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Drying Polymers During Screw Extrusion

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Partial removal of dissolved or absorbed solvent from polymers can be accomplished during the process of screw extrusion. This paper shows how two simple transport models based either on an effective diffusivity or on an empirical mass transfer coefficient can be combined with the fluid mechanical equations which describe polymer flow during screw extrusion. In this way, the drying of solvent from a polymer during extrusion can be correlated in terms of the design and operating parameters of the extruder screw and a mass-transfer coefficient or an effective diffusivity. This approach is illustrated employing data obtained by using two extruders of different size and two different solvent-polymer systems. Furthermore, it is pointed out how the results can be used to predict the extent of drying during screw extrusion as well as how modifications in extruder-screw design can permit more extensive drying of polymers during the extrusion process.

During polymer manufacture it is often necessary to remove dissolved, volatile hydrocarbon impurities from the product. Frequently, this is carried out by drying with steam or hot gas. As an example, conventional processes for polypropylene manufacture may involve several stages of drying to remove dissolved solvents such as hexane, xylene, etc., from the product.

The work described here demonstrates that it is also possible to remove some dissolved volatile material from a polymer as it undergoes screw extrusion, a final step in preparing a plastic for packaging. Thus this extrusion, aside from its conventional role, can also serve to replace part of the usual drying operation thereby decreasing the requirement for ordinary drying equipment. This study develops a design method for predicting how much drying a given extruder can provide for a given solvent-polymer system. Alternatively, the results indicate how an extruder can be modified to provide additional drying capacity.

The approach begins by focusing attention on the fluid mechanics of polymer flow within a screw extruder. This provides knowledge of the geometrical configuration and residence time of the drying polymer in the extruder. With these results it is possible to formulate drying either in terms of an empirical mass transfer coefficient or in terms of an effective diffusivity and thereby arrive at equations which predict the extent of drying. These equations involve mechanical design parameters of the extruder as well as an effective diffusivity of solvent in polymer in the case of a diffusional model, or a mass-transfer coefficient in the case of the more empirical model.

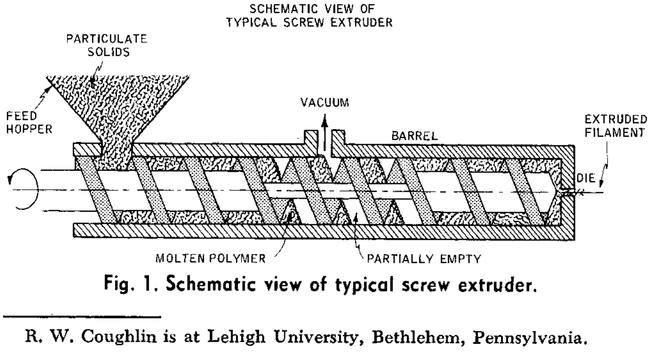

Fig. 1. Schematic view of typical screw extruder.

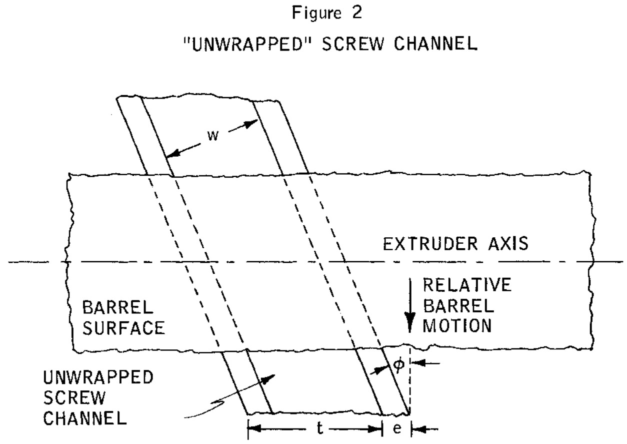

Fig. 2. "Unwrapped" screw channel.

