Mathematical Model of Shear Rate of Polymer Melt **Dynamic Extruding Through Capillary**

Yue Jun Liu,^{1,2} Jin Ping Qu,¹ Xian Wu Cao¹

¹The National Engineering Research Center of Novel Equipment for Polymer Processing, South China University of Technology, Guangzhou, China ²Key Laboratory of New Material and Technology for Package, Zhuzhou Institute of Technology, Zhuzhou, China

Received 2 December 2003; accepted 29 June 2004 DOI 10.1002/app.21281 Published online in Wiley InterScience (www.interscience.wiley.com).

ABSTRACT: With superimposing a sine vibration of displacement on the extruding direction of a polymer melt, the characterization formula of the shear rate of a polymer melt within a capillary was set up. By making use of the experimental equipment of a constant velocity type dynamic rheometer of capillary (CVDRC) designed by the authors, the calculating steps of the shear rate of the polymer melt at the wall of the capillary under a vibration force field were established. Through measuring and analyzing the instan-

taneous data of capillary entry pressure, capillary volume rate, and their phase-difference under the superimposed vibration, the polymer melt's shear rate at the wall of the capillary can thus be calculated. © 2005 Wiley Periodicals, Inc. J Appl Polym Sci 95: 1056–1061, 2005

Key words: shear rate; polymer melt; vibration force field; capillary

INTRODUCTION

Introducing a vibration force field to strengthen the forming process of a polymer is an important technique, called the dynamic forming technique.¹⁻² During the dynamic forming process, the fluid of a polymer melt becomes an unstable flow, and auxiliary stresses are superimposed on the major shear flow; thus, the rheological behaviors of polymers will be decided by the combinatorial stresses. The introducing of a vibration force field causes a deep influence on the forming process of the polymer,^{3–8} but its mechanism has not been explored until now. In this article, by using experimental equipment of a constant velocity type dynamic rheometer of capillary (CVDRC) designed by the authors,⁹ the instant entry pressure of the capillary and the instant volumetric flow rate, as well as their phase-difference, have been examined; then the distribution of the shear rate within the capillary under the vibration force field is obtained. It provides an important theory basis for investigating deeply the dynamic forming mechanism of polymers, and also for optimizing the technologic parameters

and structure parameters of dynamic forming equipment.

ANALYSIS OF THE FLOW FIELD WITHIN THE CAPILLARY

CVDRC superimposes in parallel form a sinusoidal displacement vibration on the extrusion direction of the polymer melt; thus, a periodic vibration force field is introduced into the whole extruding process of the polymer melt through the capillary (see Fig. 1).

Assuming that the length of the capillary is L and the radius is *R*, the length of the entire development region is L'. For convenience sake in this study, the cylinder coordinate system r, θ , z was adopted, in which the direction of flow velocity of the melt is on zdirection, that of the velocity gradient is on *r* direction, and the neutral direction is on θ direction, as shown in Figure 2. When theoretically analyzing the dynamic flow of the polymer melt within the capillary, it is presumed that the melt has isothermal, full developed and axial stratified flow, and is incompressible, there is no slipping on the inside wall of the capillary, and the gravitational force is omitted.

According to the above hypotheses, it may be known that the flow field bears the following distributive form:

$$\boldsymbol{v}_z = \boldsymbol{v}(r, t), \, \boldsymbol{v}_r = 0, \, \boldsymbol{v}_\theta = 0 \tag{1}$$

Under definite frequency *f* and amplitude *A* of vibration, the extrusion rod plays a sinusoidal displacement

Correspondence to: Y.-J. Liu (liu_yue_jun@tom.com) or J.-P. Ou (jpqu@scut.edu.cn).

Contract grant sponsor: National Nature Science Foundation of China; contract grant number: 20027002.

Contract grant sponsor: National Nature Science Foundation of China; contract grant number: 29904001.

