ρ = density, g./cc.
σ = interfacial tension, dynes/cm.
χ = d_j/d_t = jet contraction ratio

Subscripts and Superscripts

A = ambient medium
L = liquid (jet)
$*$ = the maximum in the breakup curve

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Viscoelastic Jet Stability

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The breakup of a low speed horizontal jet is investigated. Weber's theory for the Newtonian jet is extended to a linear viscoelastic fluid. The theory predicts a dependence of breakup length on the elasticity number. Breakup lengths are measured for low concentration solutions of polyisobutylene in tetralin. Two molecular weights, several concentrations, and five capillary diameters were studied. A single correlation is obtained for all data which gives the breakup length as a function of the elasticity number, and the parameters of Weber's theory. At constant values of the Ohnesorge number and Weber number, the breakup length decreases with increasing elasticity number. The effect of the length of the capillary is studied. At large elasticity numbers short tubes give rise to slightly shorter breakup lengths than long tubes under identical flow conditions.

This paper, one of a series (*3, 4, 10*) on the stability of liquid jets, is especially concerned with jets formed by extrusion of viscoelastic liquids from long capillaries into still air. An investigation of the role played by elasticity in altering the growth rate of infinitesimal disturbances has been conducted, and the experimental results can be rationalized through a simple viscoelastic stability analysis.

The phenomenon studied is the breakup into droplets of a horizontal laminar cylindrical jet. The primary measurement is of the breakup length, L, the distance from the capillary exit to the point where the jet is no longer coherent. Earlier studies (*4*) using Newtonian fluids have shown that data may be correlated by Weber's theory (*13*), in the form

$$L/D = C_1 \, N_{We}^{1/2} \, (1 + 3Z) \qquad (1)$$

The Ohnesorge number, Z, is the ratio of $N_{We}^{1/2}$ to N_{Re}, and is a measure of the relative importance of viscosity

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and surface tension in controlling stability. C_1 should be a constant, though Grant reports a dependence of C_1 on Z. Most of that dependence, it turns out, is due to Grant's method of correlating the data.

THEORY

The stability analysis for a viscoelastic fluid requires the assumption of a so-called "constitutive equation," the relationship of stress to rate of strain in the fluid. Once the constitutive equation is assumed an analysis similar to that in reference *10* may be carried out. Since only a linear stability analysis can be conducted one needs to examine only linear viscoelastic models. One such model is the Maxwell fluid, for which the stress tensor $\boldsymbol{\tau}$ and the rate of deformation tensor $\boldsymbol{\Delta}$ are related by

$$\boldsymbol{\tau} + \lambda_1 \frac{\partial \boldsymbol{\tau}}{\partial t} = \eta_0 \boldsymbol{\Delta} \qquad (2)$$

The constant λ_1, is a relaxation time, and η_0 is the zero-shear viscosity.

The linearity of this model immediately precludes its ability to describe viscoelastic phenomena such as normal stress development and non-Newtonian viscosity in simple shearing flows. Hence the model can be valid only in the limit of very small deformation rates, and the resulting stability analysis will be correspondingly restricted. Despite this, the model leads to useful results which guide the correlation of data obtained in the region of nonlinear viscoelastic response.

The theoretical development follows the technique of a previous work (*10*), and is given in detail elsewhere (*5*). The result is

$$L/D = C_2 N_{We}^{1/2}(1 + 3Z)/[1 + 3N_{El}/(1 + 3Z)^2] \quad (3)$$

and is subject to the approximation $N_{El} << (1 + 3Z)^2$. C_2 should have the same value as the coefficient C_1 found with Newtonian fluids. A new group, the elasticity number, appears, and is the ratio of the Weissenberg number to the Reynolds number, or

$$N_{El} = N_{Ws}/N_{Re} = \lambda_1 \eta_0/\rho D^2 \qquad (4)$$

It will prove more convenient to rearrange Equation (3) to the form

$$\frac{L/D}{N_{We}^{1/2}(1 + 3Z)} = C_1 = \frac{C_2}{1 + 3N_{El}/(1 + 3Z)^2} \qquad (5)$$

where the notation C_1 has been retained for the quantity on the far left of Equation (5). For a viscoelastic fluid, C_1 is predicted to be a decreasing function of N_{El}, at fixed Z. Thus, the theory claims that if two fluids have the same N_{We} and Z, the more elastic fluid is the less stable.