FLOW IN SCREW EXTRUDER

A conventional description of a screw extruder is that of a screw turning within a concentric cylinder. The polymer as a particulate solid is fed into a hopper at the low-pressure end; at the other end it passes through a die in the plastic state under high pressure. Heat generated by the shearing action within the extruder melts the polymer (heat can also be supplied by external sources). The relative motion of screw and barrel provides conveying action toward the die. Figure 1 depicts a typical screw extruder.

The fluid dynamical theory of isothermal screw extrusion has been presented in detail in many places (*1* to *9*). For the purpose at hand, only a cursory description and the results of the theory will be given.

The conveying action of an extruder is easily understood by considering the helical screw channel as unwrapped and laid flat as illustrated in Figure 2. With this convention, the barrel becomes a flat plate which moves over the screw channel as noted. The component of barrel motion parallel to the screw channel imparts a drag force to the molten polymer which wets the barrel. Thus, the fluid is conveyed towards the die against a pressure which is a maximum at the die. The net flow is conveniently and

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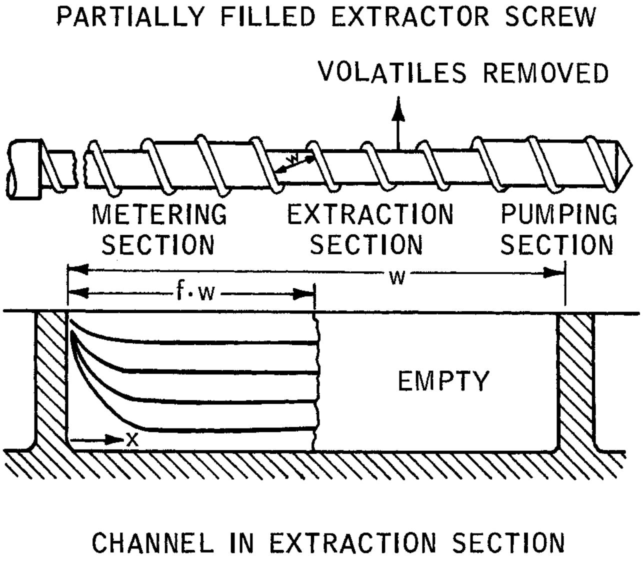

Fig. 3. Partially filled extractor screw.

accurately described as the difference between two components: the drag flow forward against zero pressure drop, and a fictitious backward flow due to pressure in the absence of drag flow. The net flow, Q, through the extruder has been given by Squires (*9*) as the difference between these two quantities:

$$Q = F_d \alpha N - F_p \frac{\beta}{\mu} \frac{dp}{d\lambda} \tag{1}$$

Here N is the speed of revolution of the extruder screw, p the pressure, λ the distance measured along the screw channel and μ the viscosity of the flowing polymer. F_d, F_p, α, and β are factors that depend only on extruder geometry or design parameters. Squires (*9, 16*) presents the analytical form of all these factors and gives values of F_d and F_p in graphical form as well. In Equation (1) the first term is the drag flow which is independent of pressure and occurs only by virtue of the forward motion imposed on the polymer by the barrel wall. The second term is the more usual kind of flow due to the pressure gradient along the screw channel, the pressure flow.

FLOW DIFFERS IN EXTRACTION SECTION

In the extraction section, where drying occurs, the screw channel is deeper than elsewhere, and, consequently, this part of the screw operates only partially filled. This condition is shown in Figure 3. At the extraction section, the extruder barrel is provided with a vent where a vacuum is applied for removing volatile substances from the polymer. There can be no axial pressure gradient in a partially filled screw section, and, therefore, there can be no pressure flow in this section. Hence, for the extraction section the pressure gradient, $dp/d\lambda$, is zero, the pressure flow is zero, the net flow is entirely drag flow and Equation (1) becomes:

$$Q = F'_d \alpha N \tag{2}$$

The quantity α which appears in Equation (1) is given by:

$$\alpha = \frac{\pi^2 d^2 h (1 - e/t) \sin \phi \cos \phi}{2} \tag{3}$$

The quantity F'_d is a shape factor for partially-filled screws. Figure 4 gives the values of F'_d as a function of the channel depth to width ratio (h/w) with f, the fraction filled, as a parameter. This graph is due to Squires (*9, 16*) who also presents the analytical form of F'_d. It should be noted that such forms of this shape factor are available which either neglect or take account of the curvature of the screw channel (*8, 9*). Such curvature has been neglected in the present work.

