Flow of Molten Polymers Through Porous Media

DALE R. GREGORY and RICHARD G. GRISKEY

Virginia Polytechnic Institute, Blacksburg, Virginia

The flow of molten polymer through porous media has been investigated at constant temperatures and flow rates. Temperatures of 375°, 400°, and 425°F., flow rates from 12 to 60 g./min. and particle sizes from 0.054 in. to 6 mm. in diameter were studied. A modified Darcy's law was developed using the Metzner-Reed modification of the Mooney-Rabinowitsch relation. This form was then used to develop a modified friction factor plot which correlated the data with an average error of 5.02%. Calculated superficial velocities from the Darcy's law equation checked experimental values generally to within 10%. A technique for estimating shear rates when curved rheological flow curves occurred was also developed.

An integral part of many polymer processing operations is the flow of molten polymers through a screen or sand pack. Such a case constitutes a complex flow problem because of the non-Newtonian nature of the polymer melts. In spite of its inherent difficulty the problem merits study because of its industrial importance.

Until recently, no studies had been carried out that involved flow of non-Newtonian fluids through porous media. Then within the past two years the investigations of Bird and Sadowski (*1, 2*), Christopher and Middleman (*3*), and McKinley, Jahns, Harris, and Greenkorn (*4*) were published. All of these studies however considered only the flow of non-Newtonian polymer solutions through porous media. No reference was found in the open literature to any such studies involving molten polymers. The present work was therefore undertaken to investigate the flow of molten polymers through porous media.

A schematic of the apparatus used is shown in Figure 1. Polyethylene pellets were fed to a 1-in. laboratory extruder where they were melted and pumped to a helical gear pump driven by a Reeves varispeed drive. The gear pump metered a constant flow of polymer to the test section.

Figure 2 is a detail drawing of the test section. Two stainless steel screens (16 × 16 and 200 × 200 mesh) were placed immediately over the holes in the bottom of the test section. The screens prevented the escape of any of the porous media through the holes.

The test section was filled to a given depth with stainless steel or glass beads. These were poured from a height of 6 in., while the test section was continuously rotated to provide a random bed packing. At ½-in. bed height intervals the bed was tamped ten times with a special device dropped from a height of about 6 in. Packed section bed depth was carefully measured with an architect's scale. This made possible the precise determination of bed porosity.

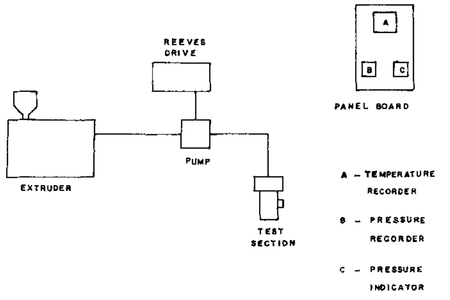

Fig. 1. Schematic of apparatus.

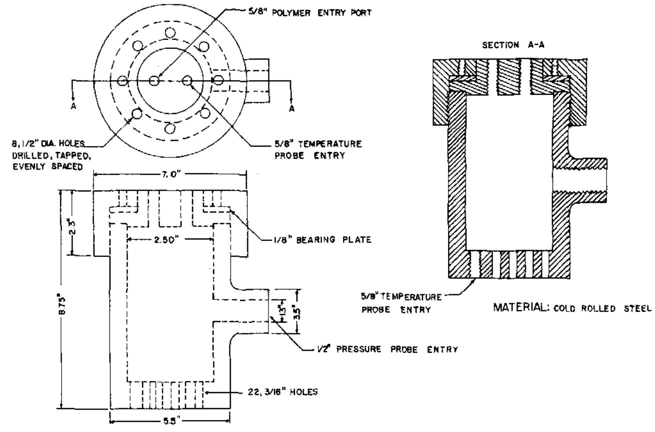

Fig. 2. Detail of test section.

Experiments were performed at constant temperatures and flow rates. Temperatures of 375°, 400°, and 425°F., flow rates from 12 to 60 g./min., and particle sizes of 0.054 and 0.125 in. and 3, 4, 5, and 6 mm. were studied.

Pressures were measured with a Taylor pressure bulb and temperatures with Aminco high-pressure thermocouples.

Data were taken both for a given bed and also for the test section under the same conditions without the bed. The pressure drop obtained in the latter case was subtracted from the overall pressure drop with the bed present to give a corrected pressure drop for the bed alone.

