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Thermal Modeling in Polymer Extrusion

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Abstract

In this paper we consider thermal effects of polymer flows through a cylindrical die. First, we derive a model for the oscillatory behavior of polymer flow in an extruder given a functional relation between the pressure and flow rate. A simple isothermal but temperature dependent model is constructed to find this relation. Unfortunately, the model is shown to be invalid in the physical regime of interest. We present several arguments to suggest that the isothermal assumption is reasonable but that a more detailed understanding of the small-scale molecular dynamics near the boundary may be required. Second, we show that a simplified model for thermoflow multiplicity in a cooled tube is inconsistent, when the stationary non-Newtonian flow is assumed to be incompressible without radial pressure gradients and without radial velocity. This inconsistency can be removed by allowing for weak compressibility effects in the down-steam area.

5.1   Introduction

A detailed understanding of polymer flow is important in the manufacturing of many modern plastics. In the extrusion process molten plastic is forced through a small aperture or "die" by a winding screw. The manufacturer would of course like to get the highest possible output through such a machine. However, when the flow rate

is too high hysteresis can occur and instabilities in the flow result in defects in the product (see Figure 5.1).

Different defects are thought to be caused by different physical mechanisms, such as a stress singularity at the outlet or hysteresis in the flow. This hysteresis comes from a multivalued relation between flow rate and applied pressure and the following hysteretic cycle can be observed experimentally (see Figure 5.2). Such a cycle has been shown to lead to an oscillatory flow regime (Molenaar and Koopmans 1994). One model for this behavior is to assume a stick-slip condition at the polymer-extruder interface while ignoring temperature effects within the polymer (Den Doelder, 1999). Alternatively, in this paper we employ a standard no-slip condition at the boundary of the die and allow, in addition, for temperature variation in the polymer.
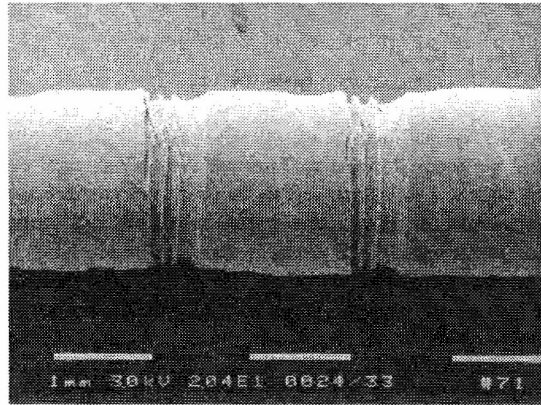
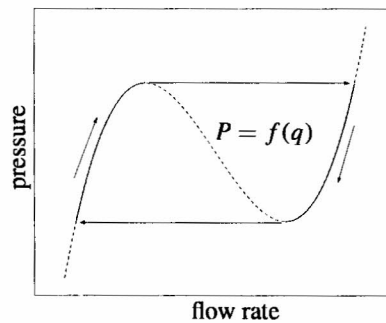Figure 5.1: Surface instabilities in an extruded polymer

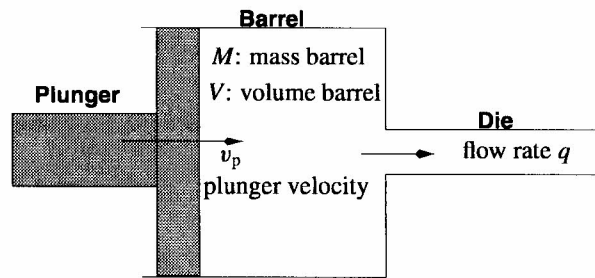Figure 5.2: Sketch of a hysteretic flow profile

Figure 5.3: Geometry of the barrel-die system

5.2   Modeling the barrel-die system

We begin by modeling the physical device not as the screw-driven extruder as used in manufacturing but rather as two chambers of different radii with a plunger moving at constant speed forcing the polymer through the aperture. This is reasonable as it is the exact geometry of experimental rheometers. Because the volume of the barrel is considerably greater than that of the die, which is entirely open at the far end. We take the polymer to be compressible in the barrel but not in the die.