DRYING IN EXTRACTION SECTION

Within the partly filled screw channel of the extraction section the helical ribbon of polymer is exposed to the vacuum on one surface only. The depth of polymer below this surface is fw, where f is the fraction filled and w is the channel width. The polymer ribbon can be thought of as flattened out, as it would be in an unwrapped screw channel. Drying of this polymer ribbon can be treated as an effective diffusion problem or by means of an empirical mass-transfer coefficient model.

According to the first approach, the polymer ribbon presents a two-dimensional, steady state problem in a moving medium. The situation is simplified by viewing it as a one-dimensional, unsteady state problem in a stationary medium. With these considerations, transport takes place in one direction only, that is, perpendicular to the surface exposed to the vacuum, and the exposure time becomes the residence time of the polymer in the extraction section. Although these assumptions are by no means rigorous, they allow the use of one of the simpler solutions (*10, 11*), to the diffusion equation:

$$\frac{C(\theta) - C^*}{C_0 - C^*} = \sum_{n=0}^{\infty} \frac{8}{\pi^2} \cdot \frac{1}{(2n+1)^2} \cdot \exp\left\{ -\frac{(2n+1)^2 \pi^2 D\theta}{4f^2w^2} \right\} \tag{4}$$

It must be recognized that D in Equation (4) is an effective diffusivity or dispersion coefficient and includes effects due to convective mixing within the molten polymer being dried. This well known expression describes isothermal, one-dimensional diffusion of a single component in a polymer slab of thickness fw. Since C^* is the equilibrium solubility of the volatile material in the polymer, this quantity is effectively zero when a good vacuum is applied to the extruder. Thus, with C^* equal to zero, Equation (4) gives $C(\theta)/C_0$, the fraction of volatile material remaining in the polymer as a function of the dimensionless group, $\pi^2 D\theta / 4f^2w^2$.

An alternative model can be constructed by assuming that the rate of drying is described by a mass-transfer coefficient, k, and, accordingly, is also proportional to the exposed surface to volume ratio, s/v, of the polymer. This can be written:

