Tube Flow of Non-Newtonian Polymer Solutions: Part I. Laminar Flow and Rheological Models

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A correlation of turbulent tube-flow friction factors for non-Newtonian polymer solutions, based upon the fluid property $\tau_{1/2}$ defined in Part I, has been found to represent data taken on seven solutions of Natrosol hydroxyethylcellulose in ½- and 1-in. I.D. smooth tubes, with an average accuracy of 10.8%. Also, for these seven solutions, this correlation gives somewhat more accurate predictions of the point of transition into turbulent flow than are made by the Ryan-Johnson stability theory.

The correlation must be considered only provisional, however, because it does not simplify to the limiting case of extremely dilute solutions. Plausible extensions based on models of steady-flow non-Newtonian viscosity behavior, and other possible correlation schemes utilizing viscoelastic fluid properties, are briefly discussed.

Three kinds of mechanical behavior of polymers and polymer solutions have been studied experimentally: the shear-dependent (non-Newtonian) viscosity η; the normal stress (Weissenberg) effect, described by the normal stress coefficient ζ; and the transient response to small amplitude displacements which can be described by the complex viscosity η^*. Recently it has been proposed (*24, 25, 26*) that the transient response of the normal stresses should also receive experimental attention.

One of the primary aims of rheology is to formulate constitutive equations capable of describing all of the above phenomena and their interrelations. To this end some very general and elegant equations have been proposed. Despite their all-inclusiveness they may not necessarily offer the most judicious description of systems encountered in engineering applications. For many applications an adequate description of a viscoelastic fluid is provided by the generalized Newtonian model (*5, 18*)

$$\underline{\tau} = -\eta \, \underline{\Delta} \qquad (1)$$

in which η is a function of the second invariant of the viscous momentum flux tensor $\underline{\tau}$ or the rate-of-deformation tensor $\underline{\Delta} = \nabla \mathbf{v} + (\nabla \mathbf{v})^\dagger$.

The non-Newtonian viscosity η appearing in Equation (1) can usually be curve fitted with a function containing a small number of constant parameters. One then states that the mechanical behavior of the fluid is characterized by specifying values of these parameters. The use of Equation (1) with a curve-fit function for η has been studied rather extensively at this and other laboratories for several fluids and flow systems; the following facts seem to emerge regarding the above method:

1. The method seems to be adequate for describing steady state laminar flow in a variety of systems; that is when the model parameters are determined in one geometry, the same parameters are able to describe the flow of the same fluid in a different geometry (*17, 10, 19, 20, 23*).
2. The method seems to be adequate for describing unsteady state laminar flow provided that the dominant time constant of the system is larger than the dominant time constant for the fluid (*2*).
3. The method provides a small number of parameters which can be used for the preparation of dimensionless correlations for steady flow in geometrically complex systems (*20, 21, 22*).
4. The parameters in the non-Newtonian viscosity function may be useful for correlating some turbulent flow phenomena (*12*).
5. The method is probably quite adequate for solving steady state heat transfer problems (*1*).
6. The generalized Newtonian model is easily adapted to the use of variational methods for obtaining analytical solutions to complex flow problems (*13, 3, 9, 27*).

Hence, although the generalized Newtonian models cannot possibly describe normal stresses or small amplitude transient response, they are nonetheless sufficiently important to warrant their further exploration and use. In this and the following paper the authors present briefly some new results regarding laminar and turbulent tube flow of polymers and polymer solutions.

