On the Explanation and Correlation of Turbulent Drag Reduction in Dilute Macromolecular Solutions

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Synopsis

A simple mechanistic picture of turbulent drag reduction is proposed, based on the recent work of Corino and Brodkey for Newtonian fluids. Possible explanations are then considered from a phenomenological viewpoint, and the applicability of the well-known dumbbell model toward this end is discussed. It is seen that the predictions of the "molecular" dumbbell model are surprisingly similar to those of the "continuum" convected Maxwell model utilized by Seyer and Metzner. Furthermore, the significance of the fluid relaxation time θ in characterizing the drag-reducing effectiveness of a polymer solution is shown to follow from a number of qualitative arguments. A correlation is proposed, closely related to the recent correlations advanced by Rodriguez, Zakin, and Patterson, and Astarita, Greco, and Nicodemo. Some new aspects of this correlation are discussed.

INTRODUCTION

Turbulent drag reduction in dilute polymer solutions, also known as the Toms effect,[1] has been studied extensively in recent years. Still, it is clear that the basic explanation of the phenomenon, although generally agreed to be of a "viscoelastic" nature, is not understood. Furthermore, there remain unanswered a number of important questions concerning the existing phenomenological theories used for the correlation of turbulent drag reduction.

In the present work, a simple explanation of drag reduction is suggested, based on the recent studies of Corino and Brodkey[2] with Newtonian fluids. We then turn our attention to correlation and discuss the applicability of the "molecular" dumbbell model toward this end. A correlation is proposed and compared and contrasted with the quite similar correlations of Rodriguez, Zakin, and Patterson[3] and Astarita, Greco, and Nicodemo.[4]

A MECHANISTIC PICTURE OF DRAG REDUCTION IN DILUTE MACROMOLECULAR SOLUTIONS

In a recent significant study, Corino and Brodkey[2] made observations of the flow field in the vicinity of the wall for a Newtonian fluid in turbulent motion through a circular pipe. The main conclusion of their study, in agreement with the related work of Kline and co-workers,[5] was the sig-

nificance of periodic "ejections" or "bursts" originating in the wall region in the generation and maintenance of turbulence. For the purposes of the present paper, the importance of their study lies in the fact that these "bursts" also accounted for most of the turbulent energy dissipation.

According to Corino and Brodkey,[2] the ejected element would leave the wall region, interact with the main flow, and essentially burst or break up into numerous very small-scale, high-frequency dissipative eddies.

"This interaction created a great deal of turbulent motion and the movements of the fluid elements within it were very intense, abrupt and chaotic. The entire structure had a very small scale. The intensity of this interaction destroyed the identity of the individual elements. The interaction not only occurred between the mean flow and the ejected element, but the continual ejection of fluid into this chaotic mass by the remainder of the original element and others which could occur at approximately the same time caused increased interaction and mixing."[2]

These studies suggest a simple mechanism for turbulent drag reduction. It is known that even very dilute (30 to 50 ppm) drag-reducing polymer solutions exhibit a rather strong resistance to breakup and droplet formation and prefer instead to form thread or filament-like structures.[6-8] Following Lumley,[8] we refer to this phenomenon as "filament formation." Filament formation is well illustrated in the recent photographs by Goldin, Yerushalmi, and Shinnar[7] of the breakup of jets of Newtonian, viscoinelastic, and viscoelastic (drag-reducing) fluids. In addition, this effect may easily be observed by simply dipping one's finger into the liquid. When the finger is withdrawn, "a very long, coherent, tenuous thread" is seen to trail from the finger to the liquid.[8]

A somewhat related phenomenon occurs when a jet of dyed drag-reducing solution is injected into a quiescent mass of the same solution, as in the studies of Gadd.[9] The dispersion of the dye is severely inhibited, the dye preferring instead to form into filamentous streaks.

Recalling now that Corino and Brodkey's studies[2] indicate turbulent energy dissipation to occur when the ejected element breaks up or bursts, causing intense and chaotic mixing on a very small scale with the external flow, it is suggested here that turbulent drag reduction in dilute polymer solutions is a result of the aversion of these systems to dispersion or breakup, as indicated in the works of Goldin et al.[7] and Gadd,[9] for example.