Journal of Applied Polymer Science, Vol. 95, 1056-1061 (2005) © 2005 Wiley Periodicals, Inc.



Figure 1 Schematic drawing of the dynamic extrusion of the capillary.

vibration; thus, the instant volumetric flow rate Q(t) of the capillary may be proposed as (see Fig. 3):

$$Q(t) = \bar{Q} \cdot (1 + \varepsilon_a \cos \omega_a t) \tag{2}$$

where Q is the average volumetric flow rate of the capillary, $\overline{Q} \cdot \varepsilon_q \cos \omega_q t$ is the pulsating part of the volumetric flow rate, ε_q is the pulsating amplitude value coefficient of the volumetric flow rate, and ω_q is the pulsating frequency of the volumetric flow rate.

Similarly, under definite frequency *f* and amplitude *A* of vibration, the instant pressure drop of the capillary may also be proposed as (see Figs. 3, 4, and 5):

$$\Delta p(t) = \overline{\Delta p} \cdot \left[1 + \varepsilon_p \cos(\omega_q t + \varphi)\right] \tag{3}$$

that is, considering the pulsating frequency of the pressure drop and the volumetric flow rate to be equal. In this equation, $\overline{\Delta p}$ is the average pressure drop of the capillary, $\overline{\Delta p} \cdot \varepsilon_p \cos(\omega_q t + \varphi)$ is the pulsat-



Figure 2 Full developed flow of the polymer melt within the capillary.



Figure 3 Time-domain analysis of the instant entry pressure of the capillary (f = 10 Hz, A = 0.1 mm, $u_0 = 1000$ mm/min).

ing part of the pressure drop, ε_p is the pulsating amplitude value coefficient of the pressure drop, and φ is the phase-difference of stress and strain. ε_p and ω_q may be obtained by using frequency-domain analysis; φ may be obtained by time-domain analyzing the signal waves collected synchronously of the vibrating displacement of the extrusion rod and the entry pressure of the capillary.

Through analyzing, it may be known that the flow field within the capillary possesses the following distribution form of shear stress:^{9,10}

$$\tau_{rz}(t)\big|_{r=0} = 0 \tag{4}$$

$$\tau_{rz}(t)\big|_{r=R} = \tau_w(t) = a \cdot \overline{\Delta p} \cdot [1 + \varepsilon_p \cos(\omega_q t + \varphi)] - b \cdot \overline{Q} \cdot \varepsilon_q \cdot \omega_q \cdot \sin \omega_q t \quad (5)$$

where coefficients *a* and *b* are, respectively, equal to

$$a = \frac{R}{2(L+N_B \cdot R)} \tag{6}$$

$$b = \frac{\rho}{2\pi R} \tag{7}$$

and N_B is the revised factor of the pressure gradient under the vibration force field,¹⁰ and ρ is the melt density.

Based on the assumption of "wall nonslipping," the boundary condition is

$$v_z|_{r=R} = v(R, t) = 0 \tag{8}$$



Measuring time (ms)

Figure 4 Gathering synchronously the analog signals of the vibrating displacement and the entry pressure of the capillary (f = 10 Hz, A = 0.1 mm)

MODEL OF SHEAR RATE AT THE WALL OF THE CAPILLARY

The axial velocity distribution of the polymer melt within the capillary under the vibration force field known from eq. (1) is

$$v_z = v(r, t) \tag{9}$$

Integrating eq. (9) and combining the boundary condition eq. (8), the instant volumetric flow rate of the capillary Q(t) under definite frequency and amplitude of vibration may be expressed as:



Figure 5 Frequency-domain analysis of the instant entry pressure of the capillary (f = 10Hz, A = 0.1mm, $u_0 = 1000$ mm/min).

$$Q(t) = \int_{0}^{R} v_{z} 2 \pi r dr = v_{z} \pi r^{2} |_{0}^{R} - \int_{0}^{R} \pi r^{2} \frac{dv_{z}}{dr} dr$$
$$= -\pi \int_{0}^{R} r^{2} \frac{dv_{z}}{dr} dr \quad (10)$$

It is known from eq. (4) and eq. (5):

$$r = R \, \frac{\tau_{rz}}{\tau_w} \tag{11}$$

The shear rate at the capillary wall should be expressed as:

$$\left. \frac{dv_z}{dr} \right|_{r=R} = -\dot{\gamma}_w(t) \tag{12}$$

where the negative sign is because flow velocity is maximum at r = 0, and the flow velocity decreases with increasing of r.