GENERAL CHARACTER OF NON-NEWTONIAN VISCOSITY CURVES

Polymers and polymer solutions seem to exhibit the same general behavior with regard to the non-Newtonian viscosity η as a function of the shear stress τ. In the limit of very small shear stress, the viscosity approaches a lower limiting viscosity η_0 (unless, of course, a gel structure is formed). With increasing shear stress the viscosity η decreases, and apparently if the shear stress can be increased sufficiently the viscosity becomes constant at an upper

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TABLE 1. VALUES OF η_0, η_∞, τ_m, α, AND $\tau_{1/2}$ FOR NATROSOL TYPE H SOLUTIONS*

Concentration, %	Parameters in Equation (2) η_0 (poise)	η_∞, (poise)	τ_m, (dyne cm.$^{-2}$)	α, (- - -)	$\tau_{1/2}$, (dyne cm.$^{-2}$)
0.3	0.23	0.0122	78	2.02	92
0.5	1.09	0.0150	76	2.19	81
0.7	3.86	0.0185	106	2.54	104
1.0	20.3	0.025	173	3.10	173

* Values taken from D. M. Meter, Doctoral dissertation, University of Wisconsin, Madison, Wisconsin (1963).

limiting viscosity η_∞. Hence η_0 and η_∞ are measurable quantities characteristic of the fluid.

Another measurable quantity is τ_m, the shear stress at which the viscosity has dropped to $\frac{1}{2}$ $(\eta_0 + \eta_\infty)$. For some polymers and solutions $\eta_\infty << \eta_0$ so that τ_m is essentially equal to $\tau_{1/2}$, the shear stress at which the viscosity has dropped to $\frac{1}{2}$ η_0. As will be seen presently, τ_m and $\tau_{1/2}$ turn out to be useful quantities for characterizing non-Newtonian fluids.

It should be emphasized that η_0, η_∞, and τ_m (or $\tau_{1/2}$) are usually measurable properties, independent of any assumed model of the fluid. It is therefore reasonable to try to introduce these quantities into any proposed empirical models. However, it may be that the procedure used to determine model parameters is such that the quantities η_0, η_∞, and τ_m (or $\tau_{1/2}$) are merely the values which give the best curve fit over the range of available data, and that they may not quite have their intended physical significance. Such would certainly be true in the case of a fluid which forms a gel structure when not in motion; then there is no lower limiting viscosity and a $\tau_{1/2}$ would be impossible to determine.

A FOUR-CONSTANT MODEL*

An analytical expression which contains three of the above-mentioned measurable quantities is that suggested by Meter (*11*):

$$\eta = \eta_\infty + \frac{\eta_0 - \eta_\infty}{1 + |\tau/\tau_m|^{\alpha-1}} \tag{2}$$

This model contains the η_0, η_∞, and τ_m described above, and in addition an exponent α which indicates the abruptness of the transition from η_0 to η_∞. Note that η_0/τ_m and η_∞/τ_m are two characteristic times of the fluid. Since η_∞ is often much smaller than η_0, Equation (2) can be rewritten as

$$\frac{\eta_0}{\eta} = [1 + |\tau/\tau_m|^{\alpha-1}] \sum_{j=0}^{\infty} [- |\tau/\tau_m|^{\alpha-1} (\eta_\infty/\eta_0)]^j \tag{3}$$

by rearranging and then expanding in powers of η_∞/η_0.

As an example of the use of Equation (2) in fitting non-Newtonian viscosity data, data on four aqueous solutions of Natrosol 250, Type H (hydroxyethyl cellulose, high viscosity grade) were analyzed. The fluids were studied in a capillary-tube viscometer and in a pipe-flow apparatus (*11*), the measured quantities being the pressure drop and the mass rate of flow. Use of the Weissenberg-Rabinowitsch equation (*16, 5*) then gave η as a function of τ, as shown in Figure 1. In addition the low shear limiting value η_0 was found by observing the rate of fall of tiny glass beads through a column of fluid. The upper limiting viscosity η_∞ was estimated from the η vs. τ curves. Then τ_m and α were adjusted by trial-and-error until a reasonable fit of the experimental curve was obtained. The values of the parameters thus obtained are presented in Table 1. Values of $\tau_{1/2}$ are also given, that is the values of τ for which $\eta = \eta_0/2$. Further details are given elsewhere (*11*).

