Criteria for the Onset of Drag Reduction

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The sudden onset of drag reduction as the wall shear rate increases is now well established (Hoyt, 1972). In a recent paper (Kohn, 1973) we proposed that drag reduction is a function of the energy stored per ml of solution by macromolecules in a fluctuating shear field (W, the strain energy density). In support of this theory we reported a correlation between the strain energy density and the drag ratio f/f_0. The strain energy density was estimated from Gaussian bead-spring theory (Beuche, 1954; Cerf, 1957, 1958; Rouse, 1953; Zimm, 1956) using an equation derived by Blatz (1967).

$$W = \frac{cRT}{2M} \sum_{i=1}^{N} \ln[1 + (\gamma\tau_i)^2]$$

The bead-spring theory gives τ_i as

$$\tau_i = \frac{b^2\zeta}{6kT\lambda_i}$$

The eigenvalues λ_i of the hydrodynamic mobility operator are given by Pyun and Fixman (1965) as

$$\lambda_i = 4 \sin^2 \left[\frac{i\pi}{2(N+1)} \right] + 4h^* N^{-3/2} [I_1(i) + I_2(i)]$$

I_1 and I_2 are tabulated functions of Fresnel integrals.

Blatz's equation has not been applied to drag-reducing systems earlier because the segmental parameters were unknown. We have rendered the equation useful by the following technique. The number of statistical segments per molecule was approximated by $N = M/pM_0$, and the segmental parameters b, ζ, h^*, and p were estimated (Kohn, 1973) by a least-square fit of computed to observed intrinsic viscosities for a wide range of molecular weights. The bead-spring theory gives the intrinsic viscosity as

$$[\eta] = \frac{N_0 b^2 \zeta}{6M\eta_0} \sum_{i=1}^{N} \frac{1}{\lambda_i}$$

The optimized parameters for several polymer-solvent systems are listed in Table 1.

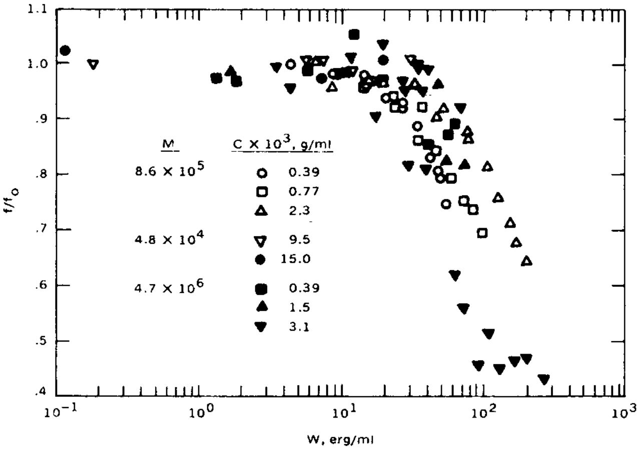

Fig. 1. Drag ratio vs. strain energy density for polyisobutylene-cyclohexane solutions. Data are from Hershey (1965) ($M = 4.8 \times 10^4$ and $M = 8.6 \times 10^5$) and from Rodriguez (1966) ($M = 4.7 \times 10^6$).

Examples of the drag ratio-strain energy density correlation for the pipe flow data of Hershey (1965), Rodriguez (1966), and Paterson and Abernathy (1970) are given in Figures 1 and 2. The strain energy density at the onset of drag reduction W_0 was determined from this correlation for several polymer-solvent systems (Kohn, 1973). The average W_0 value for several concentrations are listed in Table 2 along with their average deviations. Since the Rouse-Zimm bead-spring theory treats all polymers as ideally flexible, the difference in flexibility of real polymers is reflected in higher vaules of W_0 for stiffer polymer chains. The discrepancy may be removed by using a bead-spring model with internal viscosity (Cerf, 1958).

