Flow of Molten Poly(ethylene terephthalate) Through Packed Beds of Glass Beads

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A considerable amount of work has been done on the flow of fluids through porous media, but this work has been largely limited to the flow of common Newtonian fluids. Several papers have been published recently, however, concerning experimental investigations of the flow of polymeric systems through porous media (*1, 5, 6, 9, 10, 13, 14, 16, 18, 20, 21, 23, 24*). Of the experimental work noted, only that of Griskey and Gregory (*13*) and Siskovic, Gregory, and Griskey (*21*) considered flows of molten polymers.

Because the complex geometry of porous media prevents a detailed, easily applied solution of the equation of motion, correlative techniques are usually employed for relating pressure drop across a system to flow rate through the system. Bird and Sadowski (*1*) and Sadowski (*20*) correlated a friction factor with a modified Reynolds number based on Darcy's law and the Ellis model for nonNewtonian fluids. A similar approach was used by Christopher and Middleman (*5*) and Marshall and Metzner (*17*) except they used a power-law model for the polymer solutions evaluated. Griskey and Gregory (*13*) used the "capillary" model representation of the porous medium and developed a friction-factor-modified Reynolds number correlation based on the Mooney-Rabinowitsch equation for the flow of molten polyethylene through packed beds of glass beads. Siskovic, Gregory, and Griskey (*21*) used an approach similar to that of Christopher and Middleman (*5*) and of Marshall and Metzner (*17*) for molten polyethylene.

In this investigation, the capillary model representation of the porous medium was used in correlating the flow of molten poly(ethylene terephthalate) (PET). PET was chosen because of its availability and because its rheological behavior was well established (*11, 12*). Calculations of shear rate in the packed bed based on an estimate given by Christopher and Middleman (*5*) as

$$\dot{\gamma}_w = \frac{3n+1}{4n}\,\frac{12\,V_0}{(150\,K\epsilon)^{1/2}} \tag{1}$$

where

$\dot{\gamma}_w$ = shear rate, t^{-1}
n = flow behavior index, dimensionless
K = permeability, L^2
V_0 = superficial velocity, L/t
ϵ = porosity, dimensionless

and the fact that PET acts as a Newtonian fluid for shear stresses under 14 lb./sq.in. (*11, 12*) indicate that a Newtonian fluid model should be applicable for this investigation of the flow of molten PET through packed beds of glass beads. Therefore, a standard friction-factor-modified Reynolds number plot should correlate the experimental data and verify the well-known Blake-Carman-Kozeny (BCK) equation (*2, 3*).

$$V_0 = \frac{\Delta P D_p^2 \epsilon^3}{180\,L\mu(1-\epsilon)^2} \tag{2}$$

where

ΔP = pressure drop across packed bed, M/Lt^2
L = depth of packed bed, L
D_p = diameter of particle in packed bed, L
μ = viscosity, M/Lt

The friction-factor-modified Reynolds number plot is defined so that

$$f^* = \frac{180}{N_{Re}} \tag{3}$$

where the friction factor f^* is defined by Ergun (*8*) as

$$f^* = \frac{\Delta P}{L}\,\frac{D_p}{\rho V_0^2}\,\frac{\epsilon^3}{(1-\epsilon)} \tag{4}$$

where ρ is the density of fluid M/L^3.
The usual modified Reynolds number is (*2*)

$$N_{Re} = \frac{D_p V_0 \rho}{\mu(1-\epsilon)} \tag{5}$$

EXPERIMENT

PET produced by standard procedures (*15, 19*) was fed by gravity to a laboratory 1.5-in.-diam. melt extruder equipped with a gear pump. Molten PET was pumped through a packed bed of glass beads until the pressure drop and melt temperature were constant with time, thus assuring steady-state operation. Reynolds numbers ranged from 2×10^{-7} to 7×10^{-6}. Inherent viscosities (I.V.) of the PET used ranged from 0.3 to 0.6. Polymer melt temperatures were in the range from 290°C. to 328°C. Pressure drops ranged to 6000 lb./sq.in.gauge. Flow rates were in the 1- to 10-lb./hr. range.

Glass beads used in this experiment were obtained from Microbeads Division, Cataphote Corporation. The following sizes were used:

Mesh	Diam., in.
20-30	0.0328-0.0232
30-40	0.0232-0.0164
40-50	0.0164-0.0116
60-80	0.0097-0.0069
80-120	0.0069-0.0049

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The arithmetic average particle diameter D_p was used in subsequent calculations.

The test section used in this investigation was cylindrical, 2.12 in. in diameter by 2 in. deep. A fine mesh screen was used to retain the glass beads. Bed porosity was determined from density data supplied by the manufacturer of the glass beads and from bulk density measurements of the packed bed. Bulk density was determined by weighing the beads and measuring the occupied volume in a graduated cylinder. The same method was used to fill both the graduated cylinder and the test section.