EXPERIMENTAL PROCEDURE

Details of experimental procedure are described elsewhere (*4,5*). In simple terms, jets were produced by ejecting liquid from a constant pressure reservoir through horizontal stainless steel hypodermic tubes. A high speed flash photograph allowed measurement of breakup lengths directly from the negative.

The liquids studied were polyisobutylene (PIB) solutions in tetralin. Molecular weights had been previously determined from intrinsic viscosity measurements in benzene (*2*). Surface tension was measured with a DuNouy Tensiometer. Viscosities were determined with capillary and concentric cylinder rotational viscometers. In some cases a Cannon-Fenske viscometer was used for viscosity measurements are very low shear rates. Solution properties are given in Table 1. At shear rates consistent with the capillary flows studied here (>10^4sec. $^{-1}$), the solutions are clearly non-Newtonian with respect to viscous flow, as can be seen in Figure 1. The development of significant normal stresses is evidenced by the degree of expansion of the jets upon ejection from the capillary (*9*), as shown in Figure 1.

Capillary diameters were in the range 0.0315 to 0.137 cm., and, unless otherwise noted, lengths were sufficient to allow complete development of the equilibrium velocity and stress profiles.

TABLE 1. SOLUTION PROPERTIES AT 25.0°C.

Polymer	c (wt. %)	η_0 (Poise)	σ (dynes/cm.)	
L100				
$M_\eta = 7.3 \times 10^5$	3.78	2.00	29	
	3.17	1.13	30	all densities
	2.40	0.534	31	are 0.97
	2.30	0.480	32	g./cc.
	2.21	0.434	32	
	1.26	0.150	33	
B100				
$M_\eta = 1.25 \times 10^5$	2.59	1.76	27.6	
	2.45	1.21	28	
	1.91	0.678	30	

RESULTS

The primary data for each run consisted of a series of breakup length measurements as a function of velocity. At low velocities the breakup length is a linear function of velocity, while at higher velocities the breakup length is observed to go through a maximum and then to decrease. The appearance of the maximum is associated with the effects of the ambient medium, and has been studied in detail for Newtonian fluids, and reported elsewhere (*3*). In the study reported here, only the linear, low speed region of breakup is considered.

Newtonian Fluids

Because the zero shear viscosities of the polymer solutions were relatively large, it was thought advisable to first extend the Newtonian data of Grant (*4*) into the region of high viscosity. The Newtonian theory, given in Equation (1), was tested by Grant, and it was found that the constant C_1 was a function of Z. It is believed that part of this dependence on Z is an artifact of Grant's method of treating his data. The linear theory of Weber (*13*) was developed by considering wave growth on a stationary cylinder of fluid. Once the growth rate was found, the breakup length was calculated by assuming that the cylinder moves with the jet velocity. Thus the velocity

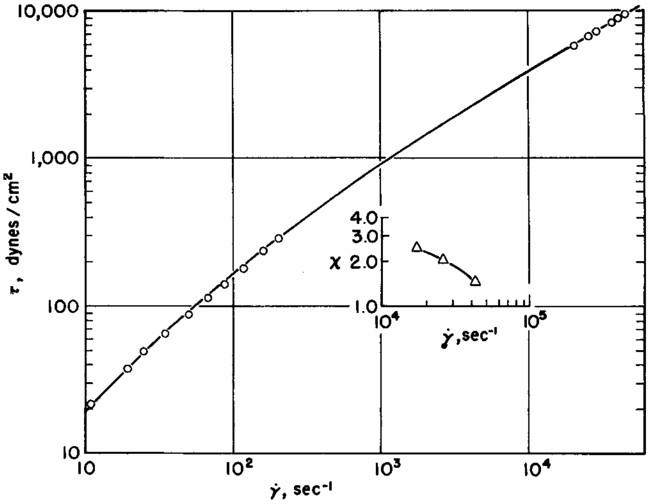

Fig. 1. Shear stress—shear rate data for 2.7% B100 in tetralin at 25.0°C. The insert shows the expansion ratio χ as a function of shear rate, in the same fluid.

