Stability of Vertical Jets of Non-Newtonian Fluids

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It is a well-known fact that certain fluids are stringy, that is, when a finger is suddenly withdrawn from these fluids long thin filaments are formed which are remarkably stable to the surface forces naturally tending to decompose them into droplets. Many polymer solutions, particularly those possessing elastic properties, exhibit this behavior. An immediate demonstration of this phenomenon is obtained by wetting the tips of the thumb and forefinger with saliva, touching the two together and then separating them abruptly a distance of 1 to 2 cm. Such filaments may remain unbroken for several ten's of seconds. When these kinds of fluids are extruded through nozzles into air, a similar kind of stability is observed: far from the nozzle the filament forms into beads connected by long thin strands of fluid.

The interesting point in all this is that water or other purely viscous fluids are not stringy. These kinds of fluids break into droplets cleanly, more or less in accordance with Rayleigh's theory. A thorough explanation of the stringiness phenomenon is not now available, but it seems clear that those perturbations normally present on the surface of the liquid column, which tend to break it up, are damped. Thus the length of jets of viscoelastic fluids extruded into air might be expected to exceed the lengths of jets of purely viscous fluids similarly extruded. Some evidence has been presented which suggests the converse is true however (*1, 2*). Because of the great commercial interest in the spinning of polymer fibers and in the atomization of certain high molecular weight fuels, it is of value to know more about this phenomenon. Therefore further examination of the breakup problem is warranted.

RELATED WORK

A theory for the breakup of cylinders of inviscid fluids by surface tension was developed by Rayleigh (*3*). He showed that there is a tendency toward the development of beadlike swellings and contractions along the column. These swellings and contractions, initially only infinitesimal in size, grow at an exponential rate until ultimately the cylinder is broken into drops. This rather idealized treatment of the problem (small perturbation, inviscid fluid) was subsequently modified by Weber (*4*) by incorporating the viscosity of the jetting fluid and also the interactions between it and an inviscid ambient fluid. He solved the Navier-Stokes and continuity equations for a jet moving with uniform velocity u_0 in a zero body force field and showed that viscosity has a stabilizing influence on the jet. Weber assumed that the unbroken length of the filament could be obtained with negligible error if the rate of growth of the disturbance is taken to hold right up to disintegration. This last assumption is generally acceptable, because once the disturbance reaches finite size, its rate of growth is such that the time from there to drop formation is but a small fraction of the total time for breakup. Thus if T_b is the time for breakup to occur and u_0 is the velocity of efflux of the jet, the unbroken length L of the jet is

$$L = u_0 T_b \tag{1}$$

T_b is determined by the rate of growth of the most critical disturbance in a manner described by Levich (*5*).

Weber subsequently showed that this relationship could be rewritten in the form

$$L/D = CA \tag{2}$$

where

$$A \equiv (N_{We})^{1/2}\left[1 + 3\frac{(N_{We})^{1/2}}{N_{Re}}\right]$$

C is a constant which Weber found from experimental data to be equal to 12 for water jets in air. Grant and Middleman (*6*) and subsequently Fenn and Middleman (*7*) showed that Equation (2) is correct for low velocities of efflux when the jets are horizontal. As we shall see shortly this equation fails for jets directed vertically, since the velocity of the jet does not remain constant. Equation (1), on the other hand, still holds if u is taken to be the convected velocity of the jet.

Middleman (*8*), paralleling Weber's development, analyzed the problem of the breakup of unaccelerated jets of slightly viscoelastic fluids ejected into air. The fluid was assumed to have a linear constitutive equation of the form

$$\tau_{ij} + \lambda_1 \frac{\partial \tau_{ij}}{\partial t} = \mu\left[d_{ij} + \lambda_2 \frac{\partial d_{ij}}{\partial t}\right] \tag{3}$$

with constants λ_1 and λ_2 small and nearly equal. The unbroken length L_{nn} of the slightly viscoelastic jet relative to the length L_n of a Newtonian fluid with the same Weber and Reynolds numbers was shown to be

$$\frac{L_{nn}}{L_n} = 1 - \frac{\lambda_2(\lambda_1/\lambda_2 - 1)u}{a\ A} \tag{4}$$

Thus the length of a jet of viscoelastic fluid is shorter, longer, or equal to one formed of a Newtonian fluid, depending on the value of λ_1/λ_2. Since for most fluids $\lambda_1/\lambda_2 > 1$, Equation (3) indicates that $L_{nn}/L_n < 1$. (Note that the relative length also depends on the diameter $2a$ of the jet.)