Figure 3 represents a typical plot of the experimental data. As can be seen, the pressure drop increases with increasing flow rate as would be expected. Higher pressure drops are also obtained for lower temperatures at a given flow rate.

Figures 4 and 5 are plots of the pressure drop vs. particle diameter at a constant temperature and a constant flow rate, respectively. Here again the behavior is as would be expected with changing flow rate in Figure 4 and changing temperature in Figure 5.

One feature of both plots however merits some discussion: the sharp rise in pressure drop as the particle diameter changed from 3 mm. (glass) to 0.054 in. (stainless steel). This sudden change could be due to one of two

---

Dale R. Gregory is with the Tennessee Eastman Corporation, Kingsport, Tennessee.

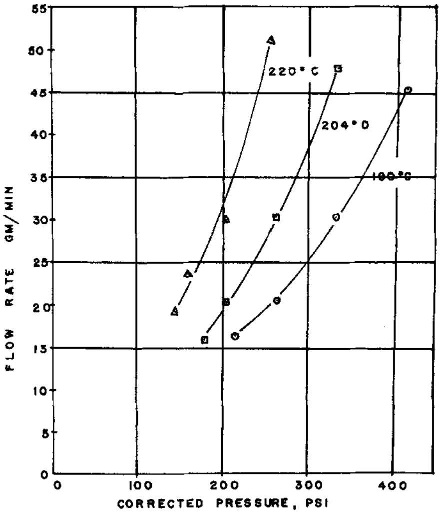

Fig. 3. Mass flow rate vs. pressure drop with temperature as a parameter for a 1.015-in. deep bed of 3-mm. glass spheres.

possibilities: an effect of the surface of the particle or a sudden increase in the shear rate of the flowing polymer due to a sudden decrease in particle size. Additional experiments were run to evaluate the possibility of a surface effect. Stainless steel spheres 0.125 in. in diameter were obtained and tested. These spheres were quite close in diameter to the 3-mm. glass spheres. Porosities differed only slightly. Figure 6 is a plot of data both for the glass and stainless steel spheres. As can be seen the data for both types of spheres essentially fell on the same lines (these data were corrected for porosity differences). The data of Figure 6 therefore indicate that the sharp break in Figures 4 and 5 is not likely due to a particle surface effect.

It was decided to correlate the data of the present study in the form of a modified friction factor vs. modified Reynolds number plot. A similar approach had been taken by previous investigators. Sadowski and Bird (*1*, *2*) developed such a relationship on the basis of the Ellis equation, while Christopher and Middleman used power law to describe the rheological system. A somewhat different approach was taken in the present work. Rheology was described by the relation derived from the Mooney-Rabinowitsch relation by Reed and Metzner (*5*):

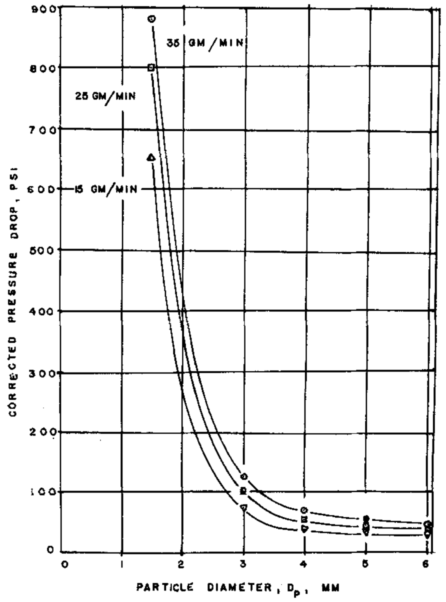

Fig. 4. Corrected pressure drop vs. particle diameter with flow rate as a parameter for a 0.5-in. deep bed at 220°C.