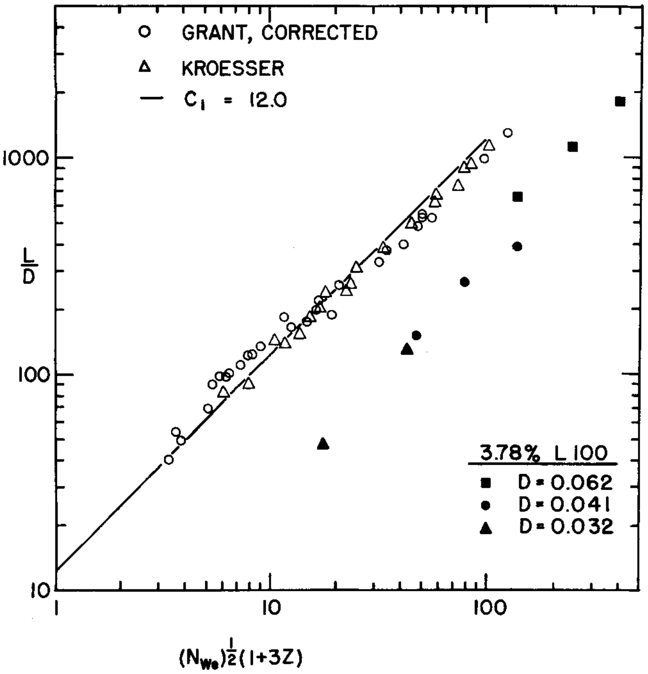

Fig. 2. Test of Equation (1) for Newtonian fluids. Filled symbols show data for a viscoelastic fluid.

and diameter used in testing the theory should be those of the jet. But Grant used the capillary diameter and the mass average velocity in the capillary in his correlations. It has been well established (*8*) that Newtonian jets undergo an expansion or contraction soon after emerging from the capillary. A more appropriate method of calculation, then, would be to use the diameter and velocity of the free jet.

Grant's data (*4*) and the additional data of this study have been plotted by using the jet values for D and V, as shown in Figure 2. A least squares line of unit slope through the data gives a value of 12.0 for C_1 .The line of unit slope is not the best fit of the data, however. The best (least squares) straight line through the data has a slope of 0.87, with a standard deviation of slope of 0.015. The slope is thus statistically different from 1.0.

If individual sets of data (that is, at fixed Z) are fitted with lines of unit slope, then a possible dependence of C_1 on Z can be tested. This is shown in Figure 3. A least squares line through these data has a slope of —0.05 and a standard deviation of a slope of 0.015. This result is considerably different from that shown by Grant (compare Figure 3 with the figure 8 of reference *4*) and simply reflects the fact that jet values for D and V should be used in correlating the data. This apparent Ohnesorge number dependence of C_1 is so weak that it probably does not warrant detailed investigation.

Viscoelastic Fluids

Breakup data obtained with viscoelastic fluids can be compared with Newtonian data, and typical results for one solution are shown in Figure 2. One observes a distinct reduction of breakup length. The largest Ohnesorge number for the viscoelastic data shown in Figure 2 is only slightly higher (2.1) than the largest Ohnesorge number for which the data in Figure 3 have been taken. As Figure 3 indicates, it is not likely that one can ascribe the marked reduction in stability to the (weak) Ohnesorge number effect. Hence it must be explained in terms of viscoelastic properties.

The non-Newtonian shear behavior of the fluids investigated is not believed to be of any significance in destabilizing the jet. If the shear rates associated with the growth of small symmetric disturbances along the jet are estimated, they are found to be well below the shear rates at which these fluids begin to deviate from Newtonian viscosity behavior. Hence the zero shear viscosity, η_0, has been used in calculating the dimensionless groups used in correlating breakup data.

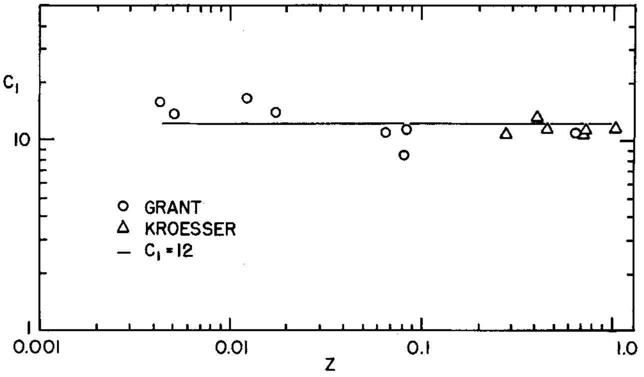

Fig. 3. Test for Ohnesorge number dependence of C_1.