CRITICAL CONCENTRATION AND CRITICAL WALL SHEAR RATE

The ratio $W_0/W' = C^*$ is the concentration below which no drag reduction is observed at the shear rate corresponding to W'. Figures 3 and 4 show how the critical concentration varies with the wall shear rate for

TABLE 1. SEGMENTAL PARAMETERS DETERMINED BY LEAST-SQUARES FIT OF CALCULATED TO OBSERVED INTRINSIC VISCOSITIES

System	b, A	$\zeta \times 10^9$, g/s	h^*	p, monomers/ segment
PIB-benzene	27.0	5.70	0.218	19.0
PIB-cyclohexane	40.5	7.92	0.182	16.6
PMMA-toluene	36.2	7.77	0.152	16.3
PEO-water	20.9	9.77	0.029	17.2

PIB = polyisobutylene, PMMA = poly(methyl methacrylate), PEO = poly(ethylene oxide).

TABLE 2. ONSET STRAIN ENERGY DENSITIES FOR DILUTE POLYMER SOLUTIONS

System	Molecular weight	t, °C	τ_1, s[a]	W_0, erg/ml
PIB-benzene	5.9×10^5	24	5.13×10^{-6}	49 ± 2
PIB-cyclohexane	8.6×10^5	25	4.17×10^{-5}	16 ± 2
	4.7×10^6	25	5.62×10^{-4}	
PMMA-toluene	1.5×10^6	30	3.73×10^{-5}	32 ± 10
PEO-water	8.0×10^6	23	3.09×10^{-3}	0.016 ± 0.005

[a] Longest molecular relaxation time computed from the Gaussian chain theory.

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several molecular weights of polyisobutylene in cyclohexane and poly(ethylene oxide) in water. The onset wall shear rates for several concentrations of these polymers were determined from these curves and compared with the observed onset shear rates. This comparison is shown in Table 3. The agreement is fairly good; the discrepancies between the computed and observed shear rates are no greater than the scatter of points for the f/f_0 versus W curves. Since the data of Table 3 are different from the data used to determine W_0, the agreement between the computed and observed onset wall shear rates may be taken as further evidence of the validity of the energy storage theory.

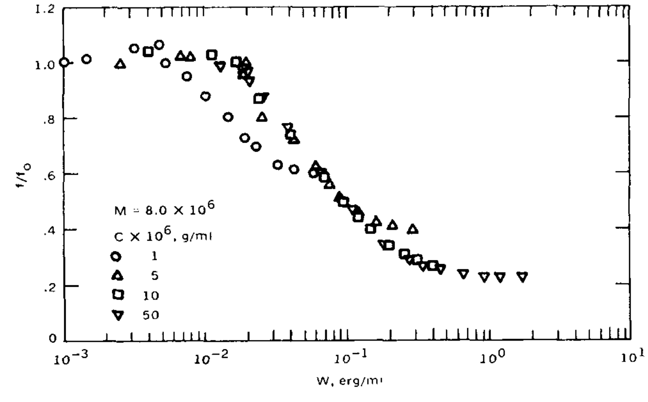

Fig. 2. Drag ratio vs. strain energy density for poly(ethylene oxide)-water solutions. Data are from Paterson and Abernathy (1970).

The tick marks on the curves of Figures 3 and 4 represent shear rates equal to the inverse of τ_1, the longest relaxation time of the polymer molecule. At shear rates less than this value the slope of the curve of log C^* versus log γ approaches -1.9. At shear rates greater than this value the slope of the curve approaches -0.7. The sharp increase of the critical concentration at shear rates less than $1/\tau_1$ may be responsible for the partial success of the Deborah number (Elata et al., 1966; Fabula et al., 1966; Hershey and Zakin, 1967) at predicting the onset of drag reduction. The high viscosity of more concentrated solutions counteracts the effects of energy

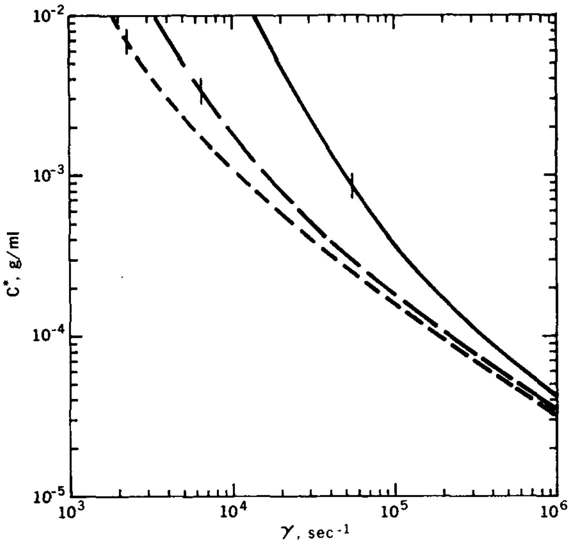

Fig. 3. Shear rate dependence of the critical concentration for polyisobutylene-cyclohexane. ————, $M = 5 \times 10^5$; — — ——, $M = 2 \times 10^6$; — — — —, $M = 4 \times 10^6$.