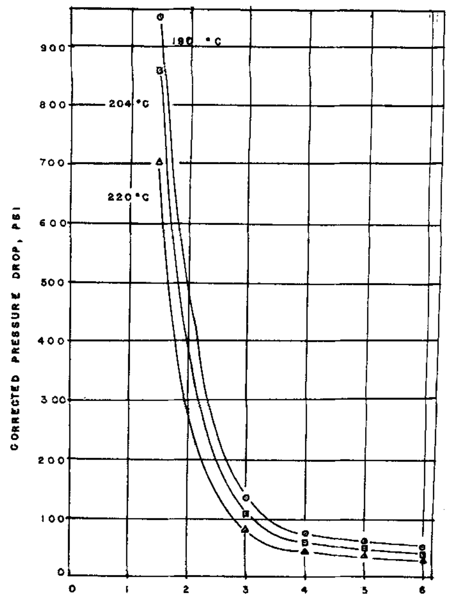

Fig. 5. Corrected pressure drop vs. particle diameter with temperature as a parameter for a bed depth of 0.5 in. and a flow rate of 17.5 g./min.

$$\frac{D\Delta P}{4L} = K'\left(\frac{8V}{D}\right)^{n'} \tag{1}$$

This equation is especially appropriate for polymer melts, since their rheological behavior is determined with capillary rheometers which gave $(D\Delta P/4L)$ vs. $8V/D$ data. In addition, the great difficulty in studying polymer melts in rotational viscometers makes the use of the Ellis equation or the direct use of the power law prohibitive.

The starting point in the development of the correlation was to cite the form of Darcy's law for Newtonians:

$$V_o = \frac{k}{\mu}\,\frac{\Delta P}{L} \tag{2}$$

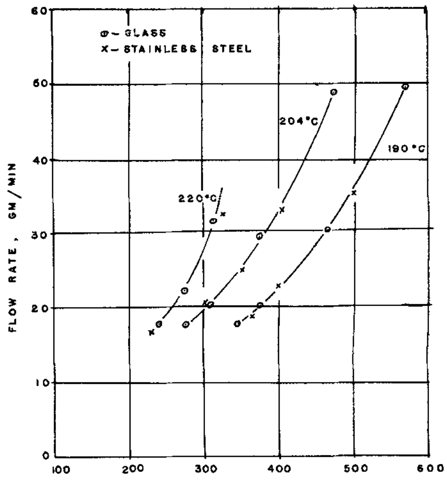

Fig. 6. Comparison behavior of stainless steel and glass spheres in a 0.5-in. deep bed.

and to indicate that the same form would be followed for non-Newtonians except that a modified viscosity η_{mod} would have to be used:

$$V_o = \frac{k}{\eta_{\text{mod}}}\frac{\Delta P}{L} \tag{3}$$

then

$$V_o = \epsilon V = \frac{\epsilon R^2}{8}\left(\frac{1}{K'}\right)^{1/n'}\left(\frac{R\Delta P}{2L}\right)^{1/n'-1}\frac{\Delta P}{L} \tag{4}$$

Now if we introduce a hydraulic radius R_H and a tortuosity factor (*1*, *6*) of $(25/12)L$ for L we obtain

$$V_o = \frac{6\,\epsilon\,R_H^2}{25}\left(\frac{1}{K'}\right)^{1/n'}\left(\frac{12\,R_H\,\Delta P}{25L}\right)^{1/n'-1}\frac{\Delta P}{L} \tag{5}$$

Comparing with Equation (3) we get

$$\frac{1}{\eta_{\text{mod}}} = \frac{6\,\epsilon\,R_H^2}{25k}\left(\frac{1}{K'}\right)^{1/n'}\left(\frac{12\,R_H\,\Delta P}{25L}\right)^{1/n'-1} \tag{6}$$

Following Blake (*7*) we obtain

$$k = \frac{\epsilon R_H^2}{C} = \frac{Dp^2\,\epsilon^3}{c(1-\epsilon)^2} \tag{7}$$

where $c = 36C$, so that

$$\frac{1}{\eta_{\text{mod}}} = \frac{c}{12}\left(\frac{2}{25K'}\right)^{1/n'}\left(\frac{Dp\,\epsilon\,\Delta P}{L(1-\epsilon)}\right)^{1/n'-1} \tag{8}$$

Now, if Ergun's (*7*) form of the friction factor equation is used

$$f^* = \left[\frac{(p_o - P_L)\rho}{G_o^2}\right]\left[\frac{D_p}{L}\right]\left[\frac{\epsilon^3}{(1-\epsilon)}\right] \tag{9}$$

where

$$(N_{Re})_{\text{mod}} = \frac{D_p\,G_o}{(1-\epsilon)\,\eta_{\text{mod}}} \tag{10}$$

In laminar flow

$$f^* = \frac{c}{(N_{Re})_{\text{mod}}} \tag{11}$$

The experimental data and rheological data (*9*) were then used to develop a plot of f^* vs. $(N_{Re})_{\text{mod}}$. This plot is shown in Figure 7. The value of c in Equation (11) was found to be 180. Carman (*10*) reported 180 for Newtonian fluids and Sadowski (*1*, *2*) 180 for a non-Newtonian system using the Ellis equation. The average error in friction factor from the present correlation was 5.02%, which compared with 6.51% for Sadowski's work.