In order to evaluate the theory developed here, it would be necessary to have an estimate of the relaxation time λ_1. Direct measurements of stress relaxation in such dilute solutions as used here are extremely difficult, since the times are of the order of milliseconds and smaller. Relaxation times can be inferred from measurements of non-Newtonian viscosity (*2*) or normal stress development (*11*). Such measurements from nonlinear phenomena do not necessarily give values consistent with the relaxation time of a linear Maxwell model. In fact, normal stress measurements show a relaxation time which depends upon shear rate (*11*).

An alternate approach is to estimate λ_1 on the basis of some molecular theory of viscoelasticity, such as the Bueche theory (*1*), which suggests that the relaxation time is given as

$$\lambda_1 = \frac{12}{\pi^2} \frac{(\eta_0 - \eta_s)M}{cRT} \tag{6}$$

The great advantage of this approach is that λ_1 may be calculated from a knowledge of the concentration, temperature, molecular weight, and the zero shear and solvent viscosities. Such information is easily obtained and generally known for the solutions of interest.

Experimental tests of the Bueche (or other molecular) theory indicate that Equation (6) is not quantitatively correct (*7*, Chap. 4). Support for the use of Bueche's λ_1 as a correlating parameter for viscoelastic response, especially in PIB solutions (*2*), is fairly positive [(*7*) Chap. 5]. Because of this, and in view of the simplicity with which Equation (6) gives an estimate for λ_1, Bueche's relaxation time will be used in the calculation of the elasticity number.

In view of the quantitative uncertainty of the value of λ_1, the elasticity number must be used here only as a rank-

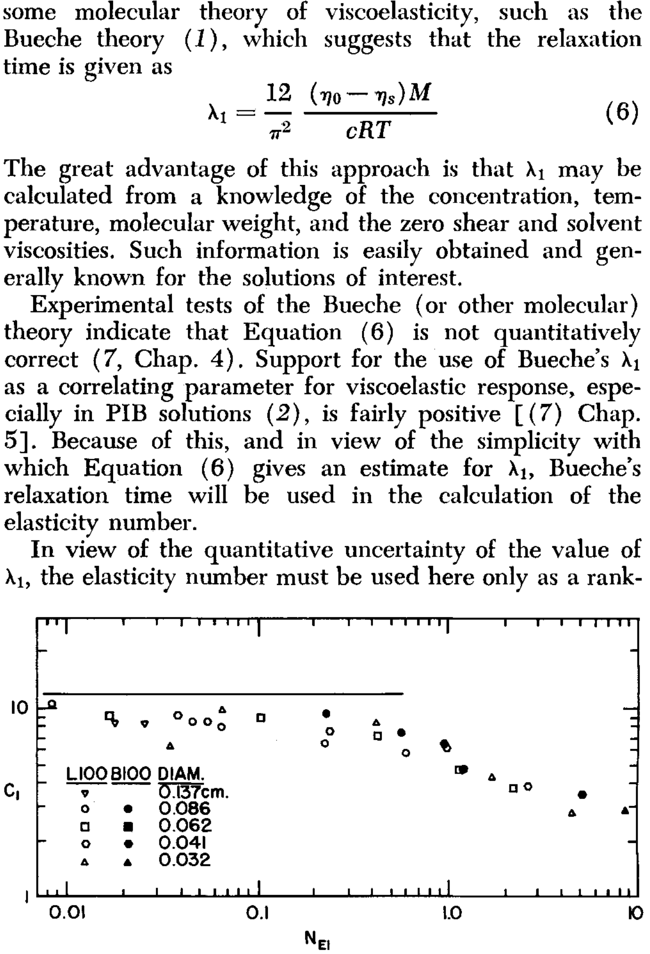

Fig. 4. Test of Equation (5) for viscoleastic fluids. The solid line is $C_1 = 12.0$

ing parameter. For a given polymer-solvent system (in this case, PIB-Tetralin)it is assumed that the elasticity number based on Equation (6) gives a measure of the relative importance of elastic effects in modifying the growth of disturbances along a jet.

The significance, and utility, of the elasticity number so defined is shown in Figure 4, where C_1, defined by Equation (5), is shown as a function of N_{El}. The use of Bueche's relaxation time allows a correlation of the data obtained with various solutions of two PIB samples of different molecular weight, for which λ_1 ranges over one order of magnitude, and N_{El} ranges over three orders of magnitude. Thus the elasticity number is shown to be a useful correlating parameter in describing viscoelastic breakup data.