Substituting eq. (11) and eq. (12) into eq. (10), we will obtain

$$\frac{\tau_w^3 Q(t)}{\pi R^3} = \int_0^{\tau_w} \dot{\gamma}(t) \tau_{r_2}^2 d\tau_{r_z}$$
(13)

Since the two sides of eq. (13) are differentiated over $\tau_{w'}$ handling with Leibnitz regulation, then we obtain

$$\frac{3[\tau_w(t)]^2 Q(t)}{\pi R^3} + \frac{[\tau_w(t)]^3}{\pi R^3} \frac{d[Q(t)]}{d[\tau_w(t)]} = \dot{\gamma}_w(t) \cdot [\tau_w(t)]^2$$
(14)

After processing eq. (14), then we obtain

$$\dot{\gamma}_{w}(t) = \frac{1}{\pi R^{3}} \left\{ 3Q(t) + \tau_{w}(t) \cdot \frac{d[Q(t)]}{d[\tau_{w}(t)]} \right\}$$
(15)

The above equation is just the calculating formula of the instant shear rate at the capillary wall under the vibration force field.

If we do not introduce the vibration force field, eq. (15) then becomes

$$\dot{\gamma}_w = \frac{1}{\pi R^3} \left[3Q + \tau_w \cdot \frac{dQ}{d\tau_w} \right] \tag{16}$$

under the stable state, $(\tau_w = R/2L \cdot \Delta p)$, then

$$\dot{\gamma}_{w} = \frac{1}{\pi R^{3}} \left[3Q + \Delta p \cdot \frac{dQ}{d(\Delta p)} \right]$$
(17)

Eq. (17) is the Weissenberg–Rabinowitsch equation, where Q and Δp are, respectively, the volumetric flow rate and the pressure drop of the capillary under stable state.

Substituting eq. (2) and eq. (5) into eq. (15), we have

$$\dot{\gamma}_{w}(t) = \frac{1}{\pi R^{3}} \cdot 3\bar{Q}(1 + \varepsilon_{q}\cos\omega_{q}t) + \frac{1}{\pi R^{3}} \cdot \{a \cdot \overline{\Delta p} \cdot [1 + \varepsilon_{p}\cos(\omega_{q}t + \varphi) - b \cdot \bar{Q} \cdot \varepsilon_{q} \cdot \omega_{q} \cdot \sin\omega_{q}t \cdot \varepsilon_{q} \cdot \sin\omega_{q}t \cdot \varepsilon_{q} \cdot \sin\omega_{q}t - \frac{\varepsilon_{q} \cdot \sin\omega_{q}t}{\bar{\Delta p} \cdot a \cdot \varepsilon_{p} \cdot \sin(\omega_{q}t + \varphi) + \bar{Q} \cdot b \cdot \varepsilon_{q} \cdot \omega_{q} \cdot \cos\omega_{q}t}$$

$$(18)$$

When eq. (18) undergoes time-average treatment

$$\overline{\dot{\gamma}_w(t)} = \frac{1}{T} \int_0^T \dot{\gamma}_w(t) dt$$
(19)