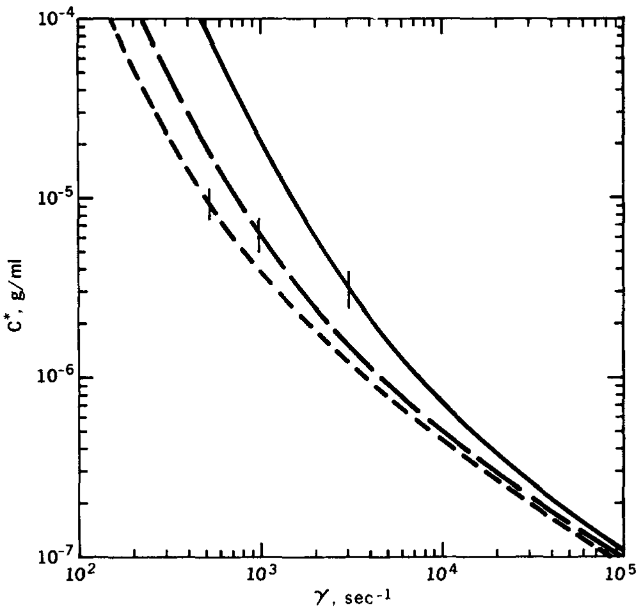

Fig. 4. Shear rate dependence of the critical concentration for poly(ethylene oxide)-water. ————, $M = 2 \times 10^6$; — — ——, $M = 4 \times 10^6$; — — — —, $M = 6 \times 10^6$.

TABLE 3. COMPUTED AND OBSERVED ONSET WALL SHEAR RATES

System	Molecular weight	Concentration, g/ml	Onset wall shear rate, sec^{-1} Calculated	Onset wall shear rate, sec^{-1} Observed	Reference
PIB-cyclohexane	2.2×10^6	5.0×10^{-4}	3.2×10^4	1.72×10^4	Rodriguez et al. (1967)
		2.0×10^{-3}	9.6×10^3	1.45×10^4	
PEO-water	4.0×10^{6a}	2.0×10^{-5}	4.9×10^2	4.1×10^2	McNally (1968)
		3.5×10^{-5}	3.6×10^2	1.7×10^2	Lowe (1969)
		5.0×10^{-5}	3.0×10^2	1.8×10^2	Goren and Norbury (1967)
	6.0×10^{6a}	3.5×10^{-6}	1.1×10^3	7.8×10^2	Tomita (1970)
				8.8×10^2	
		7.0×10^{-6}	6.3×10^2	2.7×10^2	
				4.9×10^2	
		1.5×10^{-5}	3.8×10^2	1.1×10^2	
				2.9×10^2	
	6.1×10^6	1.0×10^{-5}	4.9×10^2	3.9×10^2	Virk et al. (1967)

[a] Estimated molecular weight.

storage, and considerably less (or no) drag reduction is observed (Hoyt, 1972). For this reason, experiments are typically performed with dilute solutions.

The Deborah number theory predicts the onset of drag reduction when the wall shear rate equals the inverse of the longest relaxation time of the polymer molecule (that is, a Deborah number of 1). This work confirms our previous findings (Kohn, 1973) that the onset Deborah number approaches (and even falls below) unity as the concentration of polymer increases. At sufficiently small concentrations the onset wall shear rate exceeds $1/\tau_1$. Thus the time scale criterion does not appear to be a sufficient condition for the onset of drag reduction.

