Evaluating Flows of Non—Newtonian Fluids by the Method of Equivalent Newtonian Viscosity

E. BROYER, C. GUTFINGER, and Z. TADMOR

Technion, Israel Institute of Technology
Haifa, Israel

Many flow problems arising in engineering design of polymer processing equipment are approximated as fully developed isothermal pressure flows in variously shaped geometrical conduits such as parallel plates, tubes, etc. (Tadmor and Klein, 1970; Pearson, 1966). The rheological behavior of the melts in such flows can often be adequately described as pseudoplastic or shear dependent, but time independent and inelastic. Every polymer melt has a different shear rate dependence. In a typical flow curve (that is, shear stress or non-Newtonian viscosity versus shear rate curves), the non-Newtonian viscosity continuously drops with increasing shear rate from an asymptotic value at low shear rates. The most commonly used non-Newtonian constitutive equation, the power law model, although of great practical value, cannot accurately represent the rheological behavior of the melt over a wide range of shear rates. It fails in particular in the low shear rate range, and since the shear rates in pressure flow extend from zero at the center to a maximum value at the wall, by using the power law model to calculate the flow rate, pressure gradient relationship, an unavoidable error, is introduced into the calculation. One obvious way to partly overcome this problem would be to employ another constitutive equation which is free from the power law model deficiency, for example, the Ellis model, which is a 3-parameter model. Such models, however, being nonlinear pose some problems regarding the statistical reliability of determining the best values of the parameters. The inaccuracies in calculating flow rates with the aid of relatively simple constitutive equations can be avoided, of course, by numerical integration of the complete experimentally determined flow curve.

This is, however, a tedious or time consuming process and it will be shown below how by defining an Equivalent Newtonian Viscosity (ENV) it can be avoided after evaluating once a set of coefficients. The method will be demonstrated on parallel plate geometry.

The flow rate per unit width q between parallel plates at a distance $2h$ apart for any velocity distribution $v(y)$ is

$$q = 2 \int_0^h v(y)dy \tag{1}$$

which upon integration by parts yields

$$q = -2 \int_0^h y \dot{\gamma}\, dy \tag{2}$$

where $\dot{\gamma}$ is the shear rate dv/dy.

For a fully developed steady viscous flow the equation of motion provides the following shear stress distribution:

$$\tau = \tau_w \frac{y}{h} \tag{3}$$

where τ_w is the shear stress at the wall proportional to the local pressure gradient:

$$\tau_w = h\left(\frac{dP}{dx}\right) \tag{4}$$

By substituting Equation (3) into Equation (2), the following expression is obtained:

$$q = -\frac{2h^2}{\tau_w^2}\int_0^{\tau_w} \tau \dot{\gamma}\, d\tau \tag{5}$$

For a Newtonian fluid

$$\tau = -\mu \dot{\gamma} \tag{6}$$

and Equation (5) reduces to

$$q = \frac{2}{3}\,\frac{h^2\tau_w}{\mu} \tag{7}$$

Equation (5) provides the true flow rate for a given pressure gradient (given τ_w) for any non-Newtonian fluid. The integration in Equation (5) can be carried out subsequent to substituting a relationship between τ and $\dot{\gamma}$, that is, the flow curve. Such a relationship can be expressed to any required degree of accuracy by polynomials, such as

$$\ln \eta = a_0 + a_1 \ln \dot{\gamma} + a_{11}(\ln \dot{\gamma})^2 + a_2 T + a_{22}T^2 + a_{12}T \ln \dot{\gamma} \quad \dot{\gamma} \geqq \dot{\gamma}_0 \tag{8}$$

$$\eta = \eta_0 \quad \dot{\gamma} \leqq \dot{\gamma}_0 \tag{9}$$

where T is the temperature and η is the non-Newtonian viscosity:

$$\eta = \frac{-\tau}{\dot{\gamma}} \tag{10}$$

and $\dot{\gamma}_0$ is a certain shear rate below which Newtonian behavior can be assumed.

The coefficients a_{ij} in Equation (8) can be obtained from the experimental flow curves, measured usually at several temperatures, by multiple linear regression analysis.