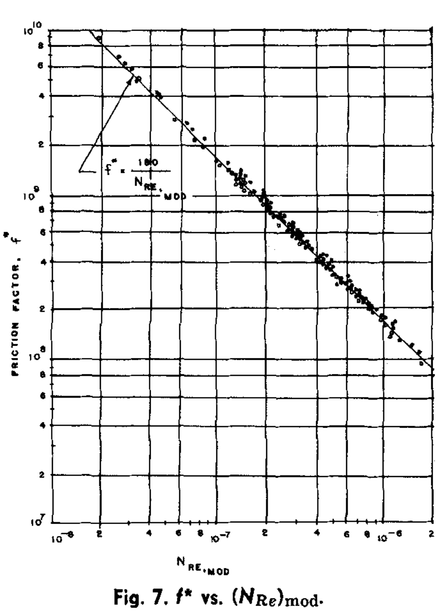

Fig. 7. f^* vs. $(N_{Re})_{\text{mod}}$.

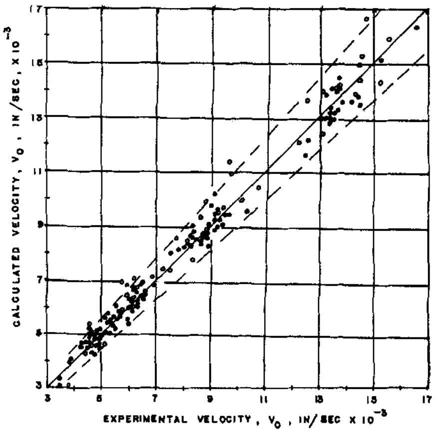

Fig. 8. Calculated vs. experimental superficial velocity.

The modified Reynolds number range in the present work covered slightly more than two decades (10^{-8} to 10^{-6}). The ranges covered by the earlier investigators were (*1*, *2*) 10^{-4} to 10^{-1} and (*3*) 10^{-1} to 10^{-2}. Although the modified Reynolds numbers are somewhat different, the present work is definitely in a range not previously studied. Direct data comparison is not possible because of the differences in fluid models.

The work of the other investigators (*4*) was not correlated on the basis of a modified friction factor plot. Here a comparison of sorts was made on the basis of predicted volumetric flow rates vs. observed rates. These data for the present work are shown in Figure 8. The behavior obtained was comparable to that found by McKinley, Johns, Harris, and Greenkorn (*4*).

A possible difficulty that could arise in the use of the correlation of this study is that the melt flow curve of log $(D\Delta P/4L)$ vs. log $(8V/D)$ might be a curve rather than a straight line. This did not occur in this work, but it is a definite possibility. In such cases it is suggested that the appropriate shear stress to be used be estimated from the relation

$$\left(\frac{D\Delta P_c}{4L_c}\right) = \frac{D_p\,\epsilon\,\Delta P}{12.5\,(1-\epsilon)\,L} \tag{12}$$

Values of K and n^1 can then be obtained by taking the tangent to the flow curve at $\log(D\Delta P_c/4L_c)$.

The present work should afford the process engineer a means of calculating pressure drops of molten polymers through porous media. It should be particularly useful since the correlation is based on the flow curve relating $(D\Delta P/4L)$ and $(8V/D)$, which is the type of data obtained with the capillary rheometer, the most widely used instrument for studying the rheology of polymer melts.

ACKNOWLEDGMENT

The authors gratefully acknowledge the Shell Fellowship awarded to one of them. In addition, the assistance of E. I. du Pont de Nemours and Company, Inc., in supplying the large quantities of polymer used is gratefully acknowledged.