This explanation is, in fact, closely related to that advanced by Gadd in 1965.[9] There are important differences, however. Gadd suggested that drag reduction was due to a reduction in the frequency of ejections. In the mechanism proposed here, only the intensity of interaction and mixing of these ejections with the external flow is reduced, the frequency of ejections being unaffected.

The turbulent flow of a dilute drag-reducing polymer solution is thus supposed to conform quite closely to the description given by Corino and Brodkey,[2] with the exception that the "bursting" of the ejected elements is less pronounced, leading to less viscous dissipation. In support of the

present arguments we may note that the frequency of ejections, as determined by Corino and Brodkey, is given by

$$\omega \sim U_*{}^2/\nu \tag{1}$$

or, alternatively, by

$$\omega \sim \frac{U}{D} N_{\mathrm{RE}}{}^{0.75}, \tag{2}$$

both expressions being equivalent in the range of Reynolds numbers studied. Equation (1) or (2) has been taken by numerous authors as representing the characteristic frequency of the dissipative eddies.[4,10,11] Our explanation is also consistent with the reduced heat transfer found in such systems.

The reduction in bursting would necessarily lead to a shift in the high wavenumber end of the turbulent energy spectrum toward smaller wavenumbers (larger eddies). One would also intuitively expect a decrease in the intensity of turbulence, at least in the region where the bursting occurs.[2] These considerations are consistent with the conclusions of Latto and Shen,[12] who studied the structure of the turbulent flow field of a dilute drag-reducing solution in the boundary layer on a flat plate. It is believed, however, that further studies are required before definite conclusions may be reached. We add finally that the present considerations are taken to apply only to very dilute solutions, for which the solution viscosity is essentially indistinguishable from that of the pure solvent.

THE PHENOMENOLOGICAL EXPLANATION OF TURBULENT DRAG REDUCTION

A continuum model of viscoelastic fluid behavior, the convected Maxwell model, has been used extensively by Seyer and Metzner[10,11] as a basis for describing viscous drag reduction. Most of the systems studied by these workers have been of the more concentrated type, exhibiting, for example, a shear-dependent viscosity. On the other hand, the very dilute solutions under consideration here are essentially indistinguishable from the base solvent and exhibit a constant viscosity.[13] A quite simple molecular model, the dumbbell model, exhibits this constancy of viscosity but allows for "elastic" effects such as normal stresses and a dynamic viscosity.[14,15] Some results of interest for both the convected Maxwell model and the dumbbell model are listed in Table I.

Here we have chosen to express the convected Maxwell model as follows:

$$\mathbf{S} = -p\,\mathbf{1} + 2\,\eta_0\,\mathbf{D} + \mathbf{S}'$$

$$\mathbf{S}' = 2\mu\mathbf{D} - \theta_M\,\frac{\delta\mathbf{S}'}{\delta t} \tag{3}$$

$$\frac{\delta\mathbf{S}'}{\delta t} = \frac{D\mathbf{S}'}{Dt} - \boldsymbol{\nabla}\mathbf{v}\cdot\mathbf{S}' - \mathbf{S}'\cdot(\boldsymbol{\nabla}\mathbf{v})^T$$

TABLE I
Comparison of the Convected Maxwell Model and the Dumbbell Model

Rheological measurement	Convected Maxwell model	Dumbbell model
1. Simple shearing flow $v_i \equiv (Gx_2,0,0)$		
(a) Viscosity $\eta = S_{12}/G$	Function of stress	Constant
(b) $N_{\mathrm{WS}} = \dfrac{S_{11} - S_{22}}{(S_{12} - \eta_0 G)}$	$2G\theta_M$	$2G\theta_D$
2. Pure deformational flow $v_i \equiv (-\bar{G}x_1,\bar{G}x_2,0)$		
$\bar{\eta} - \bar{\eta}_0 = \dfrac{S_{22} - S_{11}}{\bar{G}} - \bar{\eta}_0$	$\dfrac{4(\eta - \eta_0)}{1 - (2\theta_M\bar{G})^2}$	$\dfrac{4(\eta - \eta_0)}{1 - (2\theta_D\bar{G})^2}$
3. Small amplitude, oscillatory shearing $v_i \equiv (\gamma_0(x_2)e^{i\omega t},0,0)$		
(a) Dynamic viscosity $\eta^* - \eta_0 = (\eta' - \eta_0) - i\eta''$	$\dfrac{(\eta - \eta_0)(1 - i\omega\theta_M)}{1 + \omega^2\theta_M{}^2}$	$\dfrac{(\eta - \eta_0)(1 - i\omega\theta_D)}{1 + \omega^2\theta_D{}^2}$
(b) $\eta''/(\eta' - \eta_0)$	$\omega\theta_M$	$\omega\theta_D$