DRAG REDUCTION AND MOLECULAR BEHAVIOR

Polymer molecules subjected to a shear stress stretch along the direction of the maximum velocity gradient. This extension may be approximated by (Zimm, 1972)

$$\frac{<L>}{<L>_0} = \left[1 + \frac{16}{3\pi^2}\gamma^2 \sum_{i=1}^{N} \tau_i^2 \right]^{1/2}$$

Figures 5 and 6 show the relative extension of polyisobutylene-cyclohexane and poly(ethylene oxide)-water for several molecular weights as a function of the wall shear rate. A tick mark denotes the shear rate equal to $1/\tau_1$. This point corresponds to the onset of significant molecular elongation. The rapid increase of the critical concentration as the shear rate drops below $1/\tau_1$ must reflect the insignificant extension of the polymer coil at these shear rates. At these low relative extensions very little energy is stored in the polymer deformation modes, and high concentrations would be required to develop a strain energy density equal to W_0.

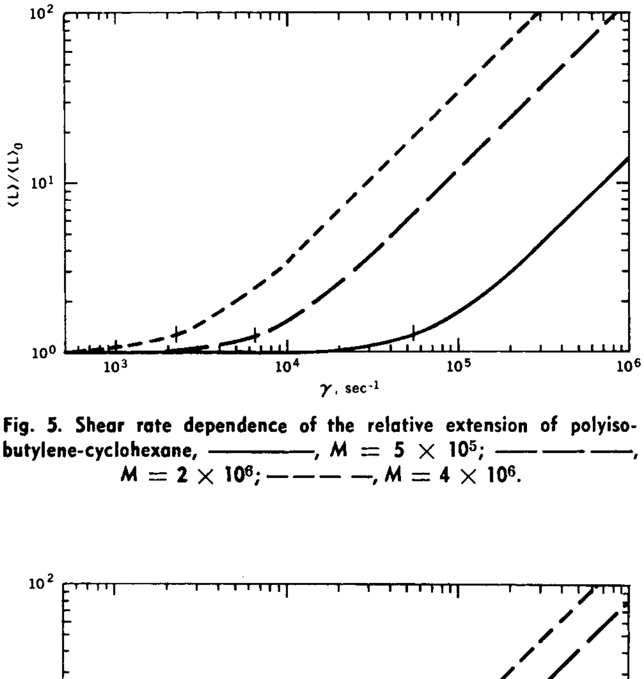

Fig. 5. Shear rate dependence of the relative extension of polyisobutylene-cyclohexane, ————, $M = 5 \times 10^5$; — — — —, $M = 2 \times 10^6$; - - - -, $M = 4 \times 10^6$.

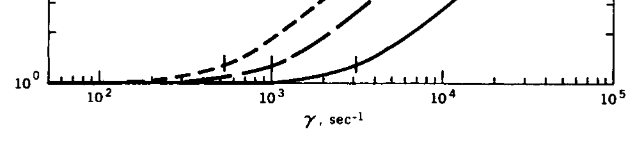

Fig. 6. Shear rate dependence of the relative extension of poly(ethylene oxide)-water. ————, $M = 2 \times 10^6$; — — — —, $M = 4 \times 10^6$; - - - -, $M = 6 \times 10^6$.

COMPARISON WITH THE WALSH THEORY

Several years ago, Walsh (1967) suggested that the onset of drag reduction occurs when H, the ratio of the rate of energy storage by macromolecules in solution to the rate of convection of turbulent energy away from the wall, is 0.01.

$$H = \frac{8cM[\eta]^2\tau_w}{RT} = 19.05\, c[\eta]\ (\eta/\eta_0)\tau_1\gamma = 0.01$$

In contrast, our energy storage theory asserts that energy is stored during periods of high shear stress and dissipated during periods of low shear stress when the polymer molecules relax. By this process, energy transferred to the near wall region by molecular collisions or by the interaction of potential energy fields is dissipated. In the absence of polymer molecules, turbulent convection would maintain the energy balance in the viscous sublayer. Thus the viscoelastic response of the polymer molecules to the fluctuating wall shear rate provides a dissipative mechanism, unavailable to the pure solvent, which obviates the need for as much turbulent mixing and results in a lowered production of turbulent energy. We believe that this mechanism is more efficient (that is, less dissipative) than turbulence itself; it helps to maintain the energy balance in the sublayer with less loss in volumetric flow rate.