where T is the vibration period. Then we have

$$\overline{\dot{\gamma}_{w}(t)} = \frac{\bar{Q}}{\pi R^{3}} \cdot \left[3 + \frac{a^{2} \cdot \overline{\Delta p}^{2} \cdot \varepsilon_{q} \cdot \varepsilon_{p} \cdot \cos \varphi}{a^{2} \cdot \overline{\Delta p}^{2} \cdot \varepsilon_{p}^{2} + 2a \cdot b \cdot \overline{\Delta p} \cdot \bar{Q} \cdot \varepsilon_{q} \cdot \varepsilon_{q}} \right]$$

$$\varepsilon_{p} \cdot \omega_{q} \cdot \sin \varphi + b^{2} \cdot \bar{Q}^{2} \cdot \varepsilon_{q}^{2} \cdot \omega_{q}^{2} \right]$$
(20)

where a and b are given by eqs. (6) and (7), respectively; eq. (20) is just the time-average value of the shear rate at the capillary wall under certain frequency f and amplitude A of vibration.

Under stable state (i.e., the amplitude $A \rightarrow 0$ or the vibration period $T \rightarrow \infty$), shear rate γ_w at the wall of the capillary may be considered as a special form of eq. (20), which will be proved as follows:

$$\lim_{A \to 0} \overline{\dot{\gamma}_{w}(t)} = \lim_{A \to 0} \left[\frac{1}{\pi R^{3}} \cdot 3\bar{Q} \right] + \lim_{A \to 0} \left[\frac{1}{\pi R^{3}} \cdot \bar{Q} \cdot \frac{a^{2} \cdot \overline{\Delta p^{2}} \cdot \varepsilon_{q} \cdot \varepsilon_{p} \cdot \cos \varphi}{a^{2} \cdot \overline{\Delta p^{2}} \cdot \varepsilon_{p}^{2} + 2a \cdot b \cdot \overline{\Delta p} \cdot \bar{Q} \cdot \varepsilon_{q} \cdot \varepsilon_{p} \cdot \omega_{q} \cdot \sin \varphi + b^{2} \cdot \bar{Q}^{2} \cdot \varepsilon_{q}^{2} \cdot \omega_{q}^{2} \right] \\ = \frac{1}{\pi R^{3}} \cdot 3Q + \frac{1}{\pi R^{3}} \cdot Q \cdot \frac{\lim_{A \to 0} a^{2} \cdot \overline{\Delta p^{2}} \cdot \varepsilon_{p}^{2} + \lim_{A \to 0} 2a \cdot b \cdot \overline{\Delta p} \cdot \bar{Q} \cdot \varepsilon_{q} \cdot \varepsilon_{p} \cdot \omega_{q} \cdot \sin \varphi + \lim_{A \to 0} b^{2} \cdot \bar{Q}^{2} \cdot \varepsilon^{2} \cdot \omega_{q}^{2}}{\lim_{A \to 0} a^{2} \cdot \overline{\Delta p^{2}} \cdot \varepsilon_{p}^{2} + \lim_{A \to 0} 2a \cdot b \cdot \overline{\Delta p} \cdot \bar{Q} \cdot \varepsilon_{q} \cdot \varepsilon_{p} \cdot \omega_{q} \cdot \sin \varphi + \lim_{A \to 0} b^{2} \cdot \bar{Q}^{2} \cdot \varepsilon^{2} \cdot \omega_{q}^{2}}$$
(21)

It is known that

$$\lim_{A \to 0} \varphi = 0 \tag{22}$$

$$\lim_{A \to 0} \omega_q = \lim_{T \to \infty} \frac{2\pi}{T} = 0$$
(23)

then eq. (21) may be processed as:

$$\lim_{A \to 0} \overline{\dot{\gamma}_w(t)} = \frac{1}{\pi R^3} \cdot 3Q + \frac{1}{\pi R^3} \cdot Q \cdot \frac{\lim_{A \to 0} a^2 \cdot \overline{\Delta p^2} \cdot \varepsilon_p}{\lim_{A \to 0} a^2 \cdot \overline{\Delta p^2} \cdot \varepsilon_p^2}$$
$$= \frac{1}{\pi R^3} \cdot 3Q + \frac{1}{\pi R^3} \cdot Q \cdot \lim_{A \to 0} \frac{\varepsilon_q}{\varepsilon_p} \quad (24)$$

The pulsating amplitude value coefficient of the volumetric flow rate of the capillary is known from eq. (2) as:

$$\varepsilon_q = \frac{|Q_{peak}| - \bar{Q}}{\bar{Q}} \tag{25}$$

where $|Q_{peak}|$ peak is the pulsating peak value of the volumetric flow rate.