where μ now represents the viscosity *increment* above that of the solvent viscosity, η_0; θ_M is the relaxation time and, like μ, is taken to depend on the invariants of the stress tensor.[16]

The relaxation time of the dumbbell model, θ_D, is found to be[15,17]

$$\theta_D = \frac{M[\eta]_0\eta_0}{RT}, \tag{4}$$

where $[\eta]_0$ is the intrinsic viscosity for a "theta" solvent.[18] In the case of a nontheta solvent, we assume eq. (4) may still be utilized, with $[\eta]_0$ replaced by

$$[\eta] = \alpha^3[\eta]_0, \tag{5}$$

where α is the molecular expansion factor.[18,19]

With reference to these results, let us now consider some phenomenological explanations of turbulent drag reduction that have been advanced:

Increased Fluid Elasticity

Metzner and Park,[20] in an early study of drag reduction, suggested that the ratio of elastic and viscous stresses be taken as measure of the drag-reducing effectiveness of a polymer solution, i.e., the quantity of importance was the Weissenberg number[10,11,20]:

$$N_{\mathrm{WS}} = \frac{S_{11} - S_{22}}{(S_{12} - \eta_0 G)} \tag{6}*$$

* In this work, we refer our discussions to the increased stress above that of the pure solvent, i.e., $\mathbf{S} - 2\eta_0\mathbf{D}$. In concentrated solutions, such as those considered by Metzner and Park,[20] the factor $2\eta_0\mathbf{D}$ is negligible.

Referring to Table I, we note that for both models, N_{WS} is determined by the relaxation time θ.

Resistance to "Stretching" Type Motions

A recent phenomenological explanation of turbulent drag reduction, advanced by Seyer and Metzner,[10,11] concerns the unusually high resistance offered by polymer solutions to pure stretching motions, as evidenced by the prediction of an infinite value for $\bar{\eta}$ at a critical shear rate $\bar{G}$, for the two models in Table I.   (It is known that in the case of the dumbbell model, the discontinuity in $\bar{\eta}$ is a result of the use of Gaussian statistics to describe the tension in the macromolecule.[15])   There are definite indications, however, that large values of $\bar{\eta}$ do arise.[8,15]   According to Seyer and Metzner, the turbulent eddy structure near the pipe wall may be considered roughly as such a stretching motion, with the three axis of the flow parallel to the pipe axis.   This idea is not incompatible with the explanation of drag reduction presented here if, as suggested by Lumley,[8] the "filament formation" tendency of drag-reducing solutions is associated with their high resistance to stretching.   Seyer and Metzner's considerations point again to the importance of the fluid relaxation time θ.

Enhancement of "Energy Storage" in Dynamic Motions

Vleggaar, Dammers, and Tels[21] have suggested that viscous drag reduction is related to the increased ability of the fluid to store energy in an oscillatory shear flow.   These authors take as a measure of drag reduction effectiveness the ratio of the storage and loss moduli, i.e.,

$$\frac{G'}{G'' - \omega\eta_0} = \frac{\eta''}{\eta' - \eta_0} = \omega\theta, \tag{7}$$

where ω is a characteristic frequency of the dissipative eddies.   Again we see the importance of the fluid relaxation time θ.