As can be seen from Figure 1, the curve fitting of the data is quite satisfactory, the only significant deviations being in the high shear-stress region. The average absolute percentage deviations of the curve fit from the experimental values at sixty-six representative points (nearly equally spaced along the logarithmic τ axis) is 4.6%. Such agreement is satisfactory for most purposes.

The parameters listed in Table 1 show that both η_0 and η_∞ decrease with decreasing polymer concentration, tending to the (Newtonian) viscosity of the solvent at infinite dilution. The parameter α also decreases with decreasing concentration, whereas τ_m appears to approach a definite value, characteristic of the polymer, as infinite dilution is approached.

RELATION TO OTHER EMPIRICAL MODELS

The model in Equation (2) may be thought of as an extension of the Peek-McLean model ($\alpha = 2$) (*14*) and the Reiner-Philippoff model ($\alpha = 3$) (*18, 15*).

Fluids for which $\eta_\infty << \eta_0$ can be approximated by Equation (2) with $\eta_\infty = 0$, or by Equation (3) with only the $j = 0$ term in the summation. Also, τ_m becomes $\tau_{1/2}$, so that

$$\eta = \frac{\eta_0}{1 + |\tau/\tau_{1/2}|^{\alpha-1}} \tag{4}$$

The resulting simple model is one attributed to Ellis (*18*), but it is here written in a form which gives more emphasis to the physical meaning of the parameters. It has been used by Gee and Lyon (*7*) in their study of viscous heating of molten polyethylene in extrusion in tubes, by Slattery and Bird (*21*) and by Turian (*23*) in the study of flow of polymer solutions around spheres, by Sadowski (*19*) to describe non-Newtonian flow in porous media,

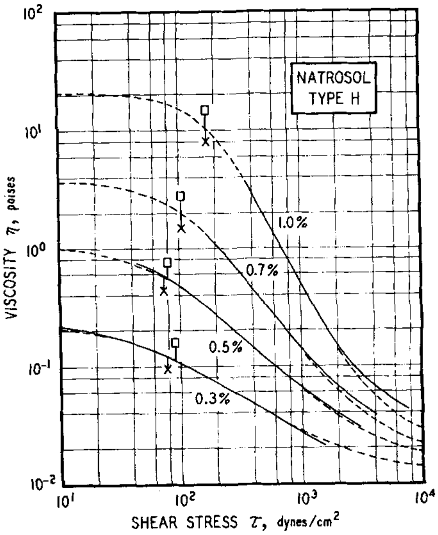

Fig. 1. Non-Newtonian viscosities of Natrosol type H solutions, as determined by Meter (*11*). Solid line: experimental values. Dashed line: curve fit with Equation (1) with parameters listed in Table 1. Values of the model parameter τ_m are marked with X's, whereas values of the fluid property $\tau_{1/2}$ are indicated by squares.

* The various models presented here are given in terms of the shear stress τ; this τ could be τ_{rz} for the flow, τ_{xy} for slit flow, etc. To generalize the formulas for use in more complex flow systems one can simply replace $|\tau|$ by $+\sqrt{\frac{1}{2}(\underline{\tau}:\underline{\tau})}$ as suggested by Hohenemser and Prager (*8*).

$$f = \frac{128}{\left(\frac{D^2 \tau_m \rho}{\eta_0^2}\right)\left(\frac{\tau_w}{\tau_m}\right)\left\{\left[1 + \frac{4}{\alpha+3}\left(\frac{\tau_w}{\tau_m}\right)^{\alpha-1}\right] - \left(\frac{\eta_\infty}{\eta_0}\right)\left(\frac{\tau_w}{\tau_m}\right)^{\alpha-1}\left[\frac{4}{\alpha+3} + \frac{2}{\alpha+1}\left(\frac{\tau_w}{\tau_m}\right)^{\alpha-1}\right] + \ldots\right\}^2} \quad (8)$$

and by Bird (*4*) in a study on heat transfer in circular tubes. There is some evidence (*2*), although not enough to be conclusive, that the characteristic time $\eta_0/\tau_{1/2}$ can be used to delineate the limits of application of a generalized Newtonian model; apparently if the characteristic time of the macroscopic flow system is substantially greater than $\eta_0/\tau_{1/2}$, then it is not necessary to include time derivatives in the constitutive equation. Since this model can reproduce experimental data over a moderately wide range, and since it does contain a lower limiting viscosity, it may be the most useful rheological model for many engineering applications.