Polymer melt temperatures were measured at the midpoint of the packed bed with an immersion type thermocouple and were recorded by a Leeds and Northrup expanded scale recorder. Melt temperature variation was $< \pm 1°C.$ after reaching steady state flow conditions. Pressure was measured 2 in. above the packed bed with a Taylor pressure transducer and was recorded. Data were taken under the same conditions for both a given bed and test section and for the test section without the bed. The pressure drop obtained for the test section was subtracted from that of the bed and test section to give the pressure drop for the bed alone.

The inherent viscosity of extrudate samples was determined by the method of Smith and co-workers (*22*), and the rheogram for the molten PET used in this investigation was based upon data reported in papers by Gregory (*11*) and Gregory and Watson (*12*).

RESULTS AND DISCUSSION

The capillary model based on the hydraulic radius concept was chosen for the correlation reported here because of its relative simplicity and because of its successful use in past correlations for Newtonian and non-Newtonian fluids, including polymer melts.

The experimental flow data were correlated using the friction factor–Reynolds number relationship given in Equation (3) and are shown in Figure 1. No trends are observed with respect to Reynolds number; however, there is considerable scatter of data points about the expected theoretical line. In Figure 2, values of pressure drop calculated from the BCK equation [Equation (2)] are shown to deviate systematically from experimental values of pressure drop, especially at test pressures above 2000 lb./sq.in.

Two possible causes of the differences between observed and calculated pressure drops investigated were viscoelastic effects and the effects of static pressure on melt viscosity. The fact that experimental pressure drops were greater than those calculated indicated the flow behavior of the molten PET was probably Newtonian, since pseudoplastic behavior would have resulted in experimental values lower than those calculated assuming Newtonian behavior.

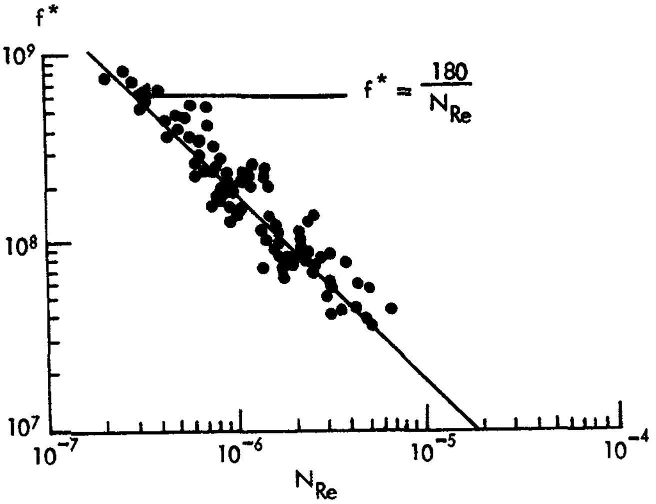

Fig. 1. Friction factor versus modified Reynolds number for glass bead data.

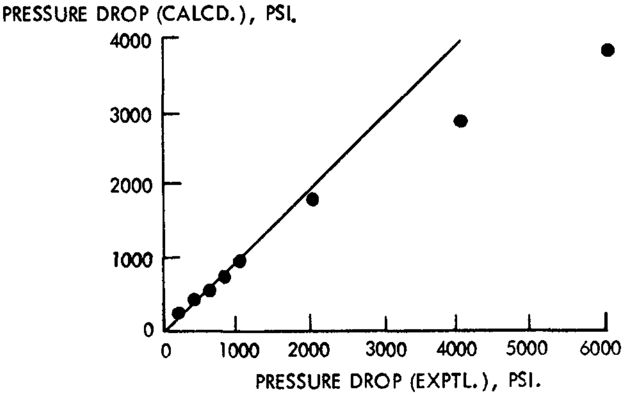

Fig. 2. Calculated versus observed pressure drop for glass beads.

The possibility of viscoelastic effects was investigated first. The measure of viscoelasticity normally used (*5, 6, 17, 21*) is the Deborah number

$$N_{Deb} = \frac{\lambda <v>}{D_p} \tag{6}$$

where

$<v>$ = average velocity, L/t
λ = relaxation time, sec.

Marshall and Metzner reported (*17*) that viscoelastic effects, evidenced by serious departures from their modified friction-factor-modified Reynolds number correlation, begin to appear at Deborah numbers of about 0.05 to 0.06 and that the pressure drop should increase to values well above those expected for purely viscous fluids at Deborah numbers of the order of 0.1 to 1.0. Christopher and Middleman (*5*), however, found no departure from a similar correlation at values of Deborah number as high as 1.0 to 1.6. Siskovic, Gregory, and Griskey (*21*) stated that the critical value of the Deborah number at which viscoelastic effects become significant for molten polyethylene is at least 0.19. Deborah numbers in this investigation ranged from 1.5×10^{-4} to 3×10^{-3}. Because the Deborah numbers are considerably less than Deborah numbers shown by others to be necessary for the onset of viscoelastic effects, we concluded that viscoelastic effects were absent and were not the cause of the increase in measured pressure drop over the calculated pressure drop.