It should be noted in Equation (5) that irrespectively of the nature of the flow curve a unique relationship between q and τ_w exists. This is, of course, the basis of the Rabinowitsch correction in non-Newtonian viscometry and the definition of the apparent Newtonian shear rate (McKelvey, 1962). This unique relationship, however, further suggests that an ENV $\bar{\mu}$ can be defined which will be a function of only τ_w and the coefficients a_{ij} and which upon substitution into the Newtonian Equation (7) yields the same flow rate q for the same given τ_w as Equation (5). An expression for the ENV $\bar{\mu}$ can be obtained by comparing Equations (5) and (7) to give

$$\bar{\mu} = -\frac{\tau_w^3}{3\int_0^{\tau_w} \tau \dot{\gamma}\, d\tau} \tag{11}$$

Correspondence concerning this note should be addressed to Z. Tadmor. E. Broyer is with the Department of Chemical Engineering, University of Minnesota, Minneapolis, Minnesota 55455.

TABLE 1. COEFFICIENTS OF EQUATIONS (8) AND (14) FOR VARIOUS POLYMERS IN PARALLEL PLATE GEOMETRY*

Polymer**	Coefficients of Equation (8)						Coefficients of Equation (14)						Temp. range, °K	Coefficient of correlation
	a_0	a_1	$a_{11} \times 10^2$	$a_2 \times 10^2$	$a_{22} \times 10^4$	$a_{12} \times 10^3$	b_0	b_1	b_{11}	b_2	$b_{22} \times 10^4$	$b_{12} \times 10^3$		
Polyethylene (Marlex 5003)	22.40880	−0.93690	−2.3750	−4.3800	0.3240	1.2420	31.6464 (31.7912)	5.42902 (5.53502)	−0.40004 (−0.40894)	−0.184538 (−0.187369)	1.20139 (1.21597)	0.484272 (0.498204)	465-535	0.99578 (0.99568)
Poly(vinyl chloride) (QGPB 5135)	0.031105	0.64496	−1.7490	5.4048	−0.6480	−2.4840	−466.3570	31.3588	−0.50670	1.29255	−8.44666	−4.3857	425-475	0.99479
Nylon 66 (Zytel 42)	10.8323	−2.29856	−3.6570	3.30952	−0.6480	4.0500	74.0889	−1.29854	−0.10228	−0.158151	0.511271	0.556109	535-645	0.97424
Poly(methyl methacrylate) (Lucite 148)	42.8283	−1.66753	−2.4229	−8.55573	0.41375	2.5371	−55.2688	22.7885	−0.66152	−0.185762	3.00283	−1.59694	465-535	0.98983

* Numbers in parentheses are for tubular geometry.

** Coefficients calculated from manufacturers' data. $\bar{\mu}$(N·s/m²); η(N·s/m²); τ_w(N/m²); $\dot{\gamma}$(s⁻¹), T(°K)

Equation (11) can be written with the aid of Equation (10) as

$$\bar{\mu} = \frac{\tau_w^3}{3\int_{\dot{\gamma}_0}^{\dot{\gamma}_w}\left(\eta^2\dot{\gamma}^2 + \eta\dot{\gamma}^3\frac{\partial\eta}{\partial\dot{\gamma}}\right)d\dot{\gamma}} \tag{12}$$

Finally substituting Equations (8) and (9) into Equation (12) gives

$$\bar{\mu} = \frac{(\eta_w\dot{\gamma}_w)^3}{3\int_{\dot{\gamma}_0}^{\dot{\gamma}_w}\eta^2\dot{\gamma}^2\,(1 + a_1 + 2a_{11}\ln\dot{\gamma} + a_{12}T)\,d\dot{\gamma} + \eta_0^2\dot{\gamma}_0^2} \tag{13}$$

In order to calculate $\bar{\mu}$ from Equation (13) for any temperature and $\dot{\gamma}_w$, Equation (13) is integrated numerically subsequent to calculating η_w from Equation (8) (by inserting $\dot{\gamma}_w$ and T) and substituting Equation (8) into Equation (13). This process can be done repeatedly for a series of $\dot{\gamma}_w$ and T values, thus generating a large number of $\bar{\mu}(\dot{\gamma}_w, T)$ values. The functional relationship between $\bar{\mu}$ and $\dot{\gamma}_w$ (or $\tau_w = -\eta_w\dot{\gamma}_w$) and T can once again be expressed in a simple polynomial form:

$$\ln\bar{\mu} = b_0 + b_1\ln\tau_w + b_{11}(\ln\tau_w)^2 + b_2T + b_{22}T^2 + b_{12}T\ln\tau_w \tag{14}$$

where the coefficients b_{ij} are also obtained by multiple linear regression analysis. This calculation procedure, for a given polymer melt and geometry, has to be done only once, and it can be done very easily on a digital computer.

