J. Crank and G. S. Park, Academic Press, London, 1968, p 142
4 Kokes, R. K., Long, F. A., and Hoard, J. C. *J. Chem. Phys.* 1952, **20**, 1704
5 Bagley, E. and Long, F. A. *J. Amer. Chem. Soc.* 1955, **77**, 2172
6 Long, F. A. and Richman, D. *J. Amer. Chem. Soc.* 1960, **82**, 513
7 Park, G. S. *J. Polym. Sci.* 1953, **11**, 97
8 Chernova, I. V. and Vasenin, R. M. *Vysokomol. Soedin.* 1964, **6**(9), 1704
9 Zhurkov, S. N. and Ryskin, G. Ya., *Zh. Tekh. Fiz.* 1954, **24**, 797–810
10 Ryskin, G. Ya. *Zh. Tekh. Fiz.* 1955, **25**, 458
11 Frensdorff, H. K. *J. Polym. Sci. (A)*, 1964, **2**, 341

Parameters affecting the onset of drag reduction in polymeric solution

R. S. Sharma*, V. Seshadri and R . C. Malhotra
Department of Applied Mechanics, Indian Institute of Technology, Delhi-110 029, India

(Received 29 March 1978; revised 8 September 1978)

INTRODUCTION

Studies on the drag reduction phenomenon in polymeric solutions indicate the existence of a threshold level of shear stress which must be exceeded before the drag reduction manifests itself. This effect is known as the onset of drag reduction and is influenced by both 'flow' and 'polymeric' parameters. White[1], and Vleggaar, Dammers and Tels[2] found analytically that the Reynolds number corresponding to the inception of drag reduction is proportional to the pipe diameter raised to the power of 8/7. However, the results of these studies do not explain all the experimental results obtained with different polymers[3,4].

In the present note, the model due to Denn has been adapted to represent the viscoelastic properties of the drag-reducing fluid. Results obtained indicate that the onset Reynolds number is a function of the pipe diameter, the type of polymer and its concentration, as well as the velocity gradient near the wall. Experimental results available in the literature have been explained qualitatively using the present analysis. Further, it is shown that the result obtained by Vleggaar *et al.*[2] forms a particular case of the present study.

ANALYSIS

A drag-reducing fluid is considered as a viscoelastic one possessing the Newtonian viscosity. Such a fluid fits the Denn model[5], represented by the following rheological equation:

$$\tau^{ij} + \tau\,(\mathrm{II}/2)^{(s-2)/2}\,\frac{\delta\tau^{ij}}{\delta t} = 2\mu e^{ij} \qquad (1)$$

where τ^{ij}, e^{ij} are the components of the deviatoric stress tensor and the rate of strain tensor, respectively. $\delta/\delta t$ represents the convected derivative and I is an invariant of e^{ij}. Further, μ,τ and s are material constants.

For one dimensional flow, equation (1) reduces to

$$\left[1 + \tau\,(\mathrm{II}/2)^{(s-2)/2}\,\frac{\mathrm{d}}{\mathrm{d}t}\right]\sigma(t) = \mu\frac{\mathrm{d}}{\mathrm{d}t}\,\gamma(t) \qquad (2)$$

where $\bar{\tau}(\mathrm{II}/2)^{(s-2)/2}$ represents the relaxation time of the fluid related to the exponentail stress decay at a constant strain. II for established flow is $4(\mathrm{d}V_z/\mathrm{d}y)^2$, where V_z is the bulk flow velocity in z-direction. For periodic strain of the type

$$\gamma = \gamma_0 \exp(i\omega t\,)$$

the stationary solution of equation (2) is

$$\sigma(t) = \gamma_0\,\frac{\mu}{\tau(1/2\mathrm{I})^{1/2(s-2)}}\left[\frac{i\omega\tau(\mathrm{II}/2)^{(s-2)/2}}{1 + i\omega\tau(\mathrm{II}/2)^{(s-2)/2}}\right]\times \exp(i\omega t) \qquad (3)$$