ON THE CORRELATION OF TURBULENT DRAG REDUCTION

From a phenomenological viewpoint, either of the three explanations just discussed might be used to argue the necessary dependence of the phenomenon on θ.   We prefer the arguments of Vleggaar, Dammers, and Tels[21] and take the following ratio as a measure of the drag-reducing effectiveness of dilute polymer solutions:

$$\frac{\eta''}{\eta' - \eta_0} = \omega\theta_D = N_{DE} \tag{8}$$

Here, N_{DE}, the Deborah number,[10,11,21] is the ratio of a characteristic fluid time and a characteristic flow time.   Based on our previous arguments, we are tacitly assuming that $\eta''/(\eta' - \eta_0)$ is a measure of resistance to bursting.   As the characteristic frequency ω, we take the frequency of ejections or bursts.   For the same bulk flow conditions (Q,D,ν), it is

assumed that this frequency is identical in the solution and the pure solvent, and hence we have

$$\omega \sim \frac{U}{D} N_{\mathrm{RE}}^{0.75}. \tag{9}$$

This is no longer equivalent to U_*^2/ν, however, as the friction velocity U_* is considerably diminished in the polymer solution. The nonequivalence of eqs. (1) and (2) under drag-reducing conditions has not been sufficiently emphasized.

From eqs. (4), (5), (8), and (9),

$$N_{\mathrm{DE}} \sim \frac{U}{D} N_{\mathrm{RE}}^{0.75} \frac{\alpha^3 [\eta]_0 \eta_0 M}{RT} \tag{10}$$

or, alternatively,

$$N_{\mathrm{DE}} \sim \frac{N_{\mathrm{RE}}^{1.75}}{D^2} \frac{\alpha^3 [\eta]_0 \eta_0 M \nu}{RT}. \tag{11}$$

Equation (11) explains the observed increases in drag-reducing effectiveness with decreasing tube diameter, increasing solvent power (greater α), and increasing molecular weight for a homologous series. It is important to note that eq. (11) was derived for a flexible, linear macromolecule, for which the dumbbell model applies, and furthermore, it is limited in its present form to monodisperse solutions.

Let us now consider the flow of a drag-reducing solution in a long, cylindrical tube. The drag ratio, D_R, is defined by Savins[13,22] as the ratio of the pressure drop for the solution to the pressure drop for the solvent at the same flow rate, in the same tube, i.e.,

$$D_R = \frac{(\Delta P)_{\mathrm{soln}, Q, D}}{(\Delta P)_{\mathrm{solvent}, Q, D}}. \tag{12}$$

If we make the assumption that the fractional reduction in pressure drop for any solution-tube-flow rate depends only on the Deborah number (again restricting the discussion to very dilute solutions), then

$$\frac{(\Delta P)_{\mathrm{solvent}, Q, D} - (\Delta P)_{\mathrm{soln}., Q, D}}{(\Delta P)_{\mathrm{solvent}, Q, D}} = \gamma(N_{\mathrm{DE}}).$$

Rearranging, we obtain

$$D_R = \beta(N_{\mathrm{DE}}), \tag{13}$$

where $\beta = 1 - \gamma$. Using the definition of the friction factor f, eq. (13) may be written as

$$\frac{f}{f_0} = \beta (N_{\mathrm{DE}}), \tag{14}$$

where f and f_0 are the friction factors for the solution and solvent, respectively, for the same flow rate and pipe diameter. Equations (11) and (14) lead to the prediction that, for any particular solution, f/f_0 is a unique function of $N_{RE}^{1.75}/D^2$.

In a recent study, Rodriguez, Zakin, and Patterson[3] have proposed a correlation between f/f_0 and the following quantity:

$$N_{DE}' = \frac{U\tau_1}{D^{0.2}} = \left[\frac{N_{RE}^{1.75}}{D^{2.10}} \nu^{1.75} \tau_1^{1.75}\right]^{4/7}$$

$$\tau_1 = \frac{M_v(\eta_{sp}/C)\eta_0}{0.586 \, RT\lambda_1}$$

where N_{DE}' is a modified (dimensional) Deborah number, and τ_1 is the first-mode relaxation time of the Zimm theory.[3] Although the above correlation is not equivalent to that presented here (even at low concentrations), it does predict a unique functional dependence of f/f_0 on $N_{RE}^{1.75}/D^{2.10}$ for any particular solution, in close agreement with the present predictions. As is seen by an examination of the results of these authors,[3] such a unique dependence is indeed found, not only at the low concentrations required in the present work, but also for the more concentrated solutions studied (up to approximately 4000 ppm).