The ENV is very convenient to use: given the pressure gradient, the shear stress at the wall is calculated from Equation (4), next the ENV is obtained from Equation (14) and then the flow rate from Equation (7) with $\bar{\mu}$ substituted for μ. Alternatively, given the flow rate Equation (7) is rewritten as

$$\ln\bar{\mu} = C_0 + \ln\tau_w \tag{15}$$

where

$$C_0 = \ln\left(\frac{2}{3}\,\frac{h^2}{q}\right) \tag{16}$$

and Equation (15) substituted into Equation (14) to result in quadratic equation for $\ln\tau_w$

$$b_{11}\ln^2\tau_w + (b_1 + b_{12}T - 1)\ln\tau_w + b_0 + b_2T + b_{22}T^2 - C_0 = 0 \tag{17}$$

and finally the pressure gradient calculated from Equation (4). All these are simple algebraic calculations.

Coefficients b_{ij} were calculated for a number of polymer melts over a wide range of temperature and a shear rate range from 1 to 10^5 s^{-1}. The coefficients of correlation, shown in the last column of Table 1, indicate the good correlation between $\bar{\mu}$ and τ_w via Equation (14), with an accuracy which is at least as good as the rheological measurements themselves.

In order to assess the effect of flow geometry on the ENV the coefficients of Equation (14) are compared for flow between parallel plates and tube. The latter can be calculated by rederiving Equation (11) for tube flow:

$$\bar{\mu} = -\frac{\tau_w^4}{4\int_0^{\tau_w}\tau^2\dot{\gamma}\,d\tau} \tag{18}$$

As shown in Table 1, the difference between the coefficients is small. This, however, should be considered only fortuitous and must not be taken as a general result. It should be noted that the ENV is a function of geometry.

The idea to use an ENV is not new. It has been previously suggested in different forms by a number of authors, for example, Alves et al. (1952) and Weeks and Allen (1962), among others. It is felt, however, that in view of the availability of digital computers today it deserves renewed attention because it combines simplicity with accuracy in engineering calculation. The present authors have used ENV in solving non-Newtonian two-dimensional flow problems (Hele-Shaw type flow) in extrusion dies (Gutfinger et al., 1974) and injection molds (Broyer et al., 1973; Broyer, 1974) with a finite element type solution (Tadmor et al., 1974). This method with Newtonian fluids simply reduces to the simultaneous solution of sets of linear algebraic equations; with non-Newtonian fluids using the power law model it would reduce to the simultaneous solution of sets of nonlinear algebraic equations. For large sets of equations this is a time consuming, nontrivial problem. The use of the ENV, however, reduced the non-Newtonian two-dimensional flow problem to the much simpler problem of repeated solutions of sets of linear algebraic equations, thus rendering the solution of two-dimensional non-Newtonian flow problems of the type considered practical for engineering calculations.

NOTATION

a_i, a_{ij} = regression constants for Equation (8)
b_i, b_{ij} = regression constants for Equation (14)
h = half gap between parallel plates, m
q = flow rate per unit width, m²/s
T = temperature, °K
v = velocity, m/s
x = coordinate, direction of flow, m
y = coordinate perpendicular to plates, m

Greek Letters

$\dot{\gamma}$ = shear rate, s^{-1}
$\dot{\gamma}_0$ = shear rate value for transition to Newtonian behavior, s^{-1}
μ = Newtonian viscosity, N s/m²
$\bar{\mu}$ = equivalent Newtonian viscosity, N s/m²
η = non-Newtonian viscosity (apparent viscosity), N s/m²
η_0 = non-Newtonian viscosity at shear rate $\dot{\gamma}_0$, N s/m²

τ = shear stress, N/m^2
τ_w = wall shear stress, N/m^2

LITERATURE CITED

Alves, G. E., D. F. Boucher, and R. L. Pigford, "Pipe-Line Design for Non-Newtonian Solutions and Suspensions," *Chem. Eng. Progr.*, **48**, 385 (1952).