for $t \gg \tau\,(\mathrm{II}/2)^{(s-2)/2}$

Introducing the complex relation modulus

$$G^*(\omega) = G'(\omega) + iG''(\omega) = \sigma(t)/\gamma(t)$$

one obtains

$$G'(\omega) = G\left[\frac{\omega^2\tau^2(\mathrm{II}/2)^{(s-2)}}{1 + \omega^2\tau^2(\mathrm{II}/2)^{(s-2)}}\right]$$

$$G''(\omega) = G\left[\frac{\omega\tau(\mathrm{II}/2)^{(s-2)/2}}{1 + \omega^2\tau^2(\mathrm{II}/2)^{(s-2)}}\right]$$

$$G'(\omega)/G''(\omega) = \omega\tau(\mathrm{II}/2)^{(s-2)/2} \qquad (4)$$

where

$$G = (\mu/\tau)\,(\mathrm{II}/2)^{(2-s)/2}$$

---

* Present address: Chemical Engineering Department, The Ohio State University, 140 West 19th Avenue, Columbus, Ohio 43210, USA.

0032–3861/79/020257–03$02.00

The storage modulus $G'(\omega)$ is proportional to the energy stored and recovered in the elastic response of the fluid to the periodic strain, while the loss modulus $G''(\omega)$ is proportional to the energy dissipated into heat through the viscous behaviour of the fluid[6]. Equation (4) shows that the elastic properties of the fluid become more and more dominant as $\omega\tau(\mathrm{II}/2)^{(s-2)/2}$ increases, thereby reducing the viscous dissipation.

The dissipation of kinetic energy into heat in the case of turbulent flow of Newtonian fluids occurs *via* the small-scale eddies, the larger eddies being non-dissipative. On the other hand, for viscoelastic fluids the energy of the high frequency disturbances associated with the small-scale eddies is preferentially converted into recoverable elastic energy[7]. This process may be responsible for the reduced friction drag in the turbulent flow of viscoelastic fluids.

It may be noted from equation (4) that the viscoelastic behaviour of a drag-reducing fluid is closely related to the value of the dimensionless quantity $\omega\tau(\mathrm{II}/2)^{(s-2)/2}$. This quantity can also be written as $\tau(\mathrm{II}/2)^{(s-2)/2}/(1/\omega)$. Following Landau and Lifshitz[8], $(1/\omega)$ can be interpreted as the characteristic flow time. Then $\omega\tau(\mathrm{II}/2)^{(s-2)/2}$ can be considered as the ratio of the relaxation time of the process to the characteristic flow time, and consequently, takes the form of the Deborah number.

According to Hershey and Zakin[9], the Deborah number at the onset of drag reduction is near unity. For the sake of generality it is assumed that, for drag-reducing fluid, the Deborah number at the onset of drag reduction is a constant. Thus,

$$[\omega\tau(\mathrm{II}/2)^{(s-2)/2}]_{\mathrm{onset}} = \text{constant} \tag{5}$$

Again, in pipe flow, following Landau and Lifshitz[8], the magnitude of the frequency corresponding to the small-scale eddies is given by

$$\omega \sim \frac{v}{D} Re^{3/4}, \text{ with } Re = \frac{\rho v D}{\mu} \tag{6}$$

where D is the pipe diameter, v is the average fluid velocity, ρ and μ are the density and viscosity of the drag-reducing fluid, respectively. For a given fluid of constant material properties, it can be shown, from equations (5) and (6), that

$$Re_{\mathrm{onset}} \propto D^{8/7} (\mathrm{II}/2)^{2(s-2)/7}$$

On substituting $\mathrm{I} = 4(\mathrm{d}V_z/\mathrm{d}y)^2$, the above expression reduces to

$$Re_{\mathrm{onset}} \propto D^{8/7} \left(\frac{\mathrm{d}V_z}{\mathrm{d}y}\right)^{(4/7)(2-s)} \tag{7}$$