It is known from eq. (3) that the pulsating amplitude value coefficient of the pressure drop of the capillary is

$$\varepsilon_p = \frac{|\Delta p_{peak}| - \overline{\Delta p}}{\overline{\Delta p}} \tag{26}$$

where $|\Delta p_{peak}|$ is the pulsating peak value of the pressure drop.

Substituting eq. (25) and eq. (26) into eq. (24), thus we have

$$\lim_{A \to 0} \overline{\dot{\gamma}_w(t)} = \frac{1}{\pi R^3} \cdot 3Q + \frac{1}{\pi R^3} \cdot Q$$
$$\cdot \lim_{A \to 0} \left[\frac{|Q_{peak}| - \bar{Q}}{|\Delta p_{peak}|} - \overline{\Delta p} \cdot \overline{\Delta p} \right] \bar{Q} = \frac{1}{\pi R^3} \cdot 3Q$$
$$+ \frac{1}{\pi R^3} \cdot Q \cdot \lim_{A \to 0} \frac{|Q_{peak}| - \bar{Q}}{|\Delta p_{peak}| - \overline{\Delta p}} \cdot \lim_{A \to 0} \frac{\overline{\Delta p}}{\bar{Q}} = \frac{1}{\pi R^3} \cdot 3Q$$
$$+ \frac{1}{\pi R^3} \cdot Q \cdot \frac{dQ}{d(\Delta p)} \cdot \frac{\Delta p}{Q} = \frac{1}{\pi R^3} \left[3Q + \Delta p \cdot \frac{dQ}{d(\Delta p)} \right] \quad (27)$$

accordingly back to the Weissenberg–Rabinowitsch equation, that is, eq. (17), where Q and Δp are, respectively, the volumetric flow rate and the pressure drop under stable state.

In eq. (18), Q, $\varepsilon_{p'}$, ω_q and φ can be obtained by experiment. To calculate, the pulsating amplitude value coefficient of the volumetric flow rate of the capillary ε_q needs to be obtained yet.

When the melt has been hypothesized to be incompressible, the dynamic extrusion process may be handled by applying the law of conservation of mass, namely

$$\int_{0}^{R_0} u_z 2\pi r dr = Q(t) \tag{28}$$

where R_0 is the inside radius of the material cylinder and u_z is the absolute velocity of the piston rod in CVDRC.

$$u_z = u_0 + \gamma_0 \cos \omega t \tag{29}$$

where u_0 is the set velocity of the linear motion of the extrusion rod moving downward relative to the material cylinder and γ_0 is the vibrating amplitude value of the velocity of the extrusion rod.

Substituting eq. (29) and eq. (2) into eq. (28), we have

$$\int_{0}^{R_{0}} (u_{0} + \gamma_{0} \cos \omega t) 2\pi r dr = \bar{Q} \cdot (1 + \varepsilon_{q} \cos \omega_{q} t) \quad (30)$$

After processing eq. (30), we obtain

$$\pi R_0^2 u_0 \cdot \left(1 + \frac{\gamma_0}{u_0} \cos \omega t\right) = \bar{Q} \cdot (1 + \varepsilon_q \cos \omega_q t) \quad (31)$$

The two sides of eq. (31) are identical, that is, we have

$$\pi R_0^2 u_0 = \bar{Q} \tag{32}$$

$$\omega = \omega_q \tag{33}$$

$$\frac{\gamma_0}{u_0} = \varepsilon_q \tag{34}$$

Through eq. (34), ε_q may be calculated; it means that the pulsating amplitude value coefficient ε_q of the volumetric flow rate of the capillary is equal to the ratio value between the velocity vibrating amplitude γ_0 and its set velocity u_0 of the extrusion rod.