The compressibility relates the pressure in the barrel, P, to the density, ρ, as

$$\dot{P} = \frac{1}{\chi}\frac{\dot{\rho}}{\rho}, \tag{5.1}$$

where χ is the coefficient of compressibility and the dot denotes differentiation with respect to time, e.g. $\dot{\rho} = d\rho/dt$. The time rate of change of the mass M in the barrel is $\dot{M} = -q$, where q is the flow rate in the die (see Figure 5.3), and the time of change of the volume V in the barrel is $\dot{V} = v_p A$ where v_p and A are the velocity and area of the plunger, respectively. Defining the barrel density as $\rho = M/V$ and with (5.1), we have

$$\frac{\dot{\rho}}{\rho} = \frac{\dot{M}}{M} - \frac{\dot{V}}{V} = \chi\dot{P}. \tag{5.2}$$

Assuming $P = f(q) = f(-\dot{M}) = \tilde{f}(\dot{M})$, we can simplify (5.2) to

$$\frac{\dot{M}}{M} - \frac{\dot{V}}{V} = \chi\,\tilde{f}'(\dot{M})\,\ddot{M}. \tag{5.3}$$

It may be seen that (5.3) has the following first integral

$$\ln\alpha = \ln M/V - \chi\,\tilde{f}(\dot{M}) \Longleftrightarrow \alpha = \frac{M}{V}\,e^{-\chi\tilde{f}(\dot{M})} \tag{5.4}$$

with integration constant α. Since $V = V_0 - v_p A t$ we have

$$\dot{V}/V = -\alpha v_p A \exp[\chi \tilde{f}(\dot{M})]/M. \tag{5.5}$$

Substitution of (5.5) into (5.3) gives the following autonomous equation

$$\ddot{M} = \frac{\alpha v_p A e^{\chi \tilde{f}(\dot{M})} + \dot{M}}{M \chi \tilde{f}'(\dot{M})} \tag{5.6}$$

with $\alpha = M(0) \exp[-\chi \tilde{f}(\dot{M}(0))]/V(0)$ and $\tilde{f}' = d\tilde{f}/d\dot{M}$. The model is closed when we specify a suitable constitutive relation $q = f(-\dot{M}) = \tilde{f}(\dot{M})$, initial data and v_p, A and χ. In the next section, we derive a constitutive relation based on several simplifying assumptions.

5.3 A temperature dependent constitutive relation

Following Stroh *et al.* (1990), we consider stationary axisymmetric, incompressible non-Newtonian flow in the cylindrical die, in which the pressure is independent of the radial direction and in which the radial velocity is zero. The incompressibility condition implies that $v = v(r)$ with v the velocity along the die and r the radial coordinate. The momentum balance in the axial z-direction and the energy equation take the form (e.g. Stroh *et al.* 1990)

$$\frac{dp}{dz} = \frac{1}{r}\frac{\partial}{\partial r}\left(\eta r \frac{\partial v}{\partial r}\right), \tag{5.7}$$

$$\rho c_p v \frac{\partial T}{\partial z} = \frac{\kappa}{r}\frac{\partial}{\partial r}\left(r\frac{\partial T}{\partial r}\right) + \eta(T, n)\left(\frac{\partial v}{\partial r}\right)^2, \tag{5.8}$$

where T is the temperature, p the pressure, c_p the specific heat at constant pressure, κ the thermal conductivity constant, and $\eta(T, n)$ the viscosity. The latter is obtained from experiments and summarized in an Arrhenius relation

$$\eta = \eta_0(T)\left|\frac{\partial v}{\partial r}\right|^{n-1} = \eta_{00} e^{\gamma \frac{T_0 - T}{T}}\left|\frac{\partial v}{\partial r}\right|^{n-1}. \tag{5.9}$$

Note that we have also assumed that the problem is steady in time, that is any temperature or pressure changes are assumed to happen instantaneously.

