

$c_a$	= heat capacity of adsorbate and condensate (kJ/mol °K)
$c_f$	= heat capacity of gas phase (kJ/kg °K)
$c_s$	= heat capacity of adsorbent (kJ/kg °K)
$h_f$	= enthalpy of gas phase (kJ/kg)
$h_s$	= enthalpy of solid phase (kJ/kg)
$K$	= Langmuir isotherm parameter, Eq. 15
$K_0$	= constant appearing in Eq. 15
$L$	= bed length (m)
$p$	= partial pressure of solute (MPa)
$P$	= total pressure (MPa)
$q$	= solid-phase concentration (mol/kg)
$q_{\max}$	= maximum solid-phase concentration (mol/kg)
$q_{\text{sat}}$	= adsorbed-phase concentration in equilibrium with saturated vapor (mol/kg)
$Q$	= Langmuir monolayer capacity (mol/kg)
$R$	= gas constant
$t$	= time
$T$	= temperature (°K)
$T_0$	= temperature of hot inlet gas (°K)
$T_{\text{ref}}$	= reference temperature (°K)
$v$	= interstitial velocity (m/s)
$v_o$	= interstitial velocity of hot inlet gas (m/s)
$v^*$	= dimensionless velocity, Eq. 5

#### Greek Letters

$\epsilon$	= void fraction of packing
$\zeta$	= dimensionless axial coordinate, Eq. 6
$\theta$	= fractional saturation of adsorbed phase
$\lambda$	= latent heat (kJ/mol)
$\lambda_d$	= isosteric heat of desorption (kJ/mol)
$\lambda_v$	= heat of vaporization of liquid (kJ/mol)
$\rho_b$	= bulk density of packing (kg/m <sup>3</sup> )
$\rho_f$	= density of gas phase (kg/m <sup>3</sup> )
$\rho_{fo}$	= density of hot inlet gas (kg/m <sup>3</sup> )
$\tau$	= dimensionless time, Eq. 7
$\phi$	= initial fractional saturation of vapor phase in bed

#### Subscript

$i$	= stage index
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# Mixing and Fluid Dispersion of Viscous Liquids

A model based on extension and secondary motions of striations in a deforming flow is used to predict details of the mixing and dispersion of viscous liquids. Generally, extension of intermaterial area and interfacial surface occurs in stagnation regions, and breakup ensues when the striated flow has left such a region or has time to form unmixing regions of smaller scale in the stretching flow. Complex criteria for the types of mixing anticipated are based on differences in viscosity, differences in density, striation scale, rate of stretch, and, for various degrees of immiscibility, interfacial tension.

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## SCOPE

Mixing and fluid dispersion of viscous liquids have been studied in terms of specific and limiting cases with results based on complex fluid mechanical analysis. Experimental observations have been even more narrow in scope and, in the case of immiscible fluids, have been obscured by coalescence. In the

present paper, a basic stretching flow is used to distort a "discontinuous phase" as though it were part of the continuous flow. At various times during this imagined distortion stresses on the boundary of the discontinuous phase are evaluated for realizability, for direction of movement of the boundary, and for rate

of movement in a secondary flow which can be generated by these forces. Forces of the same order of magnitude and movements of less magnitude and rate will arise in the real flow.

The model is used to establish criteria for mixing and dispersion and to predict the order of magnitude of space and time scales for such processes.

## CONCLUSIONS AND SIGNIFICANCE

If the fluid of largest volume fraction is designated as continuous phase-c, small scale mixing can be treated as distortion of the discontinuous phase-d striation or dispersion element in a two-dimensional stretching frame of reference. This frame is characterized by a velocity gradient  $\alpha'_d(t)$  and can be treated as a nearly inertial frame at scales less than  $2(\mu_c/\rho\alpha'_c)^{1/2}$ .

Since mixing flows of viscous liquids usually experience periodic  $\alpha'_c$  values with frequency  $\alpha'_c$ , dispersions of size less than  $g_c\sigma/2(\mu_c - \mu_d)\alpha'_c$ , where  $\sigma$  is interfacial tension, will not be

dispersed further. Striations or dispersions greater than about one-eighth the mixing frame scale, when  $\nu_d/\nu_c \ll 1$ , and greater than about one-eighth the mixing frame scale based on  $\nu_d$ , when  $\nu_d/\nu_c \gg 1$ , are also poorly mixed or dispersed.

Mixing criteria for more general conditions can also be developed, but these are complex results for various viscosity levels and viscosity differences, density levels and density differences, ranges of value of  $\sigma$ , and available magnitudes and forms of  $\alpha'_d(t)$ .