Astarita, Greco, and Nicodemo[4] have also suggested that f/f_0 may be a unique function of the Deborah number, with $\omega = 1/\theta_{flow}$ given by eq. (9) and the relaxation time of the fluid left unspecified. Their studies again indicate the unique dependence of f/f_0 on $N_{RE}^{1.75}/D^2$ over wide ranges of concentration.

In order to confirm quantitatively the other aspects of the correlation represented by eqs. (11) and (14), measurements are required on dilute solutions of rather low polydispersity. For the calculation of θ_D, one may, in place of eqs. (4) and (5), use the measured values of the extinction angle χ for the solution in a Couette viscometer, along with the following relation:[15,17]

$$\theta_D = \frac{\cotan 2\chi}{G} \qquad (15)*$$

Equation (15) applies specifically to the case of the dumbbell model. On the other hand, the relationship between χ and the Weissenberg number,

$$N_{WS} = 2 \cotan 2\chi, \qquad (16)$$

which follows directly from eq. (15) and Table I, may be shown to be valid for very general types of fluid models, over the entire range of solution con-

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* We should perhaps add the qualifying statement that measurements of χ for feebly birefringent systems would present significant experimental difficulties. In any event, eq. [15] would certainly allow accurate determination of θ to much lower concentrations, and over much wider ranges of shear rate, than conventional measurements of this quantity in the Weissenberg rheogoniometer (10,11,23–26).

centration.[17,23]   Equation (16) may prove useful in the correlation of drag reduction for more concentrated solutions.

CONCLUSIONS

A mechanistic picture of turbulent drag reduction in dilute polymer solutions has been proposed, based on the increased resistance of these materials to breakup and dispersion.   This tendency, related to the "filament formation effect" noted by Lumley,[8] may be associated with the high resistance to stretching motions, as discussed in the work of Seyer and Metzner.[10,11]   The present hypothesis, and also the assumption that the "ejection" frequency is unchanged in solution, could be tested by repetition of the Corino-Brodkey[2] experiments for drag-reducing fluids.

The importance of the fluid relaxation time in the phenomenological explanation and correlation of turbulent drag reduction follows from a number of qualitative arguments.   The present correlation, essentially equivalent to that suggested by Astarita, Greco, and Nicodemo,[4] explains many of the observed features of drag reduction.   Further tests on sufficiently dilute, monodisperse solutions are needed.

Notation

C	concentration
D	tube diameter
$\mathbf{D} = 1/2[(\nabla \mathbf{v} + (\nabla \mathbf{v})T)]$	rate of strain tensor
$\dfrac{D}{Dt}$	material derivative
D_R	drag ratio
f, f_0	friction factor
G	velocity gradient in simple shearing
$\bar{G}$	velocity gradient in pure deformation
$G^* = G' + iG''$	dynamic shear modulus
M	molecular weight
M_v	viscosity-average molecular weight
N_{DE}	Deborah number
N_{RE}	Reynolds number
N_{WS}	Weissenberg number
R	gas law constant
$\mathbf{S}, S_{ij}$	stress tensor
T	absolute temperature
U_*	friction velocity
$\mathbf{v}, \mathbf{v}_i$	velocity
$\nabla \mathbf{v}$	velocity gradient tensor
α	molecular expansion factor

η_0 — solvent viscosity

η — solution viscosity

$$\bar{\eta} = \frac{S_{22} - S_{11}}{\bar{G}}$$ — "viscosity" in pure deformational flow

$\bar{\eta}_0 = 4\eta_0$ — "viscosity" of solvent in pure deformational flow

$$[\eta] = \lim_{c \to 0} \frac{\eta - \eta_0}{C\eta_0}$$ — intrinsic viscosity

$$\eta_{sp} = \frac{\eta - \eta_0}{\eta_0}$$

θ — relaxation time

λ_1 — first eigenvalue in Zimm theory

χ — extinction angle of streaming birefringence

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