Defining the geometry of the system in Figure 5.4, all the parameters have the approximate SI or dimensionless values as in Table 5.1 (page 62). Froh *et al.* (1990) considered the system (5.7)–(5.8) numerically. Instead, we will derive a constitutive relation by simplifying this system based on the following assumptions:

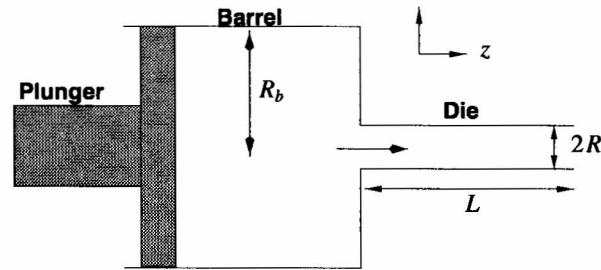- The pressure drop over the die is taken constant.

Figure 5.4: Geometry of the rheometer

- The flow in the die is isothermal which results in a parametric dependence on the temperature T in the die. The temperature T_0 of the barrel and the die walls is assumed to quickly change from T_0 to T upon entering the die.

- The heat production by the polymer in the die balances the heat loss through the die walls.

Hence, we take the pressure gradient to be constant in the die with polymer at a constant temperature T

$$\frac{dp}{dz} = \frac{\Delta P}{L}. \tag{5.10}$$

Using (5.10) and the boundary condition $v(R) = 0$, we integrate (5.7) from $z = 0$ to L and obtain

$$\frac{\Delta P}{\eta_0 L} = \frac{1}{r}\frac{\partial}{\partial r}\left(r\left|\frac{\partial v}{\partial r}\right|^n\right) \Longrightarrow \left|\frac{\partial v}{\partial r}\right|^n = \frac{\Delta P}{2\eta_0 L}r \tag{5.11}$$

$$\Longrightarrow v(r) = \frac{n}{n+1}\left(\frac{\Delta P}{2L\eta_0}\right)^{\frac{1}{n}}\left(R^{\frac{n+1}{n}} - r^{\frac{n+1}{n}}\right). \tag{5.12}$$

The flow rate q is thus given by

$$q = 2\pi\rho\int_0^R v(r)\,r\,dr$$

$$= \frac{\rho\pi n}{3n+1}\left(\frac{\Delta P}{2\eta_0 L}\right)^{\frac{1}{n}} R^{3+\frac{1}{n}}. \tag{5.13}$$

The viscous heat production, the second term on the right in the energy equation (5.8), is given by

$$2\pi L\int_0^R \eta_0\left|\frac{\partial v}{\partial r}\right|^{n+1} r\,dr = 2\pi\eta_0 L\frac{n}{3n+1}\left(\frac{\Delta P}{2L\eta_0}\right)^{\frac{n+1}{n}} R^{3+\frac{1}{n}}. \tag{5.14}$$

Parameter	Description	Value
n	power law coefficient	0.5
η_{00}	viscosity at T_0 for $n = 0.5$	10^3-10^4 kg/(m $\sqrt{s}$)
γ	Arrhenius coefficient	2.5
T_0	ambient temperature	450 K
P	pressure in barrel	$(0.1 - 10)$ MPa
L	length of the die	10^{-2} m
R	radius of the die	10^{-3} m
A	cross-sectional area of the barrel	10^{-4} m^2
v_{ave}	average exit velocity	$(10^{-3} - 10^{-1})$ m/s
v_{p}	average exit velocity	$(10^{-4} - 10^{-2})$ m/s
ρ	density of the polymer	10^3 kg/m^3
c_{p}	specific heat of the polymer	2×10^3 m^2/(s^2 K)
κ	thermal conductivity of the polymer	1 kg m/(K s^3)
h	heat conduction	$(10 - 10^4)$ kg/(K s^3)
χ	compressibility of the polymer	10^{-9} Pa^{-1}

Table 5.1: Parameter values

Setting the heat production equal to the heat lost through the boundary, i.e.