## INTRODUCTION

Mixing and dispersion of viscous liquids at limiting conditions of small striation scales and fine dispersion sizes can be described in a relatively simple way by a stretch model (Ranz, 1979) which makes use of a single, time-dependent quantity to account for all fluid mechanical action. In this type of analysis statistical averages of the results of mixing and dispersion are formed late in the scheme of description so that the precise roles of the space averaging and time averaging can be incorporated. Early distribution of the values of important variables, in particular, striation thickness and dispersion size, can be treated as initial conditions and final distributions can be obtained by following known amounts of flow subject to a particular time dependent variable for a particular contact time.

Initial volumes of a one-phase fluid being mechanically mixed remain connected in time. Cross sectioning will reveal at any time a marbled structure. If miscible fluids of different properties are involved, their parallel structures of concentration variations will be seen. Lenticular structures might be found embedded in this striation pattern if there is considerable viscosity and density variation between mixing fluids. This form of unmixing can arise from fluid mechanical instabilities in the stretching flow field. If the fluids are immiscible, at least partially, this kind of unmixing takes the form of breakup of the dispersed phase striations into droplets or flat eel-like zones which can be frozen into the mixed structure. Small enough droplets will resist further breakup because of interfacial tension. The purpose here is to demonstrate in a simple analysis these important features of the mixing and dispersion of viscous liquids.

## MIXING FRAME OF REFERENCE AND STRETCHING FLOW FIELD

Tomotika (1936) and Hinze (1955) showed how a stretching flow was the best way to cause drop breakup. The optimum conditions for pure mechanical mixing (Ottino et al., 1979) is favorable orientation in a stretching flow. At small scales near a fluid particle all flows are stretching flows, elliptically symmetrical stagnation zones, in a frame of reference which moves with and rotates with the fluid particle (Batchelor, 1967). This resolved motion is characterized by the rate of strain tensor and dilation rate. Now a small fluid volume in such a flow will stretch or compress depending on the orientation of its longest dimension with respect to the axis of stretch. Moreover, flow in another Lagrangian frame of reference based on a particle in a lamina or striation surface and the direction normal to that surface will appear to have also a local straining motion made up of appropriate components of the fluid straining motion. Because the striation and dispersion sizes of practical interest lie within the scales of the local stretching flow, it is this

special Lagrangian frame which is chosen for analysis of mixing and dispersion of viscous liquids.

There is much dynamic uniformity and symmetry in a stagnation or stretching flow. The flow appears the same to any fluid particle momentarily in the local region, that is, any particle could be the center of the frame of reference. For a pure liquid, viscous dissipation of energy within any volume is equal to viscous work rate on that volume. A straight material line remains straight and a material plane remains planar while increasing dimension in the direction of stretch. A material volume soon become a thinning slab (Fisher, 1968). If the special frame is an inertial frame for the local flow, the local flow is constrained by mechanical laws as well as by continuity to a two-dimensional stagnation flow. Such a two-dimensional flow exists in simple shear, in laminar flow, in a vortex, and in the microscales of turbulence (Batchelor, 1956). Flow in a lamina-based frame properly oriented will have the same characteristics or will soon assume the favorable orientation. The need for "turning" or "folding" a mixing flow arises from the need to keep reorienting the lamina coming from a mixing region, whose mixing strength in the lamina frame has decreased in time, when the lamina is fed into a new region of hydrodynamic stretch.

The flow field in the local mixing frame of reference can be characterized (Ranz, 1979) by a single time-dependent quantity  $\alpha(t)$ . The mixing frame flow is

$$v_x = -\alpha x, v_y = \alpha y, v_z = 0 \quad (1)$$

Arbitrarily  $x$  is chosen as the principle direction of stretch and  $y$  is chosen as the principle direction of contraction  $\alpha$  should have a negative value for mixing. If  $\alpha$  has a positive value, unmixing is occurring. In mixing and dispersion flows the integrated value of  $\alpha$  is negative over long times.

Mixing of an initial striation thickness  $s_0$  oriented in the  $y$ -direction normal to the striation surface, so long as it remains in a deformational frame with the same flow as Eq. 1, is at a maximum rate given by a constant  $\alpha$ . At time  $t$  the striation thickness  $s$  is

$$s/s_0 = \exp(-\alpha't) \quad (2)$$

where  $\alpha' = -\alpha$  is the velocity gradient in the direction of stretch.

The striation in passage through this frame thins exponentially in time, indeed, at a maximum possible rate of mixing. Coalescence of colliding bubbles and droplets is possible only because there is such a rapid thinning of continuous fluid layered between mobile interfaces. However, we are interested here only in the mixing and dispersion characteristics of this optimum flow which occurs in front of the leading edges of mixing impellers, static mixers, and egg beaters, and in unmixing tendencies in such a flow when striation instability and breakup phenomena occur.