Another possible cause of the noted behavior is the effect of static pressure on melt viscosity (*4, 7*). Carley stated (*4*) that increasing static pressure at constant pressure drop increases plastic melt viscosity for polystyrene and polyethylene. He found that his flow and static pressure data fit the exponential form given by the Eyring equation. However, they also fit a power-law function. Duvdevani and Klein found (*7*) that for the flow of molten polyethylene in capillaries the relationship between melt viscosity and static pressure was exponential and could be derived from the Eyring hole theory. The equation they derived (*7*) is

$$\frac{\mu(p)}{\mu_{at}} = e^{bp} \tag{7}$$

where

$\mu(p)$ = melt viscosity as a function of pressure, M/Lt
μ_{at} = melt viscosity measured at atmospheric pressure, M/Lt
b = constant

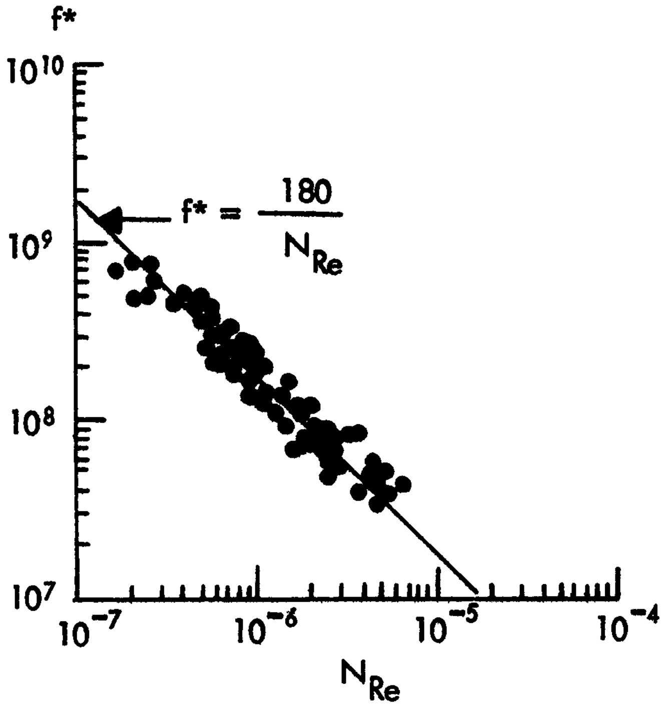

Fig. 3. Friction factor versus modified Reynolds number for glass bead data corrected for pressure effect on melt viscosity.

They found that for polyethylene b in Equation (7) was about 3×10^{-5}, depending somewhat on temperature and shear rate.

In this investigation melt viscosity was empirically made a function of pressure in order to remove the systematic differences between calculated and experimental pressure drops. Equations of both the power-law and exponential forms were fitted to the data. The exponential form gave a pressure coefficient of 5.1×10^{-5} for PET which was close to the coefficient for polyethylene reported by Duvdevani and Klein (7).

The power-law equation $\mu(p)/\mu_{at} = 0.27\ (\Delta P)^{0.2}$ rather than the exponential equation was incorporated into the BCK equation, however, so that an equation explicit in ΔP resulted:

$$\Delta P = \left[\frac{48.7\ \mu_{at} L (1-\epsilon)^2 V_0}{\epsilon^3 D_p^2} \right]^{1.25} \tag{8}$$

The power-law equation is valid for 200 lb./sq.in. $< \Delta P <$ 6,000 lb./sq.in. Use of the equation in exponential form requires iteration when solving the BCK equation for ΔP.

Figure 3 shows friction factors plotted versus modified Reynolds number using a power-law pressure-dependent melt viscosity. The scatter of data points is noticeably less than in Figure 1, where the effect of static pressure on viscosity was unaccounted for. The reduction in scatter was obviously due to the removal of the nonrandom error. Pressure drops were recalculated using the BCK equation and the pressure-dependent melt viscosity. The BCK equation with pressure-dependent viscosity predicted pressure drops within ±14% of experimental values.

As a further precaution to determine if static pressure was affecting melt viscosity, melt viscosity of PET was determined for a pressure drop of 1,000 lb./sq.in. across a capillary vented to atmospheric pressure and for a pressure drop of 1,000 lb./sq.in. for the same capillary vented to a back pressure of 4,000 lb./sq.in.gauge. Melt viscosity was about 20% higher when measured with a back pressure of 4,000 lb./sq.in.gauge than it was when measured with atmospheric back pressure. This percentage increase in melt viscosity roughly corresponds to the percentage increase necessary to account for the differences experienced in calculated and experimental pressure drops at the 4,000 lb./sq.in. level in flow through porous media.

CONCLUSIONS

The Blake-Carman-Kozeny equation was shown to describe the flow of molten PET through packed beds of glass beads under conditions used in this investigation. Calculated values of pressure drop agreed with the experimental results within an average error of 14% when melt viscosities corrected for static pressure were used.

Viscoelastic effects for flow of molten PET through the packed beds investigated were negligible.

The systematic differences between calculated and experimental values of pressure drop were shown to result from the effect of static pressure on melt viscosity. Melt viscosity of PET was empirically related to pressure drop as follows:

$$\frac{\mu(p)}{\mu_{at}} = 0.27\ (\Delta P)^{0.2}\ (200 < \Delta P < 6000 \text{ lb./sq.in.})$$

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