Broyer, E., "Numerical Methods for the Design of Polymer Processing Equipment," Doctoral Dissertation, Technion—Israel Inst. of Tech., Israel (1974).

———, Z. Tadmor, and C. Gutfinger, "Filling a Rectangular Channel with Newtonian Fluid," *Israel J. Tech.*, **11**, 189 (1973).

Gutfinger, C., E. Broyer, and Z. Tadmor, "Analysis of a Cross Head Die with the Flow Analysis Network Method," 32nd Ann. Tech. Conf. Soc. of Plastics Engrs, pp. 339-343, San Francisco (1974).

McKelvey, J. M. *Polymer Processing*, p. 75, Wiley, New York (1962).

Pearson, J. R. A., "Mechanical Principles of Polymer Melt Processing," pp. 67-126, Pergamon Press, Oxford (1966).

Tadmor, Z., E. Broyer, and C. Gutfinger, "Flow Analysis Network—A Method for Solving Flow Problem in Polymer Processing," *Polymer Eng. Sci.*, **14**, 660 (1974).

Tadmor, Z., and I. Klein, *Engineering Principles of Plasticating Extrusion*, pp. 184-308, Van Nostrand Reinhold, New York (1970).

Weeks, D. J., and W. J. Allen, "Screw Extrusion of Plastics," *J. Mech. Eng. Sci.*, **4**, 380 (1962).

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Linear Driving Force Approximations for Diffusion Controlled Adsorption in Molecular Sieve Columns

D. R. GARG and D. M. RUTHVEN

Department of Chemical Engineering
University of New Brunswick
Fredericton, New Brunswick, Canada

For systems in which the rate of mass transfer is controlled by intraparticle diffusion, theoretical prediction of the breakthrough curve for a packed bed adsorption column from the simultaneous solution of the differential fluid phase mass balance and the relevant diffusion equation requires considerable computational effort. This has led to the use of simple linear driving force expressions as an approximate representation of the mass transfer rate in diffusion controlled systems:

$$\frac{\partial \bar{\bar{q}}}{\partial t} = k_s a (q^* - \bar{\bar{q}}) \tag{1}$$

When the diffusivity is independent of concentration, it has been shown (Glueckauf, 1955) that Equation (1) with $k_s \approx 15\ D_e/R^2$ provides a good approximation for many boundary conditions. However, this approximation is not valid for adsorbents such as molecular sieves in which the diffusivity is strongly concentration dependent, and the purpose of this note is to develop the equivalent expressions for such systems.

For an isothermal plug flow system with equilibrium governed by a Langmuir isotherm and the mass transfer rate controlled by external fluid film resistance, the breakthrough curve for a step change in feed concentration at time zero is given by the solution of the following set of equations:

$$v \frac{\partial c}{\partial z} + \frac{\partial c}{\partial t} + \left(\frac{1-\epsilon}{\epsilon}\right) \frac{\partial \bar{\bar{q}}}{\partial t} = 0 \tag{2}$$

$$\frac{\partial \bar{\bar{q}}}{\partial t} = k_f a (c - c^*) \tag{3}$$

$$\frac{\bar{\bar{q}}}{q_s} = \frac{bc^*}{1 + bc^*} \tag{4}$$

Adsorption:

$$\bar{\bar{q}}(z, z/v) = 0; \quad c(z, z/v) = 0; \quad c(0, t) = c_0 \tag{5}$$

Desorption:

$$\bar{\bar{q}}(z, z/v) = q_0; \quad c(z, z/v) = 0; \quad c(0, t) = 0 \tag{6}$$

Numerical solutions have been given by Zwiebel et al. (1972) and, for the equivalent problem with a Freundlich isotherm, by Kyte (1973). The corresponding solid film linear driving force representation for intraparticle diffusion control is obtained by replacing Equation (3) by Equation 1 and Equation (4) by

$$\frac{q^*}{q_s} = \frac{bc}{1 + bc} \tag{7}$$

For adsorption under constant pattern conditions (adsorption front moving with uniform velocity), Equation (2) reduces simply to

$$\phi = c/c_0 = \bar{\bar{q}}/q_0 \tag{8}$$

D. R. Garg is with Union Carbide Corporation, Tarrytown Technical Center, Tarrytown, New York 10591.