Figure 1a shows two striations thinning in their passage through a stagnation flow. The circular frame around the striation is the mixing frame, and the mixing frame does not rotate. The entering

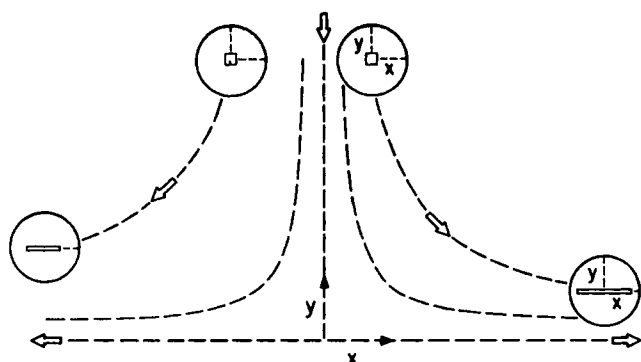


Figure 1a. Stretch of fluid volumes in a stagnation flow; mixing frame does not rotate. In mixing frame  $v_x = \alpha'_c x$  and  $v_y = -\alpha'_c y$  where  $\alpha'_c = \alpha'_c(t)$ .

striation is shown as a dispersion element. If the long axis of a striation element was normal to  $y$ , then its entering orientation would be optimal for further stretch and mixing. However, if the long axis of the entering striation was perpendicular to  $x$ , the striation would be thickened and unmixed by the flow.

### MIXING IN A SHEAR FLOW

In simple two-dimensional shear, where the hydrodynamic flow is  $v'_x = Gy'$ ,  $v'_y = v'_z = 0$ , the mixing frame of a striation thickness  $s_0$  measured initially in the  $x'$ -direction has subsequently an

$$\alpha = \frac{-G(Gt)}{1 + (Gt)^2} \quad (3)$$

Initially,  $\alpha$  is zero, but it rapidly decreases to a minimum value of  $-G/2$  at  $Gt = 1$ , after which and at long times  $\alpha$  becomes equal to  $-t^{-1}$ . This behavior of the mixing frame is a characteristic of most flows. For mixing the flow must be reoriented periodically and the lamina "folded" so that the value of  $\alpha$  is periodically restored. In the present case the striation's length is initially oriented in the direction of the velocity gradient. Had it been oriented in the direction of flow,  $s$  of a long striation would have remained  $s_0$  in its middle and no thinning would be observed except at its ends. However, if there were an interfacial tension at the striation surface or  $y$ -gradients in density and viscosity, instabilities in the striation could occur, and unmixing or breakup phenomena might ensue even in this apparent no- or little-mixing situation.

Figure 1b shows a striation thinning in a velocity gradient. Note that the mixing frame rotates and soon orients in the gradient frame in such a way that the stretching rate is inversely proportional to time. A deformation frame would continue to rotate at the initial rotational velocity of the mixing frame.

### PERIODIC PHENOMENA IN MIXING AND DISPERSION

In the mixing frame of a particular dispersion element or particular section of a striation,  $\alpha(t)$  must be generally negative in value even though it can and necessarily may have to assume positive values during passage of the mixing frame through a mixing device. In the usual mixer,  $\alpha(t)$  for a particular mixing frame assumes a periodic value that is characterized by a frequency and an amplitude with a negative time average. These separate quantities, along with the mechanical properties of the fluids being mixed, must be considered in any design for mixing and dispersion of viscous liquids.

Qualitatively, in the case of dispersion, the striation may be stretched and break apart in times of order of the reciprocal of the frequency of  $\alpha$ . The striation may have been over-extended during stretch and still break during a period when  $\alpha \approx 0$ , indicating an additional role for the form of  $\alpha(t)$ . If the amplitude is too small or the period too short for the dispersed element to reach an over-

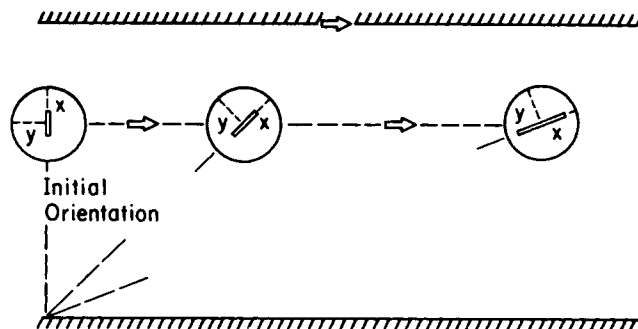


Figure 1b. Stretch of fluid volume in shear flow; mixing frame rotates at decreasing rate. In mixing frame  $\alpha'_c = G(Gt)/(1 + (Gt)^2)$  where  $G$  is shear rate.

extended shape, the dispersion merely survives shape oscillations in the mixer.