$$2\pi R L h (T - T_0)$$

with h the heat conduction, we have

$$\left(\frac{\Delta P}{L}\right)^{\frac{n+1}{n}} = h(T - T_0) \frac{3n+1}{n} \left(\frac{\eta_0 2^{n+1}}{R^{2n+1}}\right)^{\frac{1}{n}}. \tag{5.15}$$

Writing $\Delta P = P - p_{\mathrm{a}} \approx P$ with P the pressure in the barrel and p_{a} the atmospheric pressure, we can write (5.13) and (5.15) symbolically as

$$q = F_1(P, T), \qquad P = F_2(T),$$

respectively, since $\eta_0 = \eta_{00} \exp[\gamma (T_0 - T)/T]$. Hence, we have an implicit relation $P = f(q)$ which is not a cubic, but can still exhibit hysteretic behavior, as we will show. To investigate the hysteresis, we consider the zeroes and poles of

$$\frac{dP}{dq} = \frac{dP}{dT}\frac{dT}{dq} = \frac{nP}{q}\frac{(T^2 - \gamma T T_0/n + \gamma T_0^2/n)}{(T^2 + \gamma T_0 T - \gamma T_0^2)}. \tag{5.16}$$

The zeroes $T_\pm$ and pole T_{p} of (5.16) are

$$T_\pm = \frac{\gamma T_0}{2n}\left(1 \pm \sqrt{1 - 4n/\gamma}\right), \qquad T_{\mathrm{p}} = \frac{\gamma T_0}{2}\left(-1 + \sqrt{1 + 4/\gamma}\right) \qquad (5.17)$$

respectively. (Note that $T > 0$.) For the given values of γ and n this gives

$$\begin{aligned}
T_+/T_0 &= \frac{1}{2}(5 + \sqrt{5}) \sim 3.62, \\
T_-/T_0 &= \frac{1}{2}(5 - \sqrt{5}) \sim 1.38, \\
T_{\mathrm{p}}/T_0 &= \frac{5}{4}(-1 + \sqrt{2.6}) \sim 0.77.
\end{aligned} \qquad (5.18)$$

We see that there is a bifurcation at $\gamma/n = 4$ which gives $T_+ = T_- = 2T_0$. The pole $T_{\mathrm{p}} < T_0$ for every γ and there is hysteresis when $= \gamma/n > 4$, see Figure 5.5. For large γ/n the temperatures take the asymptotic form

$$\frac{T_+}{T_0} \sim \frac{\gamma}{n} - 1 + O\left(\frac{n}{\gamma}\right), \qquad \frac{T_-}{T_0} \sim 1 + O\left(\frac{n}{\gamma}\right). \qquad (5.19)$$

The relationship between applied pressure and flow rate is shown in Figure 5.6 for various values of h and η_{00}.
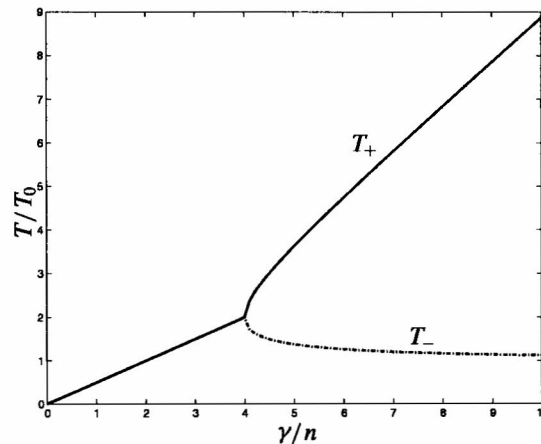