At high shear stresses, Equation (4) has power law behavior. It is well known that many polymers and polymer solutions do approximate such behavior; that is log η vs. log τ is linear over a wide range of shear stress. The power law model of Ostwald and de Waele

$$\eta = m^\alpha |\tau|^{1-\alpha} \quad (5)$$

with constants m and α does seem to describe a great variety of polymeric behavior over a wide range of shear stress; it cannot, of course, describe the lower-limiting viscosity η_0. When the constants m and α are obtained from analysis of flow in one geometry, the same constants usually cannot be used to describe flow in another geometry (*6, 10, 19, 20*). Furthermore, the model constants m and α cannot be combined in order to form a characteristic time for the fluid.

LAMINAR TUBE FLOW

Consider the flow of the fluid in Equation (2) in a circular tube of radius $R = D/2$, under the influence of a pressure difference Δp over a tube length L. The shear stress at the wall $r = R$ is $\tau_w = \Delta p R/2L$. When the four-constant model in the form of Equation (3) is used, one obtains by standard methods the following velocity distribution as a power series in the usually small quantity η_∞/η_0:

$$\frac{v_z}{\tau_w R/2\eta_0} = \left[(1-\xi^2) + \left(\frac{2}{\alpha+1}\right)\left(\frac{\tau_w}{\tau_m}\right)^{\alpha-1}(1-\xi^{\alpha+1})\right] - \left(\frac{\eta_\infty}{\eta_0}\right)\left(\frac{\tau_w}{\tau_m}\right)^{\alpha-1}\left[\left(\frac{2}{\alpha+1}\right)(1-\xi^{\alpha+1}) + \frac{1}{\alpha}\left(\frac{\tau_w}{\tau_m}\right)^{\alpha-1}(1-\xi^{2\alpha})\right] + O\left[\left(\frac{\eta_\infty}{\eta_0}\right)^2\right] \quad (6)$$

A further integration then gives the volume rate of flow Q:

$$\frac{Q}{\pi R^3 \tau_w/4\eta_0} = \left[1 + \frac{4}{\alpha+3}\left(\frac{\tau_w}{\tau_m}\right)^{\alpha-1}\right] - \left(\frac{\eta_\infty}{\eta_0}\right)\left(\frac{\tau_w}{\tau_m}\right)^{\alpha-1}\left[\frac{4}{\alpha+3} + \frac{2}{\alpha+1}\left(\frac{\tau_w}{\tau_m}\right)^{\alpha-1}\right] + O\left[\left(\frac{\eta_\infty}{\eta_0}\right)^2\right] \quad (7)$$

The latter result can be arranged to give an expression for the dimensionless friction factor ($f = R\,\Delta p/L\rho \langle v_z \rangle^2$):

Hence, the friction factor depends on four dimensionless groups: $(D^2\tau_m\rho/\eta_0^2)$, (τ_w/τ_m), (η_∞/η_0), and α.

The results in Equations (6), (7), and (8) may be easily modified for the Ellis model by setting η_∞/η_0 equal to zero and replacing τ_m by $\tau_{1/2}$.