$$\frac{dC}{d\theta} = -k \frac{s}{v} (C - C^*) \tag{5}$$

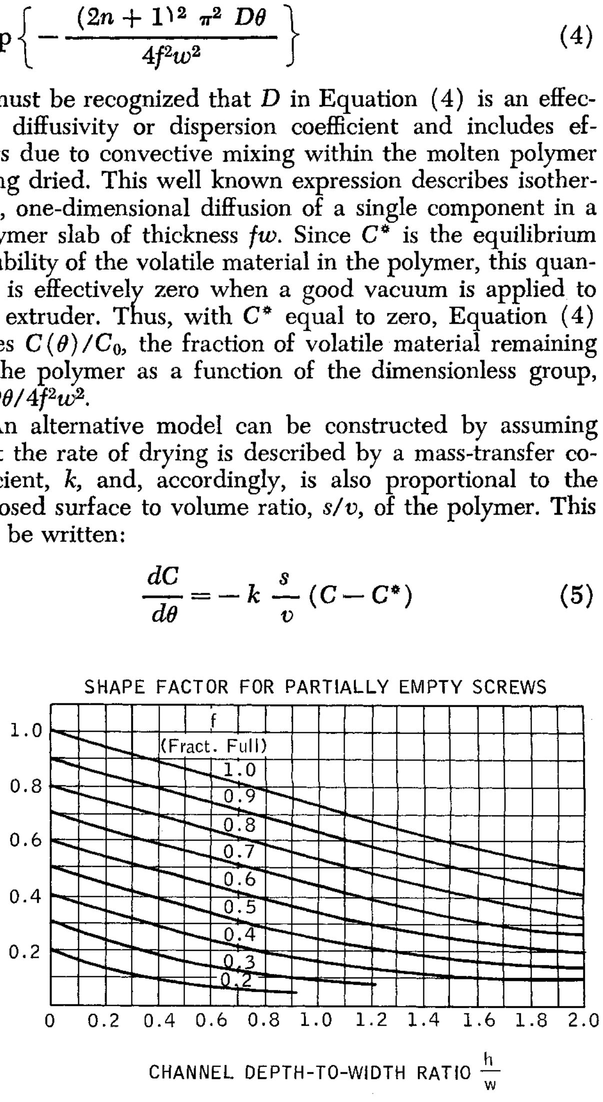

Fig. 4. Shape factor for partially empty screws.

Using $C = C_0$ at $\theta = 0$ and setting $C^* = 0$ and $v/s = fw$, Equation (5) integrates to:

$$\frac{C(\theta)}{C_0} = \exp\left(-\frac{k\theta}{fw}\right) \tag{6}$$

Equation (6) is a more empirical alternative to Equation (4). Simple calculation using measured solubility data for xylene in polypropylene indicates that C^* was so small as to be negligible compared to C_0 or $C(\theta)$ in the experiments described below.

EXPERIMENTAL RESULTS

The experiments were conducted using extruders of commercial size. The temperature of the polymer melt in the extruders was measured by thermocouples and controlled to within about ±5°F. by heating electrically. Xylene and methanol were selected for these tests as being representative of two dissimilar generic types of solvents. These tests may be summarized as follows:

Test Series	Extruder Diameter	Solvent/Polymer System
I	3½ in.	xylene/polypropylene
II	4½ in.	xylene/polypropylene
III	4½ in. modified*	xylene/polypropylene
IV	4½ in.	methanol/polypropylene

* The 4½ in. extruder originally had a ½ in. screw-channel depth; it was modified by milling the screw channel to a depth of ¾ in. The significance of this modification is discussed below.

Table 1 presents complete data for the four series of tests. Included in this Table are values of D and k computed for each experimental run using Equations (4) and (6) respectively.

Aside from effective diffusivity and mass-transfer coefficient, the other parameters in the dimensionless groups of Equations (4) and (6) were obtained as follows. The value of f, the fraction full, was obtained by using Equation (2) and Figure 4 and the value of the residence time θ, was obtained by dividing the filled volume of the extraction section by the extruder throughput. The channel width, w, is a mechanical parameter of the extruder screw. Screw curvature was not accounted for in these calculations. Had curvature effects been included this would have resulted in mean values of k and D somewhat different from those reported below. However dispersion of data about the sample mean values of k and D would have been essentially unaltered from that reported below.

In most instances, the polymer powder, as obtained from a commercial or pilot plant, contained sufficient solvent. When this was not the case, solvent was added a few days before an experiment to the polypropylene powder contained in 55 gal. drums. On the day of an experiment, this powder was thoroughly homogenized by tumbling for a few hours in a rotary mixer. Experimental samples for chemical analysis were obtained (in triplicate) from the powder in the feed hopper of the extruder and from the pellets obtained by chopping cold extrudate. Concentration of feed powder was about 1% solvent by weight. Care was taken to obtain samples only after an extruder had operated in the steady state for about 10 min. Moreover, evaporation of solvent was minimized by quenching the extrudate monofilaments in cold water and transporting all samples in filled airtight containers.

Analysis of samples was carried out by melting the polymer under vacuum, collecting the volatile materials evolved by condensing them in a cold trap and subsequently measuring the quantity of solvent by gas chromatography.

