**

Figure 1 gives representative streak photographs of tracer particles in the entrance region, namely, in the reservoir section at the slit die entrance. These pictures were taken with a 1-second camera exposure to the illuminating particles, which were suspended in a flowing molten polymer.



FIG 1. Representative streak photographs of tracer particles in the entrance region: (a) polypropylene; (b) high-density polyethylene.

For a given flow rate of melt, pictures were taken both in the entrance region and in the fully developed region (that is, downstream in the slit). This was done so that, when pictures were developed, a sufficiently large number of streaklines would be found over the entire flow field concerned. Since, in a given picture, all the steaklines were obtained with an identical exposure time, the lengths of streaklines tell us local velocities directly.

With this in mind, we can make some interesting observations as follows. First, it is seen in Figure 1 that at various axial positions the steaklines near the entrance of the die are longer than those further upstream in the reservoir. This indicates that as the melt approaches the die entrance, it gets accelerated. Second, it is also seen in Figure 1 that the streaklines along the centerline are longer than those away from the centerline. Note further the orientation of streaklines approaching the die entrance. These observations lead us to conclude that the flow field near the die entrance is converging. Earlier, similar streak photographs were reported by Metzner et al.<sup>15</sup> who investigated a dilute viscoelastic polymeric solution, and more recently by Murthy and Boger<sup>9</sup> who investigated developing velocity profiles of inelastic polymeric solutions.

In the past, the flow behavior of viscoelastic polymeric melts approaching the sharp-edged entrance of a die has attracted much attention from several investigators, in particular those<sup>16–18</sup> who were concerned with the phenomenon of polymer melt flow instability, the commonly known "melt fracture." Note, however, that these investigators were not concerned with obtaining quantitative information of the velocity distributions in the entrance region.

A minimum of 15 streak velocity pictures were taken for each flow rate and for each area investigated. The test cell was divided into two separate regions; the upstream reservoir and the downstream slit. The photographs of the reservoir area covered the region from 4 to 8 slit width lengths upstream of the slit entrance. The photographs of the slit region were taken in the fully developed flow section, between 15 and 18 slit width lengths downstream of the entrance. The entrance region photographs were enlarged between 14 and 20 times their original size, while the slit region photographs were enlarged to approximately 30 times their original size.

The slit velocity data was obtained by measuring the streak length and its vertical position with the aid of a ruler with 0.02-in. increments. The slit width was also measured and the polymer point velocity was calculated with the following equation:

$$V = \left(\frac{S_L}{M_{sw}}\right) \times \left(\frac{T_{sw}}{S_s}\right) \tag{1}$$

where  $S_L$  = streak length;  $M_{SW}$  = measured slit with;  $T_{SW}$  = true slit width; and  $S_S$  = shutter speed of the camera.

## **Determination of Velocity Profiles**

In order to assess the reliability of the experimental procedure used for the velocity measurements, we first analyzed the streak photographs in the fully developed region, that is, the downstream in the slit die section. This is because in the fully developed region we can directly compare the experimentally determined velocity profiles with the theoretically predicted ones.

If we assume that the flow is fully developed and that the power-law model is adequate for describing the fully developed flow behavior of polymeric melts, the theoretical expression for the velocity profiles in the slit die may be given by<sup>19</sup>

$$\frac{u}{V} = \frac{2n+1}{n+1} \left[ 1 - \left(\frac{2y}{h}\right)^{\frac{n+1}{n}} \right]$$
(2)

where V is the average velocity defined by V = Q/wh.



FIG. 2. Velocity profiles in the fully developed slit flow region for high-density polyethylene at 200°C; Q = 11.16 cc/min; V = 16.20 cm/min.

Figures 2 to 4 give a comparison of the experimentally determined velocity profiles with the theoretically predicted ones for the three polymers investigated. It may be said from these figures that to all intents and purposes agreement between the two is quite satisfactory. It should be noted that the values of the power-law constants used in calculating the velocity profiles were those obtained from the wall pressure measurements in Test Cell #2, as described in part I of this series.<sup>14</sup> Table I gives numerical values of the power-law constants of the three materials investigated.

Velocity data in the upstream region was obtained by superimposing a cylindrical coordinate system over the entrance section as schematically

 TABLE I

 Power-Law Constants of the Three Polymer Melts Investigated

| Material                  | Temp.,<br>°C | n<br>(dimensionless) | $ \begin{pmatrix} K, \\ \frac{dynes}{cm^2} \sec^n \\ \times 10^{-5} \end{pmatrix} $ |
|---------------------------|--------------|----------------------|---|
| High-density polyethylene | 200          | 0.402                | 0.233   |
| Polypropylene             | 200          | 0.466                | 0.557   |
| Polystyrene               | 225          | 0.935                | 1.061   |



FIG. 3. Velocity profiles in the fully developed slit flow region for polystyrene at 225°C; Q = 10.92 cc/min; V = 15.90 cm/min.

shown in Figure 5. This was done because of the radial nature of the flow lines in this region. It should be noted in Figure 1 that outside of the somewhat arbitrarily chosen wedge section, the flow lines were slightly curved near the entrance. Since the melt velocity in this outer region was so small when compared to that in the inner section, data in this outer section were ignored.

The vertex of the cylindrical coordinate system was found by extending the streaklines on the photograph with a straight edge toward the slit entrance. The point of common intersection was considered the vertex of the system. A series of circular arcs was then drawn with a compass, using the vortex as the center. The angular position of the streaks that intersected the arcs were measured with the aid of a protractor which was divided into 0.5-degree increments. The radial point velocity was then calculated by measuring the streak length and utilizing eq. (1) that applied in the slit flow region.