One of the defects of the theory presented here is its failure to account for the possible effect of normal stresses developed within the capillary shear flow. One might study this effect by comparing the stability of jets identical in every respect except the state of normal stress at the capillary exit. This may be done by using capillaries of different lengths.

Normal stresses are relatively high in the entry region of a capillary as a result of the elastic response of the fluid to acceleration. A portion of the stress relaxes as the fluid moves down the capillary, until the stress is in equilibrium with the fully developed shear field within the capillary. Normal stress relaxation within a capillary has been studied by Powell (*11*).

These ideas led to a series of stability measurements under such conditions that the only variable was tube length. Grant and Middleman (*4*) showed that the tube length has no effect on breakup lengths of Newtonian fluids so long as the data were in the linear (low speed) region of the breakup curve. Figure 5 shows C_1 (normalized to the value, $C_{1\infty}$, found in long tubes) as a function of tube length. The dependence is relatively weak, but well defined by the data. The observation seems stronger in the light of corresponding measurements of the dependence of the primary normal stress difference on tube length, also shown in Figure 5. Hence, viscoelastic breakup data differ from Newtonian data with regard to the appearance of an effect of tube length that it is not observed with Newtonian jets.

SUMMARY

A linear viscoelastic theory predicts that viscoelastic jets have shorter breakup lengths than Newtonian jets, at constant N_{We} and Z. The theory introduces the elasticity number as a key parameter.

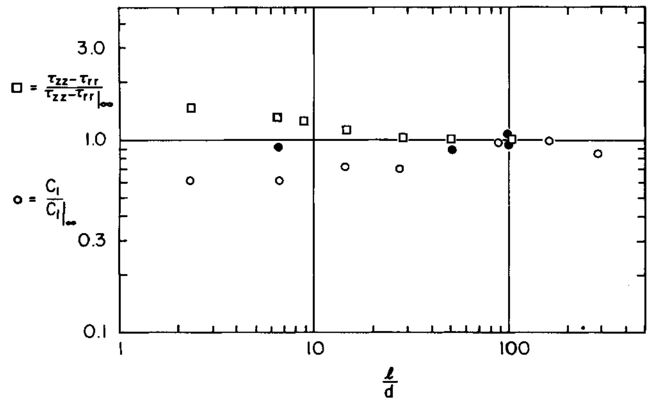

Fig. 5. Dependence of C_1 on *l/d* of capillary, compared to dependence of primary normal stress difference at capillary exit on *l/d*. Filled symbols represent dependence of C_1 on *l/d* for a Newtonian solution of comparable *Z*.

Experimental data obtained over a range of concentrations of two molecular weight samples of PIB in tetralin confirm the reduced stability of viscoelastic jets. Breakup data may be correlated with the elasticity number over a range of three orders of magnitude in N_{El}. This correlation is achieved using Bueche's molecular theory for the calculation of a relaxation time that appears in N_{El}.

The theory fails to account for the effect of normal stresses generated within the capillary. Experiments indicate a dependence of breakup length on tube length that may be associated with a normal stress effect.

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NOTATION

C_1 = constant defined in Equation (1)
C_2 = constant defined in Equation (3)
c = concentration of polymer in solution, g./cc.
d = capillary diameter, cm.
D = jet diameter, cm.
L = breakup length, cm.
l = capillary length, cm.
M = molecular weight of polymer, g./g.-mole
M_η = viscosity average molecular weight, g./g.-mole
N_{El} = $\lambda_1\eta_0/\rho D^2$ = elasticity number
N_{We} = $DV^2\rho/\sigma$ = Weber number
N_{Ws} = $\lambda_1 V/D$ = Weissenberg number
N_{Re} = $DV\rho/\eta_0$ = Reynolds number
R = 8.314×10^7 g. sq.cm./sec.^{2} g. mole °K. = gas constant
T = absolute temperature, °K.
V = jet velocity, cm./sec.
Z = $\eta_0/\sqrt{D\rho\sigma}$ = Ohnesorge number

Greek Letters

$\dot{\gamma}$ = shear rate, sec.$^{-1}$
Δ = rate of deformation tensor
η_0 = zero shear viscosity, poise
λ_1 = relaxation time, sec.
ρ = density, g./cc.
σ = surface tension, dynes/cm.
τ = stress tensor
χ = ratio of jet diameter to capillary diameter

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