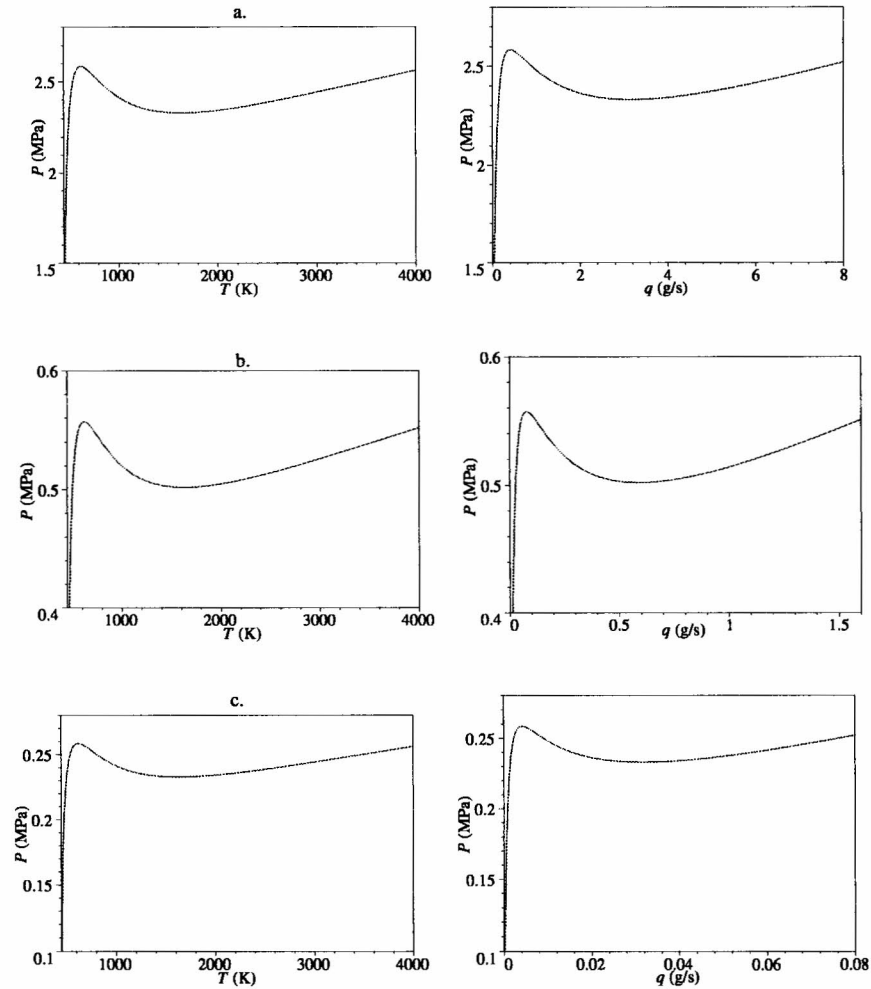Figure 5.5: Bifurcation diagram for temperature dependent polymer flows

Figure 5.6: Relation between applied pressure P and flow rate q for a) $h = 10^4 \, \mathrm{kg/(K \, s^3)}$ and $\eta_{00} = 10^4 \, \mathrm{kg/(m \, \sqrt{s})}$, b) $h = 100 \, \mathrm{kg/(K \, s^3)}$ and $\eta_{00} = 5000 \, \mathrm{kg/(m \, \sqrt{s})}$, and c) $h = 10 \, \mathrm{kg/(K \, s^3)}$ and $\eta_{00} = 10^4 \, \mathrm{kg/(m \, \sqrt{s})}$. The values of the remaining parameters are found in Table 5.1

5.4   Limitations

The assumptions we have imposed require that we check a posteriori whether the model is consistent. To that effect, we estimate the temperature difference $\Delta T = T - T_0$ by equating the work W done in the barrel with the dissipation of heat Q in the die. Using $v_p = (10^{-2} - 10^{-4})$ m/s and $P = (0.25 - 2.5)$ M Pa and where necessary other values from Table 5.1, we find

$$W = P A v_p = Q = \pi R^2 L \rho c_p \Delta T \quad \Longleftrightarrow \quad \Delta T = (0.04 - 40)\,\text{K}, \qquad (5.20)$$

respectively. With an ambient temperature of $T_0 = 450 K$ the 140% or 360% temperature change found in our model (see the estimates (5.18)) is unfortunately too large. Alternatively, the Peclet number, the ratio of convection and diffusion, for

$$Pe = \frac{\rho c_p R^2 v_{avc}}{L \kappa} \sim 0.2 - 20 \qquad (5.21)$$

for an average die velocity $v_{avc} = (10^{-3} - 10^{-1})$ m/s, respectively. The model as presented appears valid for the high flow regime, where flow rates are of the order of grams per second, although the temperature range is clearly almost two orders of magnitude too high. The magnitude of the heat conduction parameter h is very poorly estimated, while the results are clearly sensitive to h. At the higher velocities it is conceivable that there could be a boundary layer with some localized viscous heating, which analysis could lead to better estimates of h.