In the case of miscible fluids an equilibrium surface tension cannot exist, but the striation can be limited in stretch if finite flow instabilities can occur in times of the order of the reciprocal of the frequency of  $\alpha$ . Furthermore, if the diffusional time scale is much longer than the period of  $\alpha$ , mechanical unmixing will occur in the cycle whenever  $\alpha(t)$  becomes positive in value.

### LIMITING PHENOMENA

Practical results derived from the foregoing resolution of the local mixing process can be relatively simple. Some can result from detailing conditions for possible motions of intermaterial areas. First consider the scale  $\lambda_x = \lambda_y$  of the local flow in the mixing frame. Since the pressure environment has been assumed to be uniformly symmetrical and the effect of rotation negligible, the inertial stress in the mixing frame, say  $\rho_c v_x^2/2g_c$  at  $y = 0$ , must be less than the viscous normal stress, say  $-2(\mu_c/g_c)\partial v_x/\partial x$ . Here subscript-c denotes a continuous phase of largest volume fraction. Substitution of Eq. 1 gives

$$\rho_c \alpha' x^2/2 \leq 2\mu_c \alpha' \text{ or } \lambda_x = \lambda_y = 2(\mu_c/\rho_c \alpha')^{1/2} \quad (4)$$

analogous to the Kolmogoroff scale for turbulent mixing. For laminar mixing this scale can become quite large, say  $10^{-2}\text{m}$ , much more than the dispersion size of usual interest.

In the mixing of dissimilar fluids, those which have widely different viscosities and densities, those that are slowly miscible, and those that are to some degree immiscible, limiting phenomena become more complex. However, simple models can predict important features of the mixing and dispersion processes. At the largest scales of a mixing zone and at continuum model scales much larger than striation size, an effective viscosity and an average density can be approximated as the volume weighted averages of viscosities and densities of the fluids being mixed. At the scale of a single striation a more detailed model gives much more information on the limiting phenomena.

Figure 2 shows a striation of discontinuous fluid-d being stretched (or contracted) by continuous fluid-c in a mixing frame which is based on the striation. In mixing of similar miscible fluids a striation is a continuous slab; but if the striation "breaks up" or is from a previous dispersion, it will be modeled as a rectangular slab of half width- $b$  and half length- $a$  rather than a long ellipsoid or lenticular structure. The flow of the two fluids is considered to be completely dominated by the flow field outside the mixing frame. One now considers forces on the intermaterial areas of the model slab and the secondary motions which would arise to relieve these forces.

Consider first the limits of dispersion in this mixing zone where  $\alpha'$  is imposed by the far flow. A force balance on the postulated phase-d boundary at  $x = a$  should give a measure of stretch of the discontinuous region which is the maximum possible, in the sense

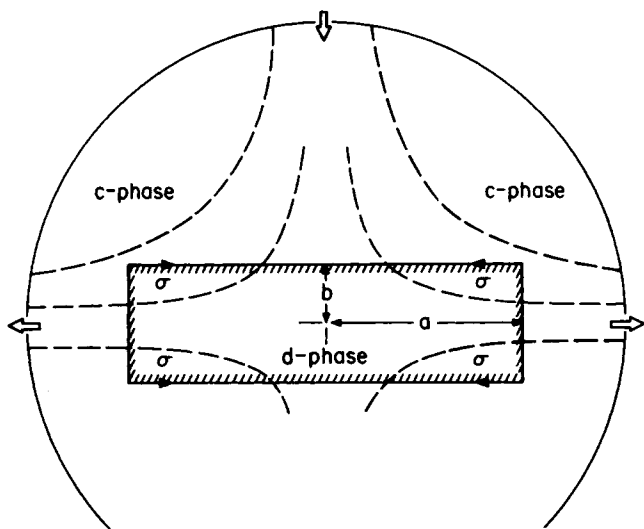


Figure 2. Simplified dispersion element in mixing frame; continuity requires that  $ab = a_0b_0 = \text{constant}$ . Assume flow field is  $v_x = \alpha'_c x$  and  $v_y = -\alpha'_c y$  everywhere and consider net stress on boundaries.

that  $b$  will grow smaller until surface tension equals the net stretching force.