Figures 6 to 8 give the velocity profiles upstream of the die entrance for the three materials investigated, at different radial distances from the vertex (refer to Fig. 5). As one would expect, the profiles indicate that as one approaches the entrance (i.e., smaller value of r), the velocity increases. Figures 6 to 8 further indicate that the velocity is largest at the



FIG. 4. Velocity profiles in the fully developed slit flow region for polypropylene at  $200^{\circ}$ C; Q = 13.68 cc/min; V = 19.98 cm/min.

centerline and decreases to zero as one moves toward the walls of the test cell. It is interesting to note that the results reported above show that the velocity profiles of polymeric melts are not uniform (or plug flow) near the die entrance. This is in contrast to the results reported by Uebler<sup>7</sup> who indicated a virtually flat profile at or near the entrance in flow of a dilute polymeric solution.



FIG. 5. Schematic diagram of the entrance region superimposed by a cylindrical coordinate system.



FIG. 6. Velocity profiles in the entrance region for high-density polyethylene at 200°C.



FIG. 7. Velocity profiles in the entrance region for polystyrene at 200°C.



FIG. 8. Velocity profiles in the entrance region for polypropylene at 200°C.

The scatter in the data of the upstream reservoir shown in Figures 6 to 8 is not entirely unexpected. It is believed that one cause of the scatter is related to the camera's depth of focus. When the data were recorded, the camera was focused on the centerline of the cell. However, when the pictures were developed, certain streaklines were shown brighter than others. This is believed due to the depth of focus (or depth of field of the camera and lens system). Since it was almost impossible to determine which particles were at the center plane, all the particles observed along any given radial position were recorded and used for plotting the data. Another possible source of error may be related to the fact that, in the upstream reservoir, the flow may not have been uniform across the entire depth of the test cell. This is because the aspect ratio of the reservoir section was about 2, whereas the aspect ratio of the slit section was greater than 10. Thus, in the upstream reservoir section, the particles moving in a plane away from the centerline and yet still in the focus may have been moving at a slower velocity than those at the centerline. This effect is more important in velocity measurements than in birefringence measurements, because in the latter one is averaging the stress across the entire test cell.

Still another source of error affecting the reservoir velocity measurements is related to the choice and measurement of the particle streaks at any given radial position. At any time when particle streaklines cut the circular arc drawn for a given radial distance, their lengths were used for the velocity calculation at that radial position, regardless of whether the arc cuts 50%, 25%, or even 10% of the streak length; that is, the total length was still used in the calculation. With regard to the error incurred in taking the actual velocity measurements from the photographs, we have estimated the maximum error to be within  $\pm 0.12$  cm/min. This is based on eq. (1), where the camera shutter speed is considered to have an error of  $\pm 5\%$  and the streak length and slit measurements have a maximum error of  $\pm 0.025$  cm.

It is appropriate at this point to give some theoretical consideration of the experimentally determined velocity profiles in the entrance region. Earlier, Metzner et al.<sup>15</sup> considered flow problems associated with converging flow of viscoelastic fluids by approximating the converging flow field with the uniaxial, extensional flow field. However, in view of the experimentally determined velocity profiles presented above, such an approximation is believed to be oversimplification of the problem. Because, as may be seen in Figure 1, the orientation of streaklines approaching the die entrance from the upstream reservoir varies as one moves from the centerline to the walls of the channel and, as may be seen in Figures 6 to 8, the velocity is largest at the centerline and decreases as one moves towards the walls of the channel.

For the particular experimental system being considered, however, it appears that use of the cylindrical coordinate system as schematically shown in Figure 5 is most appropriate, giving the velocity field represented by

$$V_{\tau} = V_{\tau}(r,\theta), \qquad V_{\theta} = V_{z} = 0. \tag{3}$$

Note that it is assumed in eq. (3) that there is no circulatory motion within the converging flow field. Substitution of eq. (3) into the continuity equation

$$\frac{\partial}{\partial r} (rV_r) = 0 \tag{4}$$

gives

$$V_r(r,\theta) = f(\theta)/r, \qquad V_\theta = V_z = 0$$
 (5)

in which  $f(\theta)$  is an as yet undetermined function depending on  $\theta$  only. The function  $f(\theta)$  in eq. (5) can be determined, however, when the equations of motion are solved for  $V_r(r,\theta)$ , subject to boundary conditions:

$$V_r(r,\,\theta\,=\,\pm\,\alpha)\,=\,0\tag{6}$$

$$\left(\frac{\partial V_r}{\partial \theta}\right)_{\theta=0} = 0. \tag{7}$$

However, there can be a theoretical argument against the use of boundary condition (6), because it is not certain if the velocity at the fluid-fluid boundary ( $\theta = \pm \alpha$ ) is really zero. Of course, use of eq. (6) can certainly be justified when the flowing fluid is confined by a converging channel (i.e., by solid walls).

In order to further pursue the theoretical approach presented above, the authors have carried out a separate study of the flow of viscoelastic polymeric melts through a *converging channel*, having a half-angle of 30 degrees at the die entrance. In a subsequent paper,<sup>20</sup> part III of this series, we shall present some representative results of the study.

## CONCLUSIONS

The technique of streak photography has been proven to be a useful tool for the measurement of local velocities of molten polymers. The technique was first checked against the theoretical prediction in the fully developed flow region in a slit die, yielding reasonable agreement. Then the technique was applied to determining the velocity profiles in the entrance region of a slit die having a sharp-edged entrance. The velocity measurements in the entrance region clearly show that, as expected, the fluid is rapidly accelerated as it approaches the entrance of a die. The present study further indicates that the technique of streak photography can be applied to flow geometries much more complicated than the one considered in this paper. This possibility is of particular significance in the study of the flow behavior of industrially important polymeric melts.

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