5.5   Compressibility effects and consistency

The model (5.7)–(5.8) turns out to be too restrictive, because it is inconsistent to insist on incompressibility, the absence of a radial velocity as well as a pressure which is independent of the radial direction. This inconsistency can be shown to arise as follows. Remember that incompressibility implies $v = v(r)$. First, since we assumed $p = p(z)$, (5.7) can be integrated and we find

$$\eta_{00}\, e^{F(T)} \left| \frac{\partial v}{\partial r} \right|^n = \frac{p(z)\, r}{2}, \qquad (5.22)$$

where $F(T) = \exp[\gamma\,(T_0 - T)/T]$. But at $z = 0$, we have $T = T_0$ so

$$\partial v / \partial r = C\, r^{1/n}, \qquad (5.23)$$

where C is an integration constant. Hence

$$\eta_{00}\, e^{F(T)}\, C^n\, r = \frac{p(z)\, r}{2} \qquad (5.24)$$

and it follows that $F(T)$ and thus is T also a function of z only. If we substitute (5.23) into (5.8) and also use the previous result that $T = T(z)$, we obtain

$$\frac{\partial T}{\partial z} = \frac{1}{\rho c_p} \frac{n+1}{n} \eta_{00} C^n e^{F(T)} \frac{(r/R)^{(n+1)/n}}{(r/R)^{(n+1)/n} - 1}.$$ (5.25)

Only the last term on the right-hand-side of (5.25) is a function of r, so we conclude that the temperature is a function of both r and z, which contradicts our previous finding that $T = T(z)$. Hence, the model is inconsistent.

We can remove this inconsistency by extending the system to include weak compressibility, as follows

$$\begin{aligned}
\frac{dp}{dz} &= \frac{1}{r} \frac{\partial}{\partial r} \left(\eta\, r \frac{\partial v}{\partial r} \right) \\
\rho c_p \frac{\partial T}{\partial z} &= \frac{\kappa}{r} \frac{\partial}{\partial r} \left(r \frac{\partial T}{\partial r} \right) + \eta \left(\frac{\partial v}{\partial r} \right)^2 \\
\rho &= \rho(p, T) \\
\frac{\partial(\rho v)}{\partial z} &= 0
\end{aligned}$$ (5.26)

plus boundary conditions at the inlet, outlet and the die wall. The specification of a (simplified) constitutive relation $\rho = \rho(p, T)$ is nontrivial and we leave its determination for future work. Although the model in Stroh *et al.* (1990) is inconsistent, their numerical solutions may be valid after all, because their numerical technique may implicitly add some weak compressibility.

5.6 Conclusions and future work

In this report we have shown that a non-isothermal, no-slip boundary condition model for extruding polymer flow can explain the observed hysteresis in pressure-flow experiments. Furthermore we have calculated that (especially in the slow flow regime) while temperature effects may exist, they do not seem crucial to the observed behavior. Instead we believe that the direction taken by the researchers at DOW - attempting to understand the dynamics at a molecular level near the boundary - is the more fruitful one.

Acknowledgment

Bibliography

[1] C.F.J. den Doelder. *Design and implementation of polymer melt fracture models*. Ph.D. thesis. TU Eindhoven, 1999.

[2] J. Molenaar and R.J. Koopmans. *Modeling polymer melt-flow instabilities*. J. Rheology. **38**(1), pp. 99–109, 1994.

[3] F. Stroh, J. Pita, V. Balakotaiah and D. Luss. *Thermoflow multiplicity in a cooled tube*. AICHE Journal **36**(3), pp. 397–408, 1990.