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# Droplet breakup mechanisms: Stepwise equilibrium versus transient dispersion

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## Synopsis

In dispersive mixing of immiscible liquids the minimum attainable dropsize is often deduced from the critical value of the Capillary number (the ratio of the shear stress to the interfacial stress) necessary for drop breakup under quasiequilibrium conditions. The critical Capillary number shows a minimum if the viscosity ratio between dispersed and continuous phase is about one. Hence, it is commonly accepted that the finest morphology is obtained if both viscosities match. In practical mixing devices, however, small drops are formed by a transient mechanism of thread breakup during extension rather than by stepwise breakup under equilibrium conditions. For Newtonian liquids, a comparison is made between the drop sizes resulting from a stepwise equilibrium and a transient breakup mechanism. Generally, the transient mechanism yields smaller drops and, more interestingly, a higher viscosity ratio between the dispersed and continuous phases results in a finer morphology, as already indicated by Tjahjadi and Ottino (1991). In the present paper the comparison is elaborated over a broad range of the relevant parameters while a compact illustrative presentation of the results is given to stress the possible consequences for practical blend morphologies.

## INTRODUCTION

Many industrial processes involve dispersion of immiscible liquid/liquid systems; the final morphology resulting from the mixing process influences the mechanical and other properties of the product. An elementary step in the mixing process is the deformation and breakup of a single dispersed drop in the flow field of a second immiscible liquid. Drop deformation is promoted by the deforming shear stress  $\tau$  imposed by the flow and counteracted by the interfacial stress  $\sigma/R$  (with  $\sigma$  the interfacial tension and  $R$  the drop radius) minimizing the interfacial area, thus tending to a spherical drop shape. The ratio between the competitive stresses is expressed by the Capillary number

$$Ca = \frac{\tau}{\sigma/R} = \frac{\eta_c \dot{\gamma} R}{\sigma} \quad (1)$$

with  $\eta_c$  the viscosity of the continuous phase and  $\dot{\gamma}$  the shear rate [ $= \sqrt{(2\mathbf{D}:\mathbf{D})}$ ,  $\mathbf{D}$  being the rate of deformation tensor]. If the Capillary number is small, the interfacial stress dominates and a steady ellipsoid drop shape exists. Upon excess of a critical value,  $Ca_{crit}$ , no equilibrium shape exists and the drop becomes unstable and finally breaks. It should be noticed that the critical Capillary number considers drop breakup

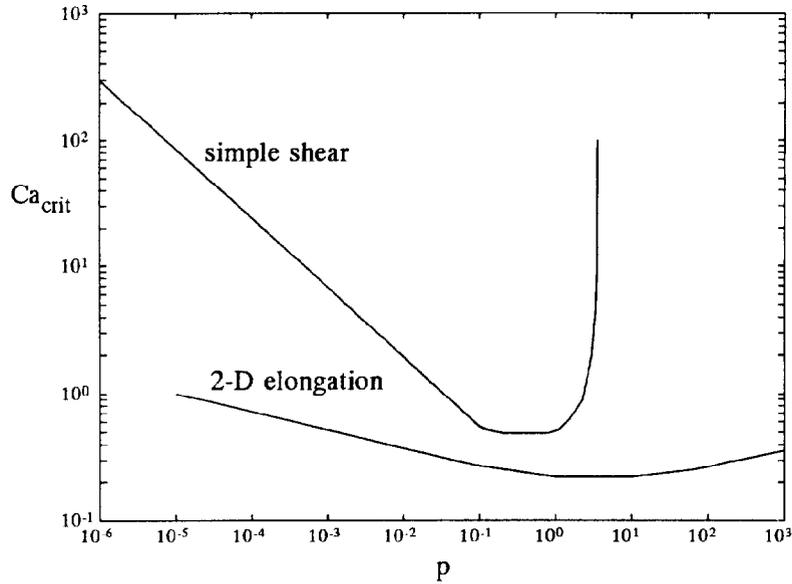


FIG. 1. Critical Capillary number vs the viscosity ratio  $p$  in simple shear and plane hyperbolic flow (after Grace, 1971).

under quasiequilibrium conditions, without including the time. Taylor (1934) and Grace (1971) showed that the critical Capillary number depends both on the type of flow and on the viscosity ratio  $p$  between dispersed and continuous phase

$$p = \eta_d / \eta_c \quad (2)$$

Figure 1 shows Grace's experimental results for Newtonian liquids. From his data it can be seen that for  $p \approx 1$  it is easiest to deform and break a drop under quasiequilibrium conditions. Furthermore, plane hyperbolic flow (2D elongation) is more effective than simple shear flow. Beyond  $p \approx 4$  it is even impossible to break a drop in simple shear caused by the rotational character of the flow. Bentley and Leal (1986) showed experimentally that intermediate flow types yield values of  $Ca_{crit}$  in between the two curves of Fig. 1. Numerical studies on the critical Capillary number in different types of flow agree with experimental data (see, e.g., Khakhar and Ottino, 1986).

The minimum attainable droplet size of a dispersion is often derived from the  $Ca_{crit}$  versus  $p$  graph: knowing the material properties and estimating the maximum deformation rate in a mixing device, the value of  $Ca_{crit}$  gives the smallest droplet size that can still be broken. Assuming the drop to break into two equal parts without any satellite droplets in between, the final droplet size is given by

$$R_{drops} = \frac{Ca_{crit}}{\eta_c \dot{\gamma} / \sigma} 2^{-1/3} \quad (3)$$

Since the value of  $Ca_{crit}$  is minimum at  $p \approx 1$ , in practice it is commonly accepted that the finest morphology is obtained if both viscosities match.

This breakup mechanism under equilibrium conditions, however, is quite unlikely to occur in real mixing processes. More realistic appears to be a transient breakup

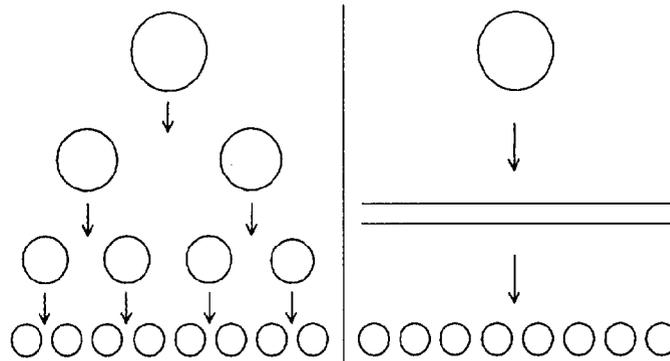


FIG. 2. Schematic representation of the two dispersion mechanisms: Left, the stepwise equilibrium mechanism of repeated breakup at  $Ca_{crit}$ ; right, the transient mechanism of thread breakup during extension.

mechanism where a drop is initially extended affinely with the flow into a thin liquid thread. The criterium for affine deformation is experimentally shown to be roughly  $Ca/Ca_{crit} > 2$  in simple shear and  $Ca/Ca_{crit} > 5$  in plane hyperbolic flow (Elemans *et al.*, 1993; Meijer and Janssen, 1993). In time, interfacial tension driven instabilities grow at the interface and finally cause the thread to disintegrate. The analysis of breakup of an extending liquid thread (Tomotika, 1936; Mikami *et al.*, 1975; Khakhar and Ottino, 1987) shows that the flow suppresses the growth of interfacial disturbances. Consequently, the thread may have thinned substantially before it finally breaks yielding a large number of small droplets.

Tjahjadi and Ottino (1991) already noted that the critical Capillary number for threads (i.e., the critical thread radius below which breakup occurs) is generally smaller than that for drops (i.e., the critical drop radius above which breakup occurs). This implies that the transient mechanism of thread breakup during extension is more effective in obtaining a fine dispersion than repeated drop breakup at  $Ca_{crit}$ . Here, the difference between the two mechanisms involved (Fig. 2) is elaborated quantitatively over a broad range of material and processing parameters. The final droplet size resulting from the stepwise equilibrium mechanism can easily be obtained from Fig. 1 using Eq. (3). The transient mechanism is somewhat more complicated and is reviewed first. For the limiting case of constant stretching rates, the results of thread breakup during extension are condensed further than was done by former authors.

### BREAKUP OF AN EXTENDING LIQUID THREAD

A liquid cylinder immersed in an immiscible fluid, quiescent or flowing, is subject to capillary instabilities at the interface. Initially, small disturbances of all wavelengths are present and develop in time. On a liquid thread in a quiescent matrix one wavelength remains dominant (i.e., has the fastest growing amplitude) during the entire disintegration process and leads to breakup. On an extending thread, however, the wavelengths are continuously stretched, hence at any moment another disturbance wavelength is dominant and the breakup of the thread is postponed when compared to a thread in a quiescent matrix. Breakup occurs as soon as the amplitude of a disturbance is magnified such that it equals the mean thread radius that continuously decreases in time.

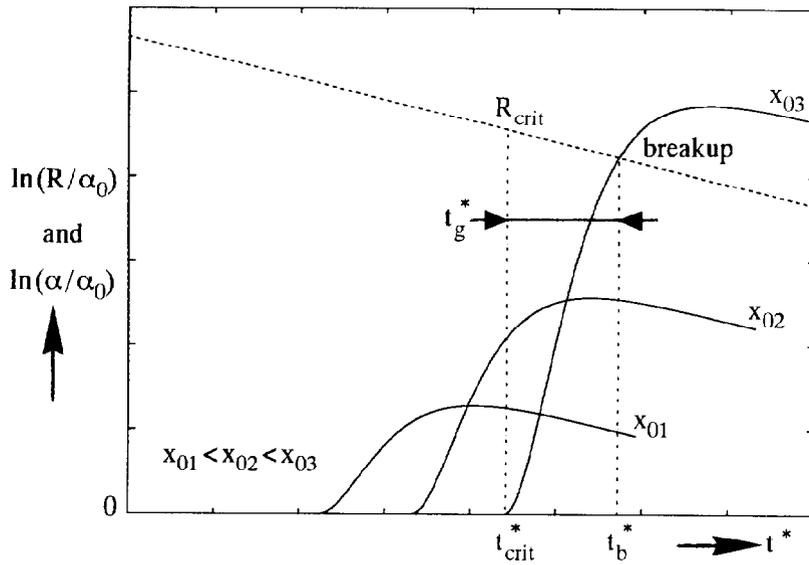


FIG. 3. Evolution of the radius  $R$  of a thread that is extending at a constant rate  $\dot{\epsilon}$  (dashed line) and of the amplitude  $\alpha$  of disturbances with different initial wave numbers (full curves).

In 1987, Khakhar and Ottino extended the theory for breakup of an infinitely long Newtonian liquid cylinder in a 3D (axisymmetric) elongational flow (Tomotika, 1936; Mikami *et al.*, 1975) to general linear flows. Any linear flow can be decomposed into a 3D elongation and a shear component. They showed that in some circumstances the shear part may be neglected so that the only relevant flow parameter in the analysis is the stretching rate  $\dot{\epsilon}$  ( $= \mathbf{D}:\vec{m}\vec{m}$ ,  $\vec{m}$  being the local orientation of the thread). Upon making the time dimensionless with respect to the shear rate ( $t^* = \dot{\gamma}t$ ) the ratio  $\dot{\epsilon}/\dot{\gamma}$  shows up in the analysis, it is proportional to the efficiency parameter  $e_f$  for line stretch (Ottino, 1989)

$$e_f = \frac{\mathbf{D}:\vec{m}\vec{m}}{\sqrt{\mathbf{D}:\mathbf{D}}} \quad \text{hence} \quad e_f = \frac{\dot{\epsilon}}{\dot{\gamma}/\sqrt{2}}. \quad (4)$$

In steady linear flow,  $\dot{\gamma}$  is constant while  $\dot{\epsilon}$  and hence  $e_f$  may vary due to a transient thread orientation  $\vec{m}$ . In 2D and 3D elongational flows, the orientation of the extending thread is permanently aligned with the principal axis of strain and  $\dot{\epsilon}$  is the classic rate of elongation; the shear rate  $\dot{\gamma}$  equals  $2\dot{\epsilon}$  or  $\sqrt{3}\dot{\epsilon}$  and thus the efficiency  $e_f$  is  $\sqrt{(1/2)}$  or  $\sqrt{(2/3)}$ , respectively. In simple shear flow, the orientation  $\vec{m}$  of the thread rotates towards the direction of the streamlines thus in the steady case  $\dot{\gamma}$  is constant while  $e_f$  and  $\dot{\epsilon}$  decay towards zero.

The main idea of the breakup process at constant stretching rate is illustrated in Fig. 3. Disturbances of all initial wavelengths  $\lambda_{0i}$  (or dimensionless wave numbers  $x_{0i} = 2\pi R/\lambda_{0i}$ ) evolve in time. At the same time, the wavelengths are stretched [ $\lambda = \lambda_0 \exp(\dot{\epsilon}t)$ ] and the thread radius is thinned [ $R = R_0 \exp(-\dot{\epsilon}t/2)$ ]. The growth rate of a small sinusoidal disturbance amplitude  $\alpha$  can be written in terms of material properties ( $\eta_c, p, \sigma$ ), flow conditions ( $e_f, \dot{\epsilon}$ ), and current geometry ( $R, x$ ):

$$\frac{d \ln \alpha}{d t^*} = 2^{-3/2} e_f \left[ \frac{(1-x^2)}{\eta_c \dot{\epsilon} R / \sigma} \Phi(x, p) - 3(p-1) \bar{\Phi}(x, p) - 1 \right], \quad (5)$$

where  $\Phi(x, p)$  and  $\bar{\Phi}(x, p)$  are complicated functions of the wave number and the viscosity ratio (Mikami *et al.*, 1975). It is convenient to use the wave number  $x [ = x_0 \exp(-3\dot{\epsilon}t/2) ]$  as the current variable instead of the time  $[ t = 2/(3\dot{\epsilon}) \ln(x_0/x) ]$  or  $t^* = 2^{3/2}/(3e_f) \ln(x_0/x) ]$ . Upon integration of Eq. (5), the evolution of an initial disturbance  $\alpha_0$  is obtained. In general, the amplitude first damps, then grows, and finally damps again. Since the amplitude cannot damp below the initial "noise" level  $\alpha_0$ , the first damping stage is omitted and, after some delay time,  $\alpha$  starts growing from  $\alpha_0$ . Breakup is achieved if the amplitude of a disturbance intersects with the continuously decreasing mean thread radius. Since a whole range of disturbances, with different wave numbers and initiation times, may lead to breakup, the initial wave number  $x_{0i}$  has to be optimized to find the disturbance that yields the shortest breakup time. The amplitude of this "fatal" disturbance starts growing at a critical time  $t_{crit}^*$ , where the thread radius has decreased to  $R_{crit}$ . After the growth time  $t_g^*$ , the amplitude reaches the actual mean thread radius at the breakup time  $t_b^*$ ; the extending thread breaks leaving a line of drops with radius  $R_{drops}$ .

For constant stretching rate  $\dot{\epsilon}$  (i.e., the external flow is either steady 2D or 3D elongational), the breakup process was analyzed over a broad range of the relevant independent parameters. The results can be condensed further than was done by former authors. The independent variables are reduced to only two relevant dimensionless groups:  $\eta_c \dot{\epsilon} \alpha_0 / \sigma$  and  $p$ , while only three dependent variables are of interest:  $R_{crit} / \alpha_0$ ,  $R_{drops} / \alpha_0$ , and  $e_f t_g^*$ . In fact, all length scales are scaled with respect to the initial amplitude  $\alpha_0$ . Moreover, the main results of the analysis are insensitive of the initial thread radius  $R_0$ : in Fig. 3,  $R_0$  may be anywhere left of  $R_{crit}$ ;  $R_0$  is only relevant for the stage of affine deformation down to  $R_{crit}$  ( $\rightarrow t_{crit}^*$ ). Figures 4, 5, and 6 give the condensed results; all other dependent variables can be derived from these plots. For example, the total breakup time  $t_b^*$  equals