TABLE 1.

Test Series	I			II		III			IV			
Solvent/Polymer System	xylene/polypropylene			xylene/polypropylene		xylene/polypropylene			methanol/polypropylene			
Type Extruder	conventional			conventional		modified*			conventional			
extruder parameters, d	3.5 in.			4.5 in.		4.5 in.			4.5 in.			
ϕ	17.67 deg.			17.67 deg.		17.67 deg.			17.67 deg.			
e	⅜ in.			½ in.		½ in.			½ in.			
t	3.5 in.			4.5 in.		4.5 in.			4.5 in.			
w	2.99 in.			3.82 in.		3.82 in.			3.82 in.			
h	0.437 in.			0.50 in.		0.75 in.			0.50 in.			
temperature, °F.	500			500		500			500			
speed, N, rev./min.	40	80	120	120	75	30	60	90	45	90	60	47.5
throughput, lb./hr.	150	290	405	425	345	198	330	458	90	90	90	120
approximate % design throughput	37	72	100	77	59	36	70	83	16	16	16	22
throughput, cu. in./min.	96.1	186	259	274	222	125	212	292	61.5	61.5	61.5	82
$F_d = Q/\alpha N$	0.381	0.359	0.343	0.177	0.229	0.216	0.183	0.168	0.106	0.053	0.080	0.102
fraction full, f (from Figure 4)	0.41	0.40	0.39	0.20	0.27	0.27	0.24	0.21	0.156	0.103	0.129	0.152
filled volume, extraction section, cu. in.	30.9	30.1	29.4	28.4	38.3	57.5	51.0	44.6	22.2	14.6	18.4	21.7
residence time, extraction section, min.	0.321	0.162	0.1135	0.1036	0.1725	0.460	0.240	0.153	0.360	0.238	0.300	0.266
percent solvent removed†	68	47	49	42	61	87	75.6	59	99.84	99.95	99.87	99.72
$\pi^2 D\Theta/4f^2w^2$ [Equation (4)]	0.93	0.43	0.47	0.34	0.73	1.85	1.20	0.69	6.30	7.40	6.65	5.75
D, sq. in./min.	1.76	1.54	2.28	0.776	1.825	1.74	1.70	1.18	2.52	2.03	2.22	2.90
$\pi^2 D\Theta/4f^2w^2$ (based on mean effective D)	0.866	0.468	0.338	0.72	0.665	1.75	1.15	0.975	6.05	8.80	7.25	4.78
k, in./min.	4.35	4.68	5.60	4.03	5.64	4.57	5.38	4.20	10.65	12.55	10.90	12.80

* For test series III a conventional 4½ in. extruder was modified by milling the screw channel to a depth of ¾ in; the original depth was ½ in.
† Initial concentrations of Solvent were between about 0.3 and 1.0% by weight.

DISCUSSION

Examination of the values of effective diffusivity, D, and mass-transfer coefficient, k, in Table 1 reveals that the drying process in the extruder appears to be represented better by an empirical, mass-transfer coefficient model like Equation (5), than by an effective-diffusion model like Equation (4). This is evident principally from the extraordinarily large values of D computed from the latter model. However this model is of value in that the large values of D suggest processes other than molecular diffusion and laminar circulation within the extruder. The mean values of k and D for the two different systems can be summarized as follows:

xylene*-polypropylene
$D = 1.60 \pm 0.43$ sq.in./min.
$k = 4.81 \pm 0.63$ in./min.

methanol-polypropylene
$D = 2.24 \pm 0.36$ sq.in./min.
$k = 11.73 \pm 1.18$ in./min.