Difference in  
viscous stress

$$\int_0^b \left\{ -\frac{2}{g_c} (\mu_d - \mu_c) \frac{\partial v_x}{\partial x} \right|_{x=a} - (\rho_d - \rho_c) \frac{v_x^2}{g_c} \Big|_{x=a} - (\rho_d - \rho_c) \frac{v_x^2 + v_y^2}{2g_c} \Big|_{x=a, y} \right\} dy \gtrsim \sigma$$

Difference in momentum flow      Difference in mixing frame pressure

which, on substituting the conditions and assumptions of Figure 2, becomes

$$\frac{2}{g_c} (\mu_c - \mu_d) \alpha'_c b + \frac{(\rho_d - \rho_c) \alpha'^2_c a_0^2 b_0^2}{2g_c b} - \frac{(\rho_d - \rho_c) \alpha'^2_c b^3}{2g_c 3} \gtrsim \sigma \quad (5)$$

where the last term on the left side of the inequality will be negligible when  $a > b$ . If  $\mu_c > \mu_d$  viscosity is aiding the stretch; if  $\mu_d > \mu_c$  viscosity is hindering the stretch. If  $\rho_d > \rho_c$  inertia is aiding stretch; if  $\rho_c > \rho_d$  inertia is hindering stretch. According to this simplified model, limits of dispersion for a mixing zone characterized by  $\alpha'$  would be given by  $b = b_0 = a_0$  or

$$2(\mu_c - \mu_d) \alpha'_c b_0 + (1/3)(\rho_d - \rho_c) \alpha'^2_c b_0^3 \approx g_c \sigma \quad (6)$$

The  $b_0$  of Eq. 6 should be able to survive a mixing zone  $\alpha'$  because interfacial tension and forces related to lack of relative flow are sufficient to keep a droplet intact.

The striation model of Figure 2 stretching in accord with Eq. 5 cannot be fully realized when  $\mu_d > \mu_c$  and  $\rho_c > \rho_d$ . Some other geometry for the striation must be considered, perhaps one which ends in a cusp with  $b = 0$ .

Consider the net forces on the boundary of the discontinuous phase- $d$  at  $y = b$ , particularly those forces which should tend to thicken the striation if it is to continue to stretch in ellipsoidal or filament form, or to thin the striation, if these forces can generate an unmixing flow in a secondary mixing frame of scale less than half length- $a$ . First, because  $\partial v_x / \partial y = 0$ , one notes that there is no shear stress on the boundary of discontinuous phase- $d$  at  $y = b$ , nor is there any normal stress due to  $\sigma$  on this flat surface which has not yet bulged or dimpled. However, the net normal stress difference

on the boundary at  $y = b$  is

$$-\frac{2}{g_c} (\mu_d - \mu_c) \frac{\partial v_y}{\partial y} \Big|_{y=b} - (\rho_d - \rho_c) \frac{v_y^2}{g_c} \Big|_{y=b} - (\rho_d - \rho_c) \frac{v_x^2 + v_y^2}{2g_c} \Big|_{x, y=b}$$

which, on substituting the conditions and assumptions of Figure 2, becomes

$$4(\mu_d - \mu_c) \alpha'_c + (\rho_c - \rho_d)(\alpha'^2_c x^2 + 3\alpha'^2_c b^2) \lesssim 0 \quad (7)$$

for a net thinning stress on the boundary at  $y = b$  and at  $x = x$  if the conditions pictured in Figure 2 were realized. Thus, if the left hand side of the relation was positive in value, the actual boundary would be at  $y > b$ . If the value was zero, the actual boundary would be at  $y = b$ . If the value was negative, the actual boundary would be at  $y < b$ , that is, the striation will be thinner at this point. The equality gives the point along the boundary at which the intermaterial surface experiences no net normal stress. This should be of the order of the half wave length of instabilities.

$$x_{\lambda/2}^2 \approx \frac{4}{\alpha'_c} \frac{(\mu_d - \mu_c)}{(\rho_d - \rho_c)} - 3b_{\lambda}^2 \quad (8)$$

a curious relationship for half wave length  $x_{\lambda/2}$  of a disturbance which will grow when the striation thickness is  $b_{\lambda}$  and the rate of stretch is  $\alpha'_c$ . Eq. 8 predicts that mixing, in the sense of continual laminar stretch, may be limited when  $\mu_d > \mu_c$  and  $\rho_d > \rho_c$  and also when  $\mu_c > \mu_d$  and  $\rho_c > \rho_d$ . If the instabilities can grow fast enough, the expected result is a line of ellipsoids, in the case of  $\sigma$  with significant value, or lens-like structures, in the case of  $\sigma = 0$ , where the line is stretching uniformly at rate  $\alpha'_c$  and where the unmixing regions or less mixed regions are moving farther apart in the  $x$ -direction. The model can be continued by assuming that  $x_{\lambda/2} = a_{\lambda}$  initially in the breakup region and by continuing the requirement that  $a_{\lambda} b_{\lambda} = a_0 b_0$ . However, speculations on subsequent events should be approached with caution. One must remember that a finite time and a particular range for  $x_{\lambda/2}$  are needed to obtain an observable instability.