The above model was modified somewhat by Kroesser and Middleman (by taking $\lambda_2 = 0$ and putting $\nu = \nu_0$, the

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Table 1. Rheological Data

Polymer	ppm.	K	n	ν_0
Separan AP-30	300	0.16	0.82	15
	600	0.45	0.73	40
Carbopol 934	1200	0.82	0.73	100
	2000	45.9	0.41	10^5

K and n are defined by the power law relationship

$$\tau = -K\left(\frac{\partial u}{\partial y}\right)^{n-1}\frac{\partial u}{\partial y}$$

zero shear rate kinematic viscosity) (*1*) and tested using tetralin solutions of polyisobutylene. Their data show that jets of these materials are indeed shorter than jets of Newtonian fluids having the same Weber and Reynolds numbers when the zero-shear viscosity is used as the characteristic viscosity for the fluid, although the solutions used were hardly only slightly viscoelastic as called for by the theory. The justification for using the zero-shear viscosity is not made clear however; it seems to be more intuitive than rigorous, particularly in the case of the concentrated solutions.

EXPERIMENTAL PROCEDURE

A transparent plastic tank (the reservoir), 20 cm. square and 1.5 m. deep, was used to generate the jets for this study. The orifice from which the fluid emerged was located centrally in the bottom of this tank and flush with the internal surface. Each orifice plate was carefully machined from 5.1 cm. diam. 0.64 cm. thick brass stock and was given a sharp edge by milling a 60-deg. cone angle in the hole on the downstream side. Diameters of 0.251, 0.338, 0.478, and 0.640 cm. were selected. The reservoir was mounted sufficiently high on a vertical wall so that jets up to 2.75 m. long could be examined.

Since the length of the jet is not constant for any given head but fluctuates with time about some average value, many observations are ordinarily required to determine this average. However, it was found that a stroboscopic light set at a frequency close to the frequency of the drop formation, which is surprisingly regular, greatly facilitated the determination of this length. Quite arbitrarily, but consistent with convention (*1, 2, 4*), the continuous length has been taken to be the average distance from the orifice to the point where distinct droplets are formed.

Even though jets of the viscoelastic fluids break-up in a manner quite different from that in which the Newtonian fluids break up, this definition for breakup length is retained.

The behavior of the viscoelastic fluid is of primary interest, of course, but since no data could be found for jets of Newtonian fluids in a vertical configuration, it was deemed necessary to collect these data as well. The Newtonian fluids investigated were water and solutions of sucrose possessing viscosities of 2.8 and 8.3 centipoise. The non-Newtonian fluids were prepared by dissolving in water 2,000 p.p.m. of Carbopol 934, a carboxyvinyl polymer of extremely high molecular weight, and 600 p.p.m. of Separan AP-30, a partially hydrolyzed high molecular weight polyacrylamide polymer. These solutions were subsequently diluted and run through the orifices again. Table 1 gives the viscosity of the polymer solutions in terms of a power law relationship. Approximate values for the zero-shear viscosity ν_0 are also tabulated. The viscosities were checked before and after each series of experiments to determine whether the polymer had been degraded. No significant changes in the shear properties of the fluid were detected. In some cases the first jet experiment was repeated in order to ascertain whether small amounts of polymer degradation, not detectable in the viscometric measurements, had any influence on the breakup length. No changes were observed. Attempts to measure the elastic properties of these fluids using the Weissenberg Rheogoniometer proved fruitless; the relaxation times (based on a Maxwellian fluid) obtained were very small and inconsistent.

Surface tensions were determined for the various solutions using two techniques: capillary rise and a variation of the Wilhelmy slide method (*9*). For the Newtonian fluids, the values were all very nearly that of water (about 73 dynes/cm) a variation of less than ± 3% was detected.