The mechanism of drying in the extruder is most probably not limited by molecular diffusion because the values of effective diffusivity computed from the data are about four orders of magnitude larger than might be expected. One indication of the influence of circulation currents on effective diffusivity is given by the work of Calderbank (*13*) who showed that the rigorous result (*14*) for diffusion in drops with internal circulation currents is numerically equivalent to the application of a factor of 2.25 to the molecular diffusivity in the transport equations written for a quiescent drop. Thus the extremely large values of effective diffusivities reported here indicate processes other than the simple laminar circulation currents known to take place within a polymer melt undergoing screw extrusion. In this regard, the formation of bubbles has been suggested (*12, 15*) but there was no evidence of boiling or frothing during operation of the extruders and solvent concentration was very low ($\sim 1\%$). It is also possible that the drying process was limited by heat transfer of the enthalpy of vaporization through the melt to the surface of the polymer. Another possible cause for the large observed values of k and D might be the presence of additional exposed surface of polymer not accounted for by the treatment above. For example, a thin layer of polymer might be produced by slippage between screw lands and barrel wall in the extraction section. Observation through the vacuum port did not reveal that this was the case, however. This might also account for the larger values of k and D for methanol which has a lower boiling point and larger heat of vaporization than xylene.

Test series I and II show that either of the two models applies equally well to extruders of different size. Test series III establishes the usefulness of these equations for predicting the improvement in drying resulting from mechanical modification of the extruder screw. Test series IV demonstrates that the models are also applicable to a different solvent-polymer system.

Equations (4) and (6) indicate that the extent of drying is a monotonically increasing function of $D\theta/f^2w^2$ or of $k\theta/fw$. When the channel depth, h is increased, extruder speed and throughput remaining constant, α is also increased proportionately and F_d decreased proportionately as indicated by Equations (2) and (3). This leads to a significantly smaller value of f (see Figure 4) which produces a more favorable value of the dimensionless quantities $D\theta/f^2w^2$ and $k\theta/fw$. Physically, deepening the extraction-section screw channel results in a thinner ribbon of polymer in that section and this means that transport takes place over a shorter distance. The results of test series III for a deepened extruder screw show this because they are in concord with the other data only when the value of f for this series is computed taking into account the deeper screw channel. That the modified extruder permits more solvent to be removed, other parameters remaining the same, is evident solely from the data for series II and III in Table 1.

It is also clear from Equations (4) and (6) that an alternative approach to increasing the drying capacity of a screw extruder is to increase the extraction-section residence time θ, by decreasing the throughput rate or by increasing the length of the extraction section. However, this may not be so effective as deepening the screw channel; doubling channel depth h doubles $k\theta/fw$ but more than doubles the quantity $D\theta/f^2w^2$ and this is clearly more effective than doubling θ by doubling the length of the extraction section.

CONCLUSIONS

This investigation has shown that screw extruders can be designed to produce a specified degree of drying. The results indicate that, if the value of k or D for solvent in polymer at the extrusion temperature is measured in a test extruder, it is a simple matter to design a different or larger extruder to produce a specified amount of drying, or to estimate the drying capacity of a particular extruder.

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NOTATION

$C(\theta)$ = concentration, moles/cu.in.
C_0 = initial concentration, moles/cu.in.
C^* = equilibrium concentration, moles/cu.in.
D = effective diffusivity or dispersion coefficient, sq.in./min.
d = screw diameter, in.
e = axial flight width, in.
F_d = shape factor for drag flow
F'_d = shape factor for drag flow in partially filled screw channel
F_p = shape factor for pressure flow
f = fraction of channel filled
h = screw channel depth, in.
k = empirical mass-transfer coefficient, in./min.
L = length of screw channel in extraction section
$L = n\sqrt{\pi^2d^2 + t^2}$
N = screw rotation speed, rev./min.
n = number of flights in extraction section
p = pressure, lb./sq.in.
Q = volumetric flow rate, cu.in./min.
s = surface of polymer exposed to vacuum, sq.in.
t = flight lead, in.
$t = \pi d \tan\phi$
v = volume of polymer in extraction section, cu.in.
w = channel width perpendicular to flight faces, in.
$w = (t - e)\cos\phi$
x = distance measured perpendicular to polymer surface in extraction section, in.