Further constraints on the possible instability predicted by Eq. 8 are provided by Eq. 4 which would have  $x_{\lambda/2} \leq \lambda_x$  and by Eq. 2 which would have  $3b_{\lambda}^2 \leq 3b_0^2/e^2$  because  $\alpha'_c$  has only a lifetime of the order of  $1/\alpha'_c$  in real mixers. The indicated range of  $\Delta\mu/\Delta\rho$  for an observable inertial-viscous interaction is  $\nu_c \gtrsim \Delta\mu/\Delta\rho \gtrsim 3\alpha'_c b_0^2/4e^2$ .

#### TIME-DEPENDENT PHENOMENA

Since small variations in the location of the intermaterial areas considered in the previous section require that  $\mu_d \approx \mu_c$  and  $\rho_d \approx \rho_c$  and since the time of action in real mixers will be only of order  $1/\alpha'_c$ , it is worthwhile to consider what happens to a striation of fluid- $d$  in the mixing frame of Figure 2 if initially it is stationary with  $b = b_0$  and if suddenly  $v_x = \alpha'_c x$  in phase- $c$  for a limited period of time  $1/\alpha'_c$ .

Now  $v_{x,c} = \alpha'_c x$  at  $y > b + \delta_c$  and  $v_{x,d} = 0$  at  $y < b - \delta_d$  where, as a crude approximation, assume that  $\delta_c = (\nu_c t)^{1/2}$  and  $\delta_d = (\nu_d t)^{1/2}$  describe the growth of the shear region. Here  $b$  will be assumed constant at  $b_0$  and  $t$  will be retained as real time for short times even though it should be warped time (Ranz, 1979) in a mixing frame with an effective  $\alpha'_d(t)$  and  $b = b(t)$ . For miscible fluids there will also be gradients in viscosity. This additional complexity, which might generate an apparent  $\sigma$ , is also neglected.

For short time actions, this simple model gives for linear velocity profiles and a  $\alpha'_d(t)$  for an averaged flow in the dispersed phase- $d$

$$\frac{\alpha'_d}{\alpha'_c} = \frac{(\nu_c t)^{1/2}/2b_0}{(\rho_d/\rho_c) + (\nu_c/\nu_d)^{1/2}} \text{ for } b > (\nu_d t)^{1/2} \quad (9)$$

One can now specify a possibly useful criterion that

$$b_0 \lesssim \frac{(1/4)(\nu_c/\alpha'_c)^{1/2}}{(\rho_d/\rho_c) + (\nu_c/\nu_d)^{1/2}} \text{ or } \frac{(1/4)(\nu_d/\alpha'_c)^{1/2}}{(\rho_d/\rho_c)(\nu_d/\nu_c)^{1/2} + 1} \quad (10)$$

for significant thinning of the striation during time  $1/\alpha'_c$ .  
 If  $\rho_d = \rho_c$  and  $\nu_d/\nu_c \ll 1$ ,  $b_0 \lesssim (1/4)(\nu_d/\alpha'_c)^{1/2}$  for thinning.  
 If  $\rho_d = \rho_c$  and  $\nu_d/\nu_c \gg 1$ ,  $b_0 \lesssim (1/4)(\nu_c/\alpha'_c)^{1/2}$  for thinning.  
 If  $\rho_d = \rho_c$ ,  $\nu_d/\nu_c = 1$ , and  $\sigma = 0$ , one knows that  $\alpha'_d = \alpha'_c = \alpha'(t)$ .

However, if  $\sigma$  is finite, the criterion suggests that  $b_0 \lesssim (1/8)(\nu_d/\alpha'_c)^{1/2}$  or  $(1/8)(\nu_c/\alpha'_c)^{1/2}$ . These considerations place a complex restriction on mixing and dispersion of viscous liquids. If  $\nu_d$  is low and  $\nu_c$  is high,  $\alpha'_c$  is usually also low because  $\nu_c$  is high. For example, if  $\nu_d = 10^{-2}$  cm<sup>2</sup>/s and  $\nu_c/\nu_d \gtrsim 10^2$  and if  $\alpha'_c = 10$  s<sup>-1</sup>, a  $b_0$  larger than about 80  $\mu$ m would resist being mixed or dispersed in that mixing element. If  $\nu_d$  is high and  $\nu_c$  is low,  $\alpha'_c$  is usually also high because  $\nu_c$  is low. For example, if  $\nu_c = 10^{-2}$  cm<sup>2</sup>/s and  $\nu_d/\nu_c \gtrsim 10^2$  and if  $\alpha'_c = 100$  s<sup>-1</sup>, a  $b_0$  larger than about 25  $\mu$ m would resist being mixed or dispersed in that mixing element. Furthermore, if the flow is experiencing a periodic distortion and the striations do not achieve reorientation between stages where  $\alpha'_c$  is effective only for time  $1/\alpha'_c$ , the striations will tend merely to oscillate in thickness and intermaterial area.

If a breakup regime with finite  $\sigma$  is established, an unmixing frame of scale  $a_\lambda$  forms. This is a secondary flow with negative  $\alpha_\lambda$  or positive  $\alpha$ . It is embedded in the  $\alpha'$  flow or continues independently when the fragmenting striation moves into a region where  $\alpha'$  tends to zero. Using a reversed flow interpretation of Figure 2, an  $\alpha'_\lambda$  can be established from balanced forces in Eq. 5. Initial conditions are  $b = b_\lambda$  when  $t = 0$ . Such an exercise determines only a time scale in which breakup can occur. If the time is longer than observable time, breakup may never be observed. Figure 3 pictures the regions in which droplets are forming by the postulated mechanism. If  $a_0 = a_\lambda$  and  $b_0 = b_\lambda$  in the equality of Eq. 5 and if one notes that  $db/dt = -\alpha'_\lambda b$ , a relationship for  $b(t)$  results. For example, if  $\rho_d = \rho_c$  and  $\mu_d > \mu_c$ ,  $db/dt = g_c \sigma/2 \mu_d$ , giving a characteristic droplet forming time of order  $2\mu_d(a_\lambda b_\lambda)^{1/2}/g_c \sigma$ .

#### RELATIONSHIP OF STRESS DIFFERENCE AND STRESS RATIO ANALYSIS TO RECENT EXPERIMENTS AND THEORY

Experimental results with finite  $\sigma$  have been presented and reviewed by Grace (1971). Unfortunately the presentation of data does not include detailed information on  $\rho_c$  and  $\rho_d$ . The model of constant  $\alpha'_c$  for a period  $1/\alpha'_c$  was only roughly achieved in Couette experiments, but a constant  $\alpha'_c$  for  $t > 0$  was achieved for very long times and breakup times for extended striations were observed in a multirotor device. The foregoing order-of-magnitude analysis appears to be consistent with these results. Acrivos and Lo (1978), in a complex development of the shape and flows of a slender drop in an axisymmetrical extensional flow have introduced the role of

inertia to formal treatment of a particular case of steady shape and breakup configuration. The foregoing analysis can anticipate the salient features of such a particular case.

Here possible actions are evaluated in terms of stress differences, stress ratios, and stress equalities. The idea is to develop means whereby a large number of mixing and dispersion cases can be surveyed without too much effort. One identifies cases of most practical interest, for further study, and finds ways of accomplishing mixing, for difficult systems.

#### MIXING IN THE STAGNATION REGION OF A MIXING ELEMENT

The stagnation region in front of mixing elements is not only a deformational frame of reference but a mixing frame as well, especially in a relative flow of viscous liquids. A complication is the boundary layer near the surface of the element. A stretching and oriented striation passing through the boundary layer may be stretched less even though it spends more time in the mixing region.

Consider a greatly simplified stagnation region ahead of some mixing element having an effective half-width  $L$  with a flat surface at  $y = 0$ . The main flow is two-dimensional.  $v_x = \alpha'x$  and  $v_y = -\alpha'y$  with  $\alpha' = V_0/L$  where  $V_0$  is the relative velocity between element and fluid. The Reynolds number of the flow is  $\alpha'L^2/\nu$ , and the boundary layer thickness is of order  $(\nu/\alpha')^{1/2}$ . The ratio of the boundary layer thickness to  $L$  is the inverse of the square root of the Reynolds number. If  $L = 1$  cm and  $\alpha' = 10^2$  s<sup>-1</sup>, the stagnation region will have been completely modified to a boundary layer in the  $y$ -direction if  $\nu = 10^2$  cm<sup>2</sup>/s.

If the boundary layer flow can be neglected, flow enters the mixing region at  $y = L$  for  $-L < x < L$  and leaves at  $x = \pm L$  for  $0 < y < L$ . Any  $s_0$  oriented in the  $y$ -direction entering the stretching region at  $x(t = 0)$  will leave at  $y = x(t = 0)$  in the same orientation with  $s = s_0(x(t = 0)/L)$ . If the striation passes through  $(L, L)$ ,  $s$  remains  $s_0$ . If it passes through  $(L, 0)$ ,  $s$  tends to zero as it forever approaches the stagnation point. For uniform concentration of striation  $s_0$  and equally probable entry points,  $s = s_0/2$  on the average in the flow from the stretch region. This model can be extended to develop distributions of striation and dispersion sizes based on successive passages through the same or similar mixing regions. Figure 1a illustrates this particular case.

In the case of a deep boundary layer,  $y(t)$  for the favorably oriented striation continues to be equal to  $y(t = 0)\exp(-\alpha't)$  where  $\alpha'$  is constant in time. The striation retains the same orientation of being stretched in the  $x$ -direction, but the stretching  $\alpha'_x$  of the mixing frame following the striation decreases with time as the striation moves farther into the boundary layer.

With a boundary layer of significant depth the mixing region dimension in the  $y$ -direction will tend to increase to allow for the necessary outflow. In general, the effect of viscosity will have to be included to discover how much fluid is stretched and how it is stretched in passing through a mixing zone.

#### QUESTIONS POSED BY STRETCH MODEL WHICH MAY FIND ANSWERS THROUGH FURTHER ANALYSIS

A major purpose of the foregoing development is to show how truly complex are mixing and dispersion of viscous fluids or of fluids of widely different viscosities and densities. Apparently no simple rules will be found by massive experimentation. An adequate analysis for experimental testing becomes a logical maze in which one can become easily lost.

Consider, for example, the question of which fluid is dispersed and which is continuous. It has been assumed tacitly that the dispersed fluid is the one of much less volume fraction in the mixture. If the volume fractions are nearly equal, not only is the dispersed phase designation dependent on how the mixing is begun, but phase inversion may eventually result on repeated coagulation and redispersion. The phase which preferentially wets the walls of a mixing device or coagulates on a wall will be continuous near the

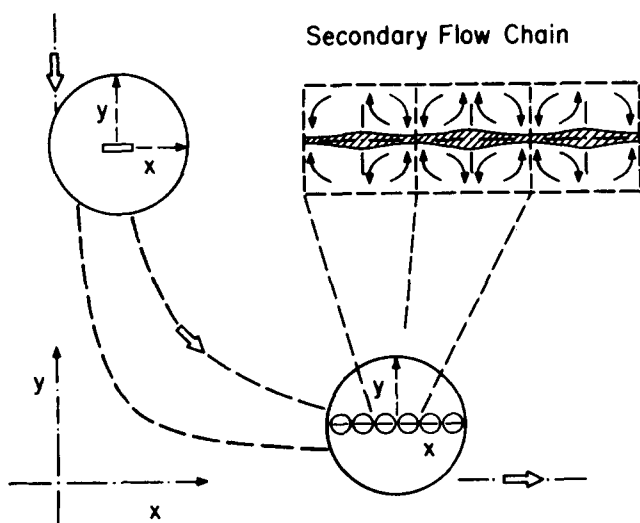


Figure 3. Formation of secondary flows and breakup of a striation being stretched.

wall. Initial and boundary conditions can affect the subsequent mixing and dispersing flow. More than one mixing flow regime may be possible, with unexpected switching among regimes.

Non-uniformity of striation thickness is the result of initial or boundary conditions which is preserved as average thickness decreases and intermaterial area increases. Diffusional uniformity, which is supposed to be hastened by mechanical thinning, actually may be retarded by the fact that largest lamina of different fluids find themselves eventually far apart in a sea of merged striations of lesser size. This particular complexity may be unique to mixing processes and may be more of a problem than practitioners of mixing arts realize.

It should be emphasized that a shear flow does not represent a region of best mixing. Indeed, this is where coagulation (gradient coagulation of dispersed droplets) occurs. Droplets, encountering one another because of relative motion in a shear flow, form a mixing frame with  $y$ -direction between drop centers with a striation of continuous phase between approaching drop surfaces. The striation thins during the first half of the encounter and thickens during the second half, if the striation does not become so thin during the earlier half that it ruptures. Figure 2 is appropriate to picture the encounter, but the stagnation flow is now axisymmetrical.  $\alpha(t)$  would depend on the fluid mechanics of the encounter, being at first negative and then positive, tending to zero value before and after the encounter and at the moment of passing. If coagulation ensues, the encounter ends with a single drop rotating in the shear field.

Concentrations of surfactant in the intermaterial surfaces may impart rigidity to the phase interface. Thinning rates could be highly retarded. This leads to a final caution that surfactants affect mixing processes by altering the mechanical properties of the intermaterial areas.

#### NOTATION

$a$	= half length of striation
$b$	= half width of striation
$G$	= velocity gradient in shear flow
$s$	= striation thickness
$t$	= time
$v_x, v_y$	= component fluid velocities in mixing (deformational) frame
$v'_x, v'_y$	= component fluid velocities in inertial frame
$x, y$	= coordinates in mixing frame
$x', y'$	= coordinates in inertial frame

#### Greek Letters

$\alpha$	= Stretch parameter in mixing frame
$\alpha'$	= Velocity gradient in the direction of stretch in a two-dimensional stagnation flow
$\delta$	= Laminar boundary layer thickness in a striation being impulsively stretched
$\lambda$	= Scale of mixing frame
$\mu$	= Fluid viscosity
$\nu$	= $\mu/\rho$
$\rho$	= Fluid density
$\sigma$	= Interfacial tension

#### Subscripts

$c$	= Continuous phase
$d$	= Discontinuous and dispersed phase

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