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NOTATION

D = diameter of tube
f = friction factor
L = length of tube
m = parameter in power law
p = pressure
Q = volume flow rate
r = radial coordinate
R = radius of tube
v = velocity
$\langle v_z \rangle$ = average velocity in tube flow
∇ = nabla operator
$\dagger$ = transpose of a tensor

Greek Letters

α = parameter in several rheological models
$\underline{\Delta}$ = rate of deformation tensor
η = non-Newtonian viscosity
η^* = complex viscosity
η_0 = lower limiting viscosity
η_∞ = upper limiting viscosity
ξ = dimensionless radial coordinate ($= r/R$)
τ = scalar value of shear stress
$\underline{\tau}$ = shear stress tensor
τ_m = parameter in Equation (2)
$\tau_{1/2}$ = parameter in Equation (4)
τ_w = shear stress at tube wall

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Tube Flow of Non-Newtonian Polymer Solutions: Part II. Turbulent Flow

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The correlation must be considered only provisional, however, because it does not simplify to the limiting case of extremely dilute solutions. Plausible extensions based on models of steady-flow non-Newtonian viscosity behavior, and other possible correlation schemes utilizing viscoelastic fluid properties, are briefly discussed.

The importance of the viscoelastic properties of fluids in determining their turbulent flow behavior has been suggested by Atkinson (*1*), Ward (*22*), and more recently by Dodge and Metzner (*4*). Recent studies (*10, 14*) have indicated, however, that for many polymer solutions the normal stresses in the two directions perpendicular to the direction of flow are equal, and furthermore (*12, 23*) that for such fluids a definite correspondence exists between viscoelastic and steady flow non-Newtonian viscosity properties. The primary purpose of this paper is to suggest ways in which non-Newtonian viscosity information may be used to correlate turbulent friction factor data for the tube flow of viscoelastic polymer solutions. It is felt that this approach is of value, even though it is not appropriate for extension to other types of non-Newtonian fluids, such as soft gels and suspensions.

DIMENSIONAL ANALYSIS OF TUBE FLOW FOR PSEUDOPLASTIC POLYMER SOLUTIONS

A four-constant model of pseudoplastic behavior, applicable to polymer solutions, has been proposed in Part I (*13*). There the integration of this model for laminar flow in circular tubes shows that the friction factor f is dependent upon the following four dimensionless groups, which include the four model parameters:

$$f = f\left(\frac{D^2 \tau_m \rho}{\eta_0^2}, \frac{\tau_w}{\tau_m}, \alpha, \frac{\eta_\infty}{\eta_0}\right) \tag{1}$$

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It is to be expected that the same dimensionless groups will be important in turbulent flow, though, of course, the exact relationship between the groups will be different from that in laminar flow.

None of the groups in Equation (1) is a Reynolds number. A Reynolds number based upon this model may be defined as the group $(D <v_z> \rho)$ divided by the tube-apparent viscosity, or ratio $(\tau_w)/(8 <v_z>/D)$ of laminar-flow consistency variables; it is

$$N_{Re} = \frac{D <v_z> \rho}{\eta_0} \left\{ \left[1 + \frac{4}{\alpha + 3}\left(\frac{\tau_w}{\tau_m}\right)^{\alpha-1} \right] - \left(\frac{\eta_\infty}{\eta_0}\right)\left(\frac{\tau_w}{\tau_m}\right)^{\alpha-1} \left[\frac{4}{\alpha+3} + \frac{2}{\alpha+1}\left(\frac{\tau_w}{\tau_m}\right)^{\alpha-1} \right] + \ldots \right\} \tag{2}$$

Note that this definition forces evaluation of the tube-apparent viscosity at the prevailing wall shearing stress, whether the flow is laminar or turbulent. If N_{Re} above were to be used, only three other dimensionless groups would be needed to specify f; probably α, τ_w/τ_m, and η_∞/η_0 would be chosen.

A priori, it would be expected that the group η_∞/η_0 would not be important except at extremely high shear stresses where the upper limiting viscosity of the fluid is being approached.

The Reynolds number of Equation (2) yields the relation $f = 16/N_{Re}$ for the laminar region; for turbulent flow an f vs. N_{Re} relation would require the introduction of additional dimensionless groups as indicated above. Investigations (*4, 12*) with a Reynolds number, along with