RESULTS AND DISCUSSION

For the Maxwellian fluid, the value of s is 2 and therefore, equation (7) simplifies to the result obtained by Vleggaar *et al.*[2]. The parameter 's' is the gradient of the first normal stress difference with respect to the velocity gradient on a log–log plot. The magnitude of this parameter depends on the type of polymer and its concentration. The stability criterion investigated by Hasegawa and Tomita[5] leads to a maximum value of 1.62 for the parameter s. Oliver[10] determined the values of s for various drag-reducing polymeric solutions by measuring the thrust of a jet and found them to be below 1.5 for all solutions investigated. It is therefore apparent that the simulation of polymeric fluids by the Maxwell model to investigate the critical Reynolds number is not accurate and hence is likely to lead to anomalous conclusions.

It is now possible to explain the available experimental results on the onset of drag reduction. Results obtained by Oliver[10] show that, for drag-reducing additives such as guar gum, sodium carboxymethylcellulose (SCMC), the value of s remains constant over a wide range of concentration (e.g., $s = 1.02$ for 1% and 2% SCMC). Thus for a given pipe diameter, equation (7) predicts that the inception of drag reduction for such polymers would be independent of concentration. This conclusion is supported by the experimental data of Whitsitt *et al.*[3] for guar gum (J2-FP) (see *Table 1*).

Equation (7) also predicts that for a given pipe and velocity gradient, an increase in s lowers the $\mathrm{Re}_{\mathrm{onset}}$. The results of Oliver[10] show that the value of s for a solution of polyacrylamide is higher than that for guar gum solution of equal concentration. Thus equation (7) predicts that the onset of drag reduction should occur earlier in the case of polyacrylamide solution as compared to guar gum solution of same concentration. This conclusion is supported by the observations of Whitsitt *et al.*[3] as can be seen from *Table 1*. The observations of Virk and Baher[11] also show that the onset of drag reduction occurs earlier in polyacrylamide (E-198) solution as compared to polyethylene oxide (W-205)

Table 1

References	Polymer type	Concentration wppm	Pipe diameter (mm)	$\mathrm{Re}_{\mathrm{onset}}$
3	Guar gum (J2-FP)	250	4.5	11 200
	Guar gum (J2-FP)	1250	4.5	11 200
3	Polyacrylamide (AP-30)	250	4.5	6650
	Guar gum (J2-FP)	250	4.5	11 200
11	Polyacrylamide (E-198)	10	9.45	6050
	Polyethylene oxide (W-205)	10	9.45	7300
				$(\tau_w)_{\mathrm{onset}}$ (dyn/cm^2)
13	Polyethylene oxide	3.0	6.60	25
	Polyethylene oxide	8.6	6.60	20
	Polyethylene oxide	57.3	6.60	11

solution. This finding can again be attributed to a higher value of s in case of a polyacrylamide solution[10]. Recently, in contradiction to the findings of Virk *et al.*[12], Hansen and Little[13] have reported that for Polyox solution of different concentrations, the wall shear stress at the onset of drag reduction, $(\tau_w)_{\rm onset}$, decreases with increasing concentration. This result can be explained qualitatively by considering the fact that the value of s for any given polymeric solution increases with increasing concentration and hence should result in lowering of $\mathrm{Re}_{\rm onset}$.

The assumption of similar velocity profiles for different diameter pipes leads to the result that the critical Reynolds number is a single parametric function of pipe diameter with an exponent of 8/7. Experimental results of Elata and Tirosh[14], White[1], and Hershey and Zakin[9] confirm this conclusion. The experimental data obtained by Sharma[15] with solutions of various concentrations (up to 2000 ppm weight) of hydroxylpropylmethyl cellulose (Trade Name: Methofas, manufactured by ICI, UK) in different diameter pipes indicate similar diameter and concentration effects at the threshold of drag reduction.