While the Walsh theory has been shown to be inadequate (Kohn, 1973; Virk and Merrill, 1969), it continues to hold a fascination for researchers in the field of drag reduction. It is therefore of interest to compare the predictions to the two theories. Walsh's theory predicts the critical concentration to be given by

$$C^* = \frac{0.00125RT}{M[\eta]^2\eta\gamma}$$

which results in values of C^* which are as much as two orders of magnitude too low. The behavior of C^* as a function of γ as predicted by the two theories are compared (with an H value of 1) in Figure 7. Unlike the present theory, the curves for the Walsh theory do not show the rapid increase of C^* as γ falls below $1/\tau_1$. Walsh's theory is based on the tacit assumption that the polymer molecules are strained to the same degree (and hence at the same rate) as the fluid. In contrast, the bead-spring theory predicts that the molecular strain lags behind the fluid strain since there is a minimum strain rate for significant molecular extension (Figures 5 and 6). Since the H parameter does not predict that a minimum fluid strain rate is necessary for significant molecular elongation, the resulting energy storage is proportional to γ^2 for all values of γ (Walsh, 1967). (The slight curvature at high C^* for the $M = 6 \times 10^6$ curve in Figure 7 is due to the increased solution viscosity.) This behavior is not in agreement with the observed data.

SUMMARY

The energy storage theory of drag reduction as formulated from the Gaussian bead-spring theory of polymer

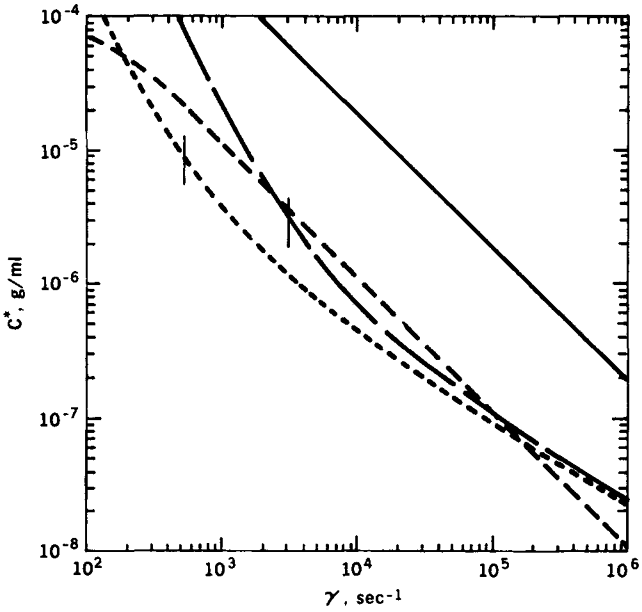

Fig. 7. Comparison of onset theories for poly(ethylene oxide)-water. For $M = 2 \times 10^6$: ————, Walsh theory; ——— ———, present theory. For $M = 6 \times 10^6$: — — — —, Walsh theory; — — — —, present theory.

structure and mechanics has been successful in correlating the polymer concentration and wall shear rate at the onset of drag reduction. Both the time scale theory and Walsh's energy storage theory are inadequate in this respect. We believe that these results support our view of the role of energy storage in the mechanism of friction reduction.

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NOTATION

b = root mean square segment length
c = polymer concentration
C^* = critical concentration
f = observed Fanning friction factor
f_0 = Fanning friction factor from Karman-Prandtl equation using solution Reynolds number
H = Walsh's onset parameter
h^* = segmental hydrodynamic interaction parameter
k = Boltzmann's constant
$<L>$ = root mean square end-to-end distance of the polymer coil in a sheared solution
$<L>_0$ = root mean square end-to-end distance of the unperturbed polymer coil
M = polymer molecular weight
M_0 = monomer molecular weight
N = number of statistical segments per polymer molecule
N_0 = Avogadro's number
p = number of monomer units per statistical segment
R = gas constant = kN_0
T = absolute temperature
W = strain energy density
W_0 = onset strain energy density
W' = strain energy per polymer molecule

Greek Letters

γ = average wall shear rate
ζ = segmental friction coefficient
η = solution viscosity
η_0 = solvent viscosity
$[\eta]$ = polymer intrinsic viscosity
λ_i = ith eigenvalue of the hydrodynamic mobility operator
τ_i = relaxation time of the ith deformation mode of the polymer molecule
τ_w = wall shear stress

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