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* The xylene was a mixture of the three geometric isomers; the relative concentrations of these isomers were not measured.

Greek Letters

α = drag-flow constant
β = pressure-flow constant
ϕ = helix angle
λ = distance measured along screw channel, in.
μ = viscosity, lb./in.-min.
θ = residence time, min.

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A Phenomenological Interpretation and Correlation of Drag Reduction

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Drag reduction data for five concentrations of a water-soluble polymer and three tube diameters are presented. The data are correlated by a single curve relating two dimensionless parameters; these have been obtained from a phenomenological analysis of the mechanism of drag reduction.

Drag reduction, which is observed in turbulent flow of polymer solutions, has recently attracted much attention, and both data and tentative interpretations are fairly abundant in the literature. Nonetheless, no correlation of predictive value and reasonably simple form has so far been proposed. This paper reports the results of a work done in the attempt to establish such a correlation, on a strictly phenomenological basis.

THEORY

It is well known that, if pressure drop data for drag-reducing solutions are plotted in dimensionless form, that is, as friction factor vs. Reynolds number, different curves are observed for different tube diameters. The larger the tube diameter, the smaller is the drag reduction effect. It is therefore necessary, in order to correlate the data, to introduce, in addition to the Reynolds number, a second dimensionless group. This group should contain the tube diameter, in order to be able to correlate the observed diameter effect; therefore, this dimensionless group cannot simply be a dimensionless rheological parameter of the solution considered.

Astarita (*1*) proposed an interpretation of the drag reduction phenomenon based on the comparison of the smallest frequency of the noninviscid (or energy-dissipating) eddies, ν, and some characteristic time scale of the fluid considered, T. Following arguments of Levich (*2*) and of Lumley (*3*), the relevant frequency ν was evaluated as:

$$\nu = \frac{U}{D} N_{Re}^{0.5} \tag{1}$$

The proper dimensionless group to be considered, following this line of thought, is given by the product νT. This group can be identified with the Deborah number of the flow, as discussed in some recent papers (*4* to *6*).

Seyer and Metzner (*6*) have re-elaborated Astarita's discussion, and, following arguments of Hinze (*7*), propose to calculate ν by the equation:

$$\nu = \frac{U}{D} N_{Re}^{0.75} \tag{2}$$

Equation (2) seems to be preferable as compared to Equation (1) on the basis of the following considerations. It has often been reported (*8*, *9*) that drag reduction is observed only at wall shear stresses τ_w exceeding some critical value, τ_{cr}. This can also be stated in reverse form, say, that no drag reduction is observed if:

$$\tau_w < \tau_{cr} \tag{3}$$

Therefore, as long as Equation (3) is satisfied, τ_w can be calculated through the usual friction factors correlations, say by means of Blasius equation:

$$\tau_w = \frac{f}{2} \rho U^2 = 0.0395 \; \rho U^2 \; N_{Re}^{-0.25} \tag{4}$$

Equation (3) can be rearranged as follows:

$$\tau_w = 0.0395 \; \mu_w \frac{U}{D} \; N_{Re}^{0.75} \tag{5}$$

where μ_w is the viscosity at the tube wall. Equation (5) is based on the simplifying hypothesis that, for non-Newtonian drag-reducing solutions, the Reynolds number is defined on the basis of the wall viscosity, and that Equation (4) is still valid with this definition when no drag reduction occurs. This is of course only an approximation, as shown by Dodge and Metzner's correlation (*10*) of friction factors for nondrag reducing non-Newtonian liquids, but presumably a quite satisfactory one for slightly non-Newtonian liquids, as most drag-reducing dilute polymer solutions are.

If Equation (5) is substituted into Equation (3), the condition for the onset of drag reduction is obtained as

$$\frac{U}{D} N_{Re}^{0.75} < 25.3 \frac{\tau_{cr}}{\mu_w} \tag{6}$$