CALCULATION OF THE SHEAR RATE AT THE WALL OF THE CAPILLARY

Adopting the experimental equipment of CVDRC designed by the authors, the calculating steps of the shear rate of the polymer melt at the wall of the capillary under a vibration force field can be described as:

- 1. Setting the falling velocity of the piston rod u_0 and the vibrating displacement $A \sin \omega t$.
- Real-time collecting the instant entry pressure of the capillary *p*(*f*,*A*,*t*).
- 3. Time-domain analyzing the instant pressure drop $\Delta p(t)$, calculating the mean pressure drop (see Fig. 3).
- 4. Frequency-domain analyzing the instant pressure drop $\Delta p(t)$, and obtaining the pulsating amplitude value coefficient of the pressure drop ε_p and the pulsating frequency ω_q (see Fig. 5).
- 5. Calculating the average volumetric flow rate of the capillary
- Calculating the pulsating amplitude value coefficient of the volumetric flow rate of the capillary ε_q.
- 7. Experimentally specifying the revised factor of the pressure gradient N_B under certain frequency *f* and amplitude *A* of vibration,¹⁰ and at velocity u_0 .
- 8. Through time-domain analyzing the signal waves gathered synchronously of the vibrating displacement of the extrusion rod and the entry pressure of the capillary, we obtained the approximate value of the phase-difference φ (see Fig. 4).
- 9. Finally, calculating the shear rate of the polymer melt at the wall of the capillary under a vibration force field according to eq. (20).

In the above experiment, Sign Q200 Low-Density Polyethylene (LDPE) was adopted as the experimental material, and the melt temperature of LDPE was set to be 165°C.

CONCLUSION

Until now, there was not a constitutive equation that could accurately describe the nonlinear rheological behaviors of polymer melts under vibration force fields; thus, it is a fully essential question to explore the response mechanism of rheological behaviors of polymer melts to vibration force fields, and to describe it mathematically. This article only studies the shear rate of polymer melts under the superimposing vibration, and has obtained the following fruits: (1) the characterization model of the shear rate of polymer melt under a vibration force field was set up on the basis of rheological measurement; (2) the calculating steps of the above shear rate were also set up by the authors; and (3) making use of the experimental equipment of CVDRC designed by the authors, the shear rate at the capillary wall was thus calculated, which is quite useful to explain the dynamic forming mechanism of polymers.

The authors wish to acknowledge financial support provided by the National Nature Science Foundation of China (Project 20027002) and the National Nature Science Foundation of China (Project 29904001).

References

- 1. Ibar, J. P. Polymer Engineering and Science 1998, 38, 1.
- 2. Liu, Y. J.; Qu, J. P.; Ren, H. L. China Plastics 2001, 15, 12.
- 3. Dunwoody, J. J Non-Newtonian Fluid Mech 1996, 65, 195.
- 4. Miroshnychenko D.; Clarke, N. Physical Review E 2002, 66, 1.
- 5. Qu, J. P.; Xu, B. P. Plastics, Rubber and Composites 2002, 31, 432.
- 6. Giacomin, A. J.; Jeyaseelan, R. S. Polymer Engineering and Science 1995, 35, 768.
- Panov, A. K.; Dorokhov, I. N.; Shulaeva, T. V.; Kafarov, V. V. Doklady Akademii Nauk 1992, 322, 560.
- Martins, M.; Covas, J. A. Key Engineering Materials 2002, 230– 232, 300–302.
- 9. Liu, Y. J. Dissertation, South China University of Technology, Guangzhou, 2002.
- 10. Liu, Y. J.; Qu, J. P. J of Vibration and Shock 2004, 23, 55.