Equation (7) indicates a possibility of controlling the onset of drag reduction by suitably adjusting the governing parameters. For a given type of polymer and its concentration in the solution, the drag reduction will occur earlier as the size of the pipe decreases. For given D and s, the threshold of drag reduction can be made to occur earlier by decreasing the velocity gradient in the region of maximum dissipation of small-scale eddies. Thus the inception of drag reduction occurs earlier as the velocity profile in the wall region becomes flatter. One possible method to achieve this may be by injecting the concentrated solution of drag-reducing polymer next to the wall. In this connection it may be noted that addition of drag-reducing polymers results in thickening of the viscous sublayer which in turn tends to reduce the velocity gradient in the wall region.

REFERENCES

1 White, A. *J. Mech. Eng. Sci.* 1966, **8**, 452
2 Vleggaar, J., Dammers, W. R. and Tels, M. *Chem. Eng. Sci.* 1968, **23**, 1159
3 Whitshitt, N. F., Horrington, L. J. and Crawford, H. R. 'Effect of Wall Shear Stress on Drag Reduction of Viscoelastic Solutions', in Viscous Drag Reduction, (Ed. C. S. Wells) Plenum Press, New York, 1969, p 256
4 Virk, P. S. and Merrill, E. W., 'The Onset of Dilute Polymer Solution Phenomenon', in Viscous Drag Reduction, (Ed. C. S. Wells), Plenum Press, New York, 1969, p 107
5 Hasegawa, T. and Tomita, Y. *Bull. Jpn Soc. Mech. Engrs.*, 1974, **17**, 73
6 Ferry, J. D., 'Experimental Techniques for Visoelastic Bodies', in Rheology, (Ed. F. R. Eirich), Academic Press, 1958
7 Patterson, G. K. and Zakin, J. L. AIChE J., 1968, **14**, 434
8 Landau, L. D. and Lifshitz, E. M. 'Fluid Mechanics', Pergamon Press, 1963
9 Hershey, H. C. and Zakin, J. L. *Chem. Eng. Sci.* 1967, **22**, 1847
10 Oliver, D. R. *Can. J. Chem. Eng.*, 1966, **14**, 100
11 Virk, P. S. and Baher, H. *Chem. Eng. Sci.* 1970, **25**, 1183
12 Virk, P. S., Merrill, E. W., Mickley, H. S., Smith, K. A. and Mollo-Christensen, E. L. *J. Fluid Mech.* 1967, **30**, 305
13 Little, R. C., Hansen, R. J., Hunston, D. L., Kim, oh-Kil, Patterson, R. L. and Ting, R. Y., *Ind. Eng. Chem. Fund.* 1975, **21**, 625
14 Elata, C. and Tirosh, J. *Israel J. Tech.* 1966, **4**, 87
15 Sharma, R. S., Ph. D. Thesis, Indian Institute of Technology, New Delhi, 1977

Concentration dependence of the diffusion coefficient of polystyrene at theta-condition

Charles C. Han
Center for Materials Science, National Bureau of Standards, Washington, DC 20234, USA

(Received 30 August 1978)

INTRODUCTION

Despite many experimental and theoretical studies, the concentration dependence of diffusion coefficient, k_D, of polymers at their theta-conditions still remain an unsettled problem. Many investigators found experimentally that k_D has a negative value[1–7]; furthermore, Cantow[8] found that the diffusion coefficient D is independent of the concentration at theta temperature. Meanwhile, theoretical investigations have been carried out by Yamakawa[9,10] and Imai[11] and also by Pyun and Fixman[12]. The Pyun and Fixman (P–F) approach uses a spherical model instead of the more realistic beads and spring model which was employed by Yamakawa and Imai (Y–I). But it has been pointed out by Pyun and Fixman that Yamakawa's earlier approach is incorrect due to the approximation made by assuming independent inter- and intramolecular distributions and consequently resulting in the neglect of correlations due to the intermolecular interactions. In the later work of Yamakawa[10], a negative concentration dependence of D was introduced through the change of coordinate system from actual polymer velocity to the drift velocity. Even though this model leads to a different magnitude for the k_D than the one obtained from P–F model, nonetheless both theories predict a negative and molecular weight independent k_D at the theta condition.

On the other hand, Kotaka *et al.*[13] has found that in the

0032–3861/79/020259–03$02.00