It was not possible to measure the surface tension of the non-Newtonian fluids unambiguously by the capillary rise method. For these fluids a modified Wilhelmy slide method, which gives values of surface tension from measurements of the force required to pull a cylinder through the surface, was found to be more reliable. By this method of testing, the Carbopol 934 additive did not affect the surface tension of the water even at concentrations up to 2,000 p.p.m. For the 600 p.p.m. Separan AP-30 solutions, however, values ranging from 70 to 73 dynes/cm. were measured, depending on the rate of withdrawal of the cylinder. (For more concentrated solutions the values were even more rate dependent.)

RESULTS AND DISCUSSION

Newtonian Fluids

It is important to realize at this point that Equation (2) applies specifically to a column of liquid having initially a flat velocity profile, moving at a constant velocity u_0, and having an average radius a, such that at any axial position it is equal to the initial radius of the jet. Because of the extremely long lengths obtained with viscoelastic fluids for even the smallest diameter orifices used in this work, it was necessary to orient the jets vertically (in the gravity field) to avoid problems associated with the general curvature of the jet. In so doing, the jets were subjected to accelerating forces. This problem has not been solved analytically. However, observations seem to show that no new phenomena were introduced by the presence of the acceleration terms in the equations of motion; the Newtonian jets were still broken up at the low velocities by symmetrical disturbances. This fact may be demonstrated in a more rigorous manner by calculating the time for breakup for the jets. According to Rayleigh's argument a column of any given fluid is most unstable to one particular disturbance which has a prescribed rate of growth (and which depends on the diameter of the column) and so, a prescribed breakup time T_b. Thus the distance downstream of the orifice where breakup occurs should depend only on the velocity with which the fluid travels. For the

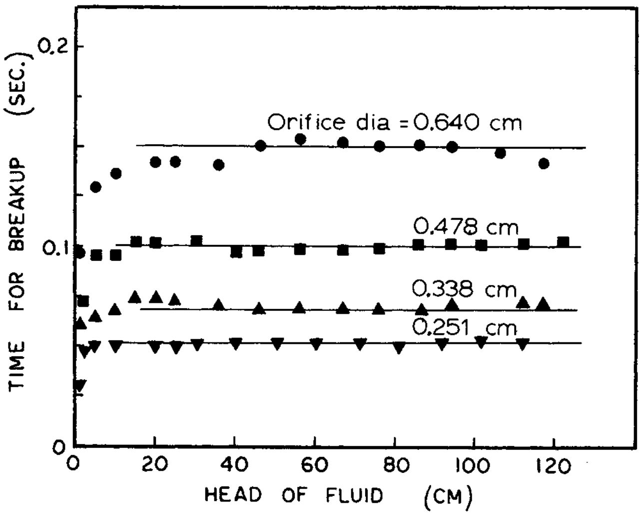

Fig. 1. Constancy of the time for breakup for the water jets.

horizontal jet the time for breakup is obtained by multiplying Equation (2) by D/u_0. If the definitions for the Weber and Reynolds numbers are introduced, the result is

$$T_b \equiv \frac{L}{u_0} = C\left[\left(\frac{\rho D^3}{\sigma}\right)^{1/2} + 3\frac{\rho \nu}{\sigma}\right] \qquad (5)$$

Inspection of the data of references 6 and 7 show T_b = constant for all but the highest velocities for any given tube in accordance with the Rayleigh mechanism for breakup.

For the accelerated jet the velocity along it is given by

$$u_z = [2g(H+z)]^{1/2} \qquad (6)$$

This is Bernoulli's equation applied to the flow; H is the head of fluid in the tank and z is the distance along the jet measured from the orifice. Since $u_z = dz/dt$, the above equation may be integrated to obtain the relationship between time and distance. The result is

$$T_b = \left[\frac{2}{g}(H+L)\right]^{1/2} - \left[\frac{2}{g}H\right]^{1/2} \qquad (7)$$

If Rayleigh's argument is to apply to the accelerated jet, the value of T_b calculated from this equation must be independent of H since this quantity only determines the efflux velocity u_0. This independence is demonstrated in Figure 1 for water for several orifice sizes, and is in fact obtained for all the fluids examined for values of $H > 15$ cm. Thus it may be concluded that the Rayleigh mechanism controls the breakup process for low velocity jets regardless of their orientation.

Since exact solutions for the vertical orientation are unavailable for any kind of fluid and since there seem to be no data in the literature for this configuration, we thought it necessary for this work to make measurements on jets of Newtonian as well as non-Newtonian liquids.