NOTATION

a = bed wetted surface/volume of bed, ft.$^{-1}$ = $a_v(1-\epsilon)$
a_v = total bed surface/volume of bed, ft.$^{-1}$ = $6/D_p$
c = constant

C = constant
D = tube diameter, ft.
D_p = particle diameter, ft.
f = friction factor
f^* = modified friction factor
G_o = superficial mass velocity, lb./(sq.ft.) (sec.)
k = permeability, sq.ft.
K = consistency index, lb.(sec.^{n})/sq.ft.
L = depth of bed, ft.
L_c = length of capillary, ft.
n^1 = flow behavior index
$(N_{Re})_{mod}$ = modified Reynolds number
P_c = pressure drop in capillary, lb./sq.in.abs.
R = tube radius, ft.
R_H = hydraulic radius, ft.
V = average tube velocity, ft./sec.
V_o = superficial velocity, ft./sec.
ϵ = porosity
η_{mod} = modified viscosity, lb./(sec.) (ft.)
μ = viscosity, lb./(sec.) (ft.)

LITERATURE CITED

1. Sadowski, T. J., Ph.D. thesis, Univ. Wisconsin, Madison (1963).
2. ———, and R. B. Bird, *Trans. Soc. Rheol.*, 9, No. 2, 243 (1965).
3. Christopher, R. H., and Stanley Middleman, *Ind. Eng. Chem.*, 57, 93 (1965).
4. McKinley, R. M., H. O. Jahns, W. W. Harris, and R. A. Greenkorn, *A.I.Ch.E. J.*, 12, 17 (1966).
5. Metzner, A. B., and J. C. Reed, *ibid.*, 1, 434 (1955).
6. Bird, R. B., W. E. Stewart, and E. N. Lightfoot, "Transport Phenomena," pp. 180-207, Wiley, New York (1960).
7. Blake, C. F., *Trans. Am. Inst. Chem. Engrs.*, 14, 415 (1922).
8. Ergun, Sabri, *Chem. Eng. Progr.*, 48, 89 (1952).
9. Bernhardt, E. C., "Processing of Thermoplastic Materials," p. 602, Reinhold, New York (1959).
10. Carman, P. C., *Trans. Inst. Chem. Engrs.*, 15, 150 (1957).

Manuscript received March 28, 1966; revision received July 5, 1966; paper accepted July 7, 1966. Paper presented at A.I.Ch.E. Dallas meeting.

Transient Response and Feed-Forward Control of a Distillation Tower Subject to a Sequence of Upsets

G. P. DISTEFANO, F. P. MAY, and C. E. HUCKABA
University of Florida, Gainesville, Florida

A mathematical model of dynamic distillation was verified experimentally for transients resulting from a sequence of upsets in operating conditions, that is, for upsets occurring in the unsteady state before the effects of previous upsets died out.

The equations for feed-forward control of the top product composition by reflux ratio control action were derived and a computer program was written in FORTRAN for solution on a digital computer. This control model was employed in the feed-forward control of an experimental distillation column when subjected to an upset in feed composition.

Experimental data were obtained from a twelve-plate, 10-in. diameter distillation column with a methanol-tertiary butyl alcohol system. The numerical technique employed is a completely general divided difference method which can be utilized in the solution of any system of first-order differential equations.

The work described herein is an extension of that previously reported by Huckaba and co-workers (*1*, *2*). These studies involved comparison of computed predictions with experimentally observed dynamic responses to upsets in reflux ratio, feed composition, thermal input to the reboiler, and simultaneous upsets in feed composition and reboiler heat duty. The model developed in these studies was based on assumptions of perfect mixing, constant mass of holdup, and plate efficiencies independent of time. This model, however, is not restricted by the frequently used assumption of constant molal overflow. Since the resulting differential equations are nonlinear, numerical integration on a digital computer was used to generate the predicted column dynamic response.

Distillation dynamics and control have attracted considerable attention in both academic and industrial circles. Complete reviews of the literature on distillation dynamics and control have been presented by Archer and Rothfus (*3*), Williams (*4*), and Buckley (*5*).

Rosenbrock (*6* to *8*) made considerable progress toward a general mathematical representation of transient distillation and the corresponding solution techniques associated with such complex systems of differential equations. Utilizing a model based on constant molal overflow, Rosenbrock presented a comparison of calculated and experimental results.

G. P. Distefano is with Electronic Associates, Inc., Princeton, New Jersey. C. E. Huckaba is at Drexel Institute of Technology, Philadelphia, Pennsylvania.