Figures 2 and 3 contain some of the data on the breakup for the jets formed of water and of water solutions of sucrose. These data are plotted according to Equation (2) even though it may not apply. The second term contained in the brackets of this equation is at most of magnitude 6×10^{-2} (for the 8.3 centipoise sucrose solution and the 0.251-cm. diam. orifice). Thus viscosity would appear to be an unimportant parameter for these jets under these conditions of flow. For the computations the velocity is taken to be the velocity of discharge from the orifice and the diameter is that of the orifice.

The data are seen to approach asymptotically the line for the unaccelerated jet [Equation (2) with $c = 12$] as the Weber number approaches zero. Thus the deviation from the line may be interpreted as the effect of the acceleration forces, at least for values of $A < 20$ where the data fall more or less along a single curve. For larger values of this parameter the data for each orifice size follow distinctly different curves. It appears that the deviations from Equation (2) decrease with increasing fluid viscosity and, possibly, with decreasing orifice diameter. The reason for the breaks in some of the curves for values of $A \approx 20$ is not understood (they are not, for example, laminar-turbulence transitions) but they seem to be associated with flow conditions at the orifice. With square-edged orifices, separation was observed at the trailing edge for small values of A and from the leading edge for larger values of this parameter. Distinctly different curves were obtained for the two conditions.

Some of the data from Figures 2 and 3 are replotted in Figure 4, this time using values for A based on the velocity of the fluid at the point of breakup rather than the discharge velocity. This velocity is given by Equation (4).

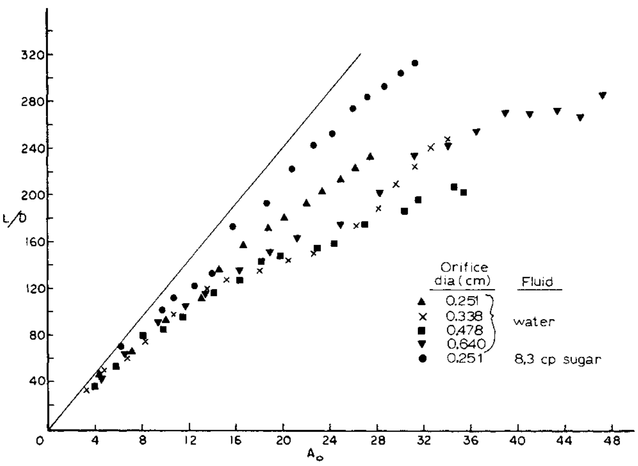

Fig. 2. The effect of acceleration forces on the length of the jets of Newtonian fluids. The line represents Equation (2) for the unaccelerated jets.

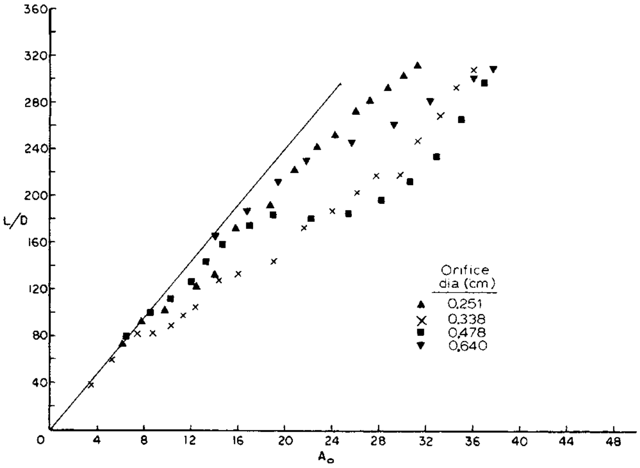

Fig. 3. The effect of acceleration forces on the length of the jets formed from the 8.3 centipoise sucrose solution. The line represents Equation (2) for the unaccelerated jets.

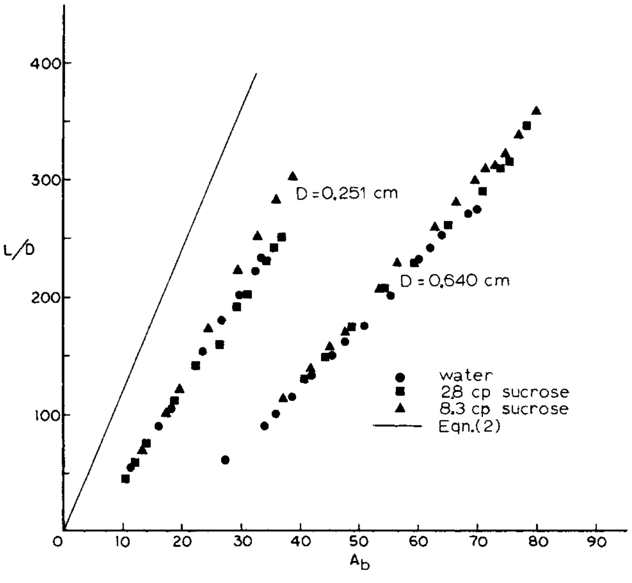

Fig. 4. Jet length correlated according to Equation (2) using the velocity of the fluid at the point of breakup.

It is evident that the dimensionless length of the jet is more nearly a linear function of A_b than it is of A, although now the data no longer are fitted by a universal function; the diameter of the orifice appears as a parameter. The fact that lines drawn through the data of Figure 4 do not pass through the origin is of no particular consequence, because, for example, at very small discharge velocities the fluids drip from the orifices. What is significant is that the slopes of lines drawn through these data are less than that of the line which represents Equation (2). Thus, for the same efflux conditions, the accelerated jets tend to be shorter than the unaccelerated ones. Since the significance of the relationships shown in Figure 4 may not be demonstrated analytically, no more shall be said about it here. We shall see shortly however that similar relationships appear to hold for the non-Newtonian fluids as well.

Non-Newtonian Fluids

If Equation (7) is used to calculate the breakup time for the jets of non-Newtonian fluids, the results are as shown in Figure 5. For a significant range of discharge conditions then, Rayleigh instabilities may be assumed to control the breakup of these jets as well. Note that the breakup time appears to increase as the elastic properties of fluid increase but not as the zero-shear viscosity increases.

For purposes of comparison it is desirable to plot the data for the non-Newtonian fluids in the manner of Figures 2 and 3. However two problems arise: the second term in the brackets of Equation (2) may become important, but the two parameters, viscosity and surface tension, which appear in this term are not necessarily identifiable in a way that is not arbitrary. Kroesser and Middleman (*1*) suggest the use of the zero-shear viscosity ν_0, and in addition introduce another parameter λ_1, a relaxation time for the fluid. Equation (2) is thus modified. Golden, Pfeffer, and Shinnar (*2*), on the other hand, suggest that the proper choice for the viscosity is that viscosity which corresponds to the shear rate at the origin of the jet flow for those fluids having characteristic times which are large compared to the breakup time. For those fluids having characteristic times small compared to the breakup time, the zero-shear viscosity is to be used.

Some of the data obtained with the two Carbopol 934 solutions jetting from the 0.64-cm. diam. orifice are presented in Figure 6; viscous effects [that is, the second term in the brackets of Equation (2)] have been neglected. For comparison the results for the Newtonian fluids (from Figure 4) are included. Comparison is made using the velocity of the fluids at the point of breakup. The use of this velocity is arbitrary at this time; the discharge velocity could just as well have been used, but the data presentation would have been less linear. The measurements show the unbroken length of these jets to increase more or less linearly with the velocity at breakup just as it did for the Newtonian fluids. More significantly, however, the unbroken lengths are changed very little from those obtained with water. Even the jets formed from the 2,000 p.p.m. solution, which has a zero-shear viscosity somewhat in excess of 10^5 centipoise, have lengths which barely exceed those formed of the 8.3 centipoise sucrose solutions and then only at the highest rates of discharge by this comparison.

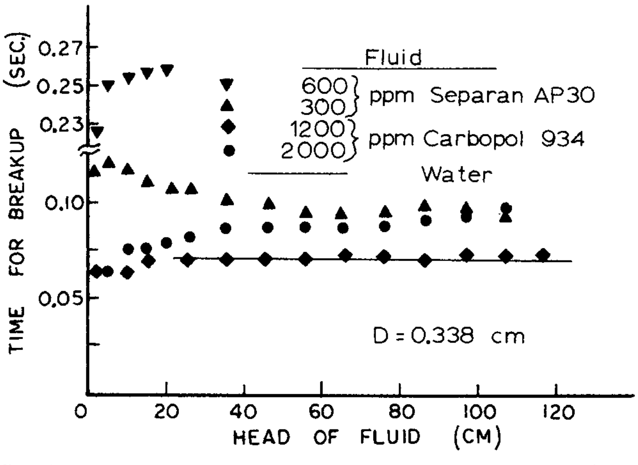

Fig. 5. Constancy of the time for breakup for the jets of non-Newtonian fluids.

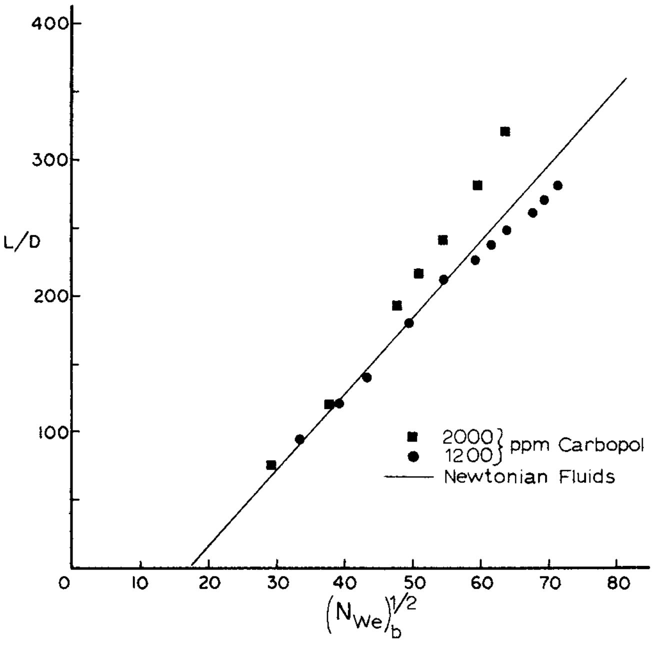

Fig. 6. The length of the jets formed of the Carbopol 934 solutions are compared with the length of jets formed of water. Viscous effects are neglected and the velocity at the point of breakup is used.

For the Carbopol solutions $\lambda_1 \approx 0$ (since these solutions are effectively inelastic), and so, following Kroesser and Middleman (*1*), Equation (2), with the viscosity replaced by the zero-shear viscosity, applies. But these viscosities are 100 and 10^5 centipoise, respectively, for the 1,200 and the 2,000 p.p.m. solutions. Thus if the second term in the brackets of Equation (2) is retained and the zero-shear viscosities inserted, it is obvious that the data for these solutions will not overlay the data for the Newtonian fluids but will fall far to the right and below the curve in Figure 6. Thus, when the jet lengths are compared for flows having the same Reynolds and Weber numbers, those formed of the Carbopol solutions are shorter as predicted (*1*, *8*). It should be noted however that a separate curve will be obtained for each solution and each orifice diameter.

If λ_1 is taken to be the characteristic time for the Carbopol solutions, then of course the Golden, Pfeffer, and Shinnar model (*2*) gives the same results as does that of Kroesser and Middleman (*1*). If however some other characteristic time which is large (and undefined) is more appropriate, then, according to the first-mentioned authors, the apparent viscosity must be used instead of the zero-shear viscosity. For the efflux rate considered in this work the corresponding viscosities are only of the order of a few centipoise for these solutions and so the viscous contribution becomes negligible and the lengths are once again as shown in Figure 6.

The water solutions containing the Separan AP-30 additive behave differently from both the Newtonian and the non-Newtonian inelastic fluids. The series of photographs presented in Figures 7 through 9 of reference *2* demonstrate the ability of these fluids to resist destructive capillary forces to form long thin filaments of fluid connecting consecutive beads. (Similar photographs were obtained in this work.) Ultimately, when the connecting threads do break, the broken sections invariably flow back into the droplets. No daughter drops, those small drops formed by the independent breakup of the connecting filaments, appear as is usually the case with the water and Carbopol solutions.

The stability of these jets is shown quantitatively in Figure 7. Once again the viscous effects have been neglected in order to obtain some basis for comparison. If the viscous term is retained and the zero-shear viscosity inserted, the effect is only to shift the data to the right, somewhat without materially altering either the shape or the relative positions of the curves since these viscosities are not large for these fluids. The evidence then is that at least for a range of Separan AP-30 concentrations, jets of these fluids are longer than jets of Newtonian fluids if they are compared at the same N_{We} (and N_{Re}).

As indicated earlier, although these solutions are elastic (at least they are in higher concentrations), relaxation times based on the assumption of a Maxwellian fluid are too small to be measured. Even those relaxation times calculated from the Bueche theory turn out to be too small to give elasticity corrections of sufficient magnitude to correct Equation (2) as described in reference *1*. Thus one might conclude that elasticity plays no role in the breakup of these jets. However if this conclusion is accepted, then it must also be accepted that Equation (2) and its modifications are invalid for these flows. Clearly more work and more thought is needed on this problem.

Plots of L/D versus A are not shown because no new information is contained in them (and further their significance is unclear). Suffice it to say that the weak tendency seen in Figure 4 for the data to separate into distinct curves with positions depending on the properties of the fluid (for a given size orifice) is no longer weak. Each curve does lie above the curve for water however, indicating that jets of these fluids are longer than the equivalent water jets.

Finally, the relationship between the breakup time and the orifice diameter for the accelerated jets is presented in Figure 8 and is seen to be approximately linear over the range of flow rates studied. A 3/2 power variation is predicted by Equation (4) for the horizontal jet. The importance of the viscometric properties of the fluid to the flow phenomenon is brought out most clearly in this figure. The breakup times for the 600 p.p.m. Separan solution are shown to be from two to three times as large as those for any of the other fluids, even the 2,000 p.p.m. Carbopol solution which has a zero-shear viscosity of 10^5 centipoise.

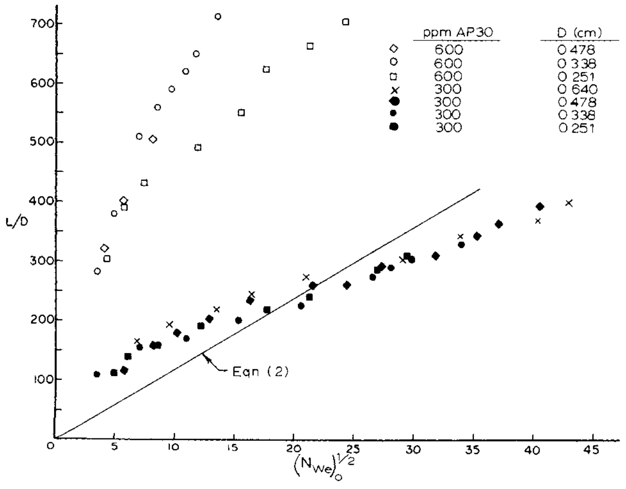

Fig. 7. The length of the jets formed of the Separan AP-30 solutions are compared with the length of jets formed of water. Viscous effects are neglected and the velocity at the point of discharge is used.

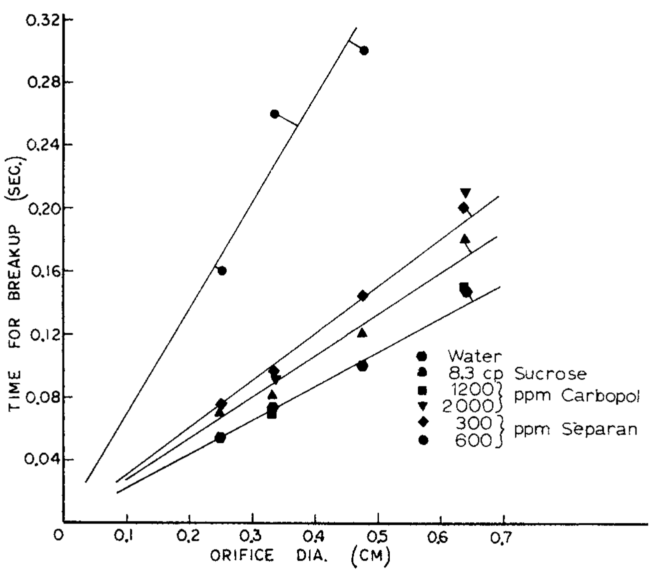

Fig. 8. Dependence of the breakup time on the orifice size for the various fields.

SUMMARY

The lengths of vertical jets formed of various kinds of fluids have been measured and, in the case of the Newtonian fluids, compared with the theoretical (and experimental) results for the length of horizontal jets. Not unexpectedly the length was found to be smaller for the vertical orientation.

More significantly however the lengths of the vertical jets were found to be strongly influenced by the character of the fluid. Purely viscous non-Newtonian fluids, like those formed by adding a carboxyvinyl polymer to water, were found to have lengths only slightly different than those obtained with the pure water. The addition of small quantities of a high molecular weight polyacrylamide polymer, on the other hand, caused a marked increase in the length of the jet. Since the latter solutions possess significant elastic properties (at least they do at moderate levels of polymer concentration), whereas the former solutions do not (even at high concentrations), normal stresses may be expected to play a role in the breakup process. Since the equations of motion have not been solved for these kinds of flow, the idea remains unevaluated.

Whether the jets of non-Newtonian fluids are longer or shorter than jets of Newtonian fluids depends on the basis for comparison. If we accept the analysis given by Middleman and co-workers and compare the jets on the basis of equal Reynolds and Weber numbers, then the jets formed of the carboxyvinyl polymer solutions are significantly shorter than the water jets. Contrary to their experimental work with polyisobutylene solutions in tetralin and with horizontal jets, however, the jets formed of the polyacrylamide solutions are longer than the water jets for sufficiently high concentrations and rates of efflux. No explanation is offered for this discrepancy.

NOTATION

A $\equiv (N_{We})^{1/2}\,[1 + 3(N_{We})^{1/2}\,(N_{Re})^{-1}]$
a = average radius of jet, cm.
d_{ij} = rate of deformation tensor, sec.$^{-1}$
D = diameter of orifice, cm.
g = gravitational constant
H = height of liquid above orifice, cm.
K = viscosity coefficient in Ostwald-de-Waele model, dyne/(sq.cm.) (sec.^{n})
L = unbroken length of the jet, cm.
L_n = unbroken length of Newtonian jet, cm.
L_{nn} = unbroken length of viscoelastic jet, cm.
n = viscosity index in Ostwald-de-Waele model, dimensionless
N_{Re} = Reynolds number, dimensionless = (Du/ν)
N_{We} = Weber number, dimensionless = $(\rho U^2 D/\sigma)$
t = time, sec.
T_b = breakup time, sec.
u_z = velocity in z direction, cm./sec.
u_0 = initial axial velocity, cm./sec.
z = axial direction, cm.

Greek Letters

λ_1 = relaxation time, sec.
ρ = density, g./cu.cm.
σ = surface tension, dyne/cm.
τ_{ij} = stress, dynes/sq.cm.
ν_0 = zero-shear kinematic viscosity, sq.cm./sec.

Subscripts

0 = values based on orifice data
b = values based on data at the breakup point

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Tubular Reactor Steady State and Stability Characteristics

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I. Effect of Axial Mixing

This paper reports results of a numerical study of the stability characteristics of the nonadiabatic tubular reactor with axial mixing. It differs from much of the prior work on similar systems by its inclusion of a heat transfer term in the modeling equations. Assuming that the tubular reactor with axial mixing (TRAM) has a flat velocity profile, is not packed, and has no radial concentration and temperature gradients, mass and energy conservation statements are, respectively

$$\frac{\partial y}{\partial \tau} = \frac{1}{N_{Pe'}}\frac{\partial^2 y}{\partial z^2} - \frac{\partial y}{\partial z} - \mathcal{R}(y, \eta) \tag{1}$$

$$\frac{\partial \eta}{\partial \tau} = \frac{1}{N_{Pe}}\frac{\partial^2 \eta}{\partial z^2} - \frac{\partial \eta}{\partial z} + \mathcal{R}(y, \eta) + U_r(\eta_w - \eta) \tag{2}$$

The commonly used boundary conditions are

$$z = 0, \quad \left.\frac{1}{N_{Pe'}}\frac{\partial y}{\partial z}\right|_0 = y(0) - 1 \tag{3}$$

$$\left.\frac{1}{N_{Pe}}\frac{\partial \eta}{\partial z}\right|_0 = \eta(0) - \eta_F$$

$$z = 1, \quad \left.\frac{\partial y}{\partial z}\right|_1 = \left.\frac{\partial \eta}{\partial z}\right|_1 = 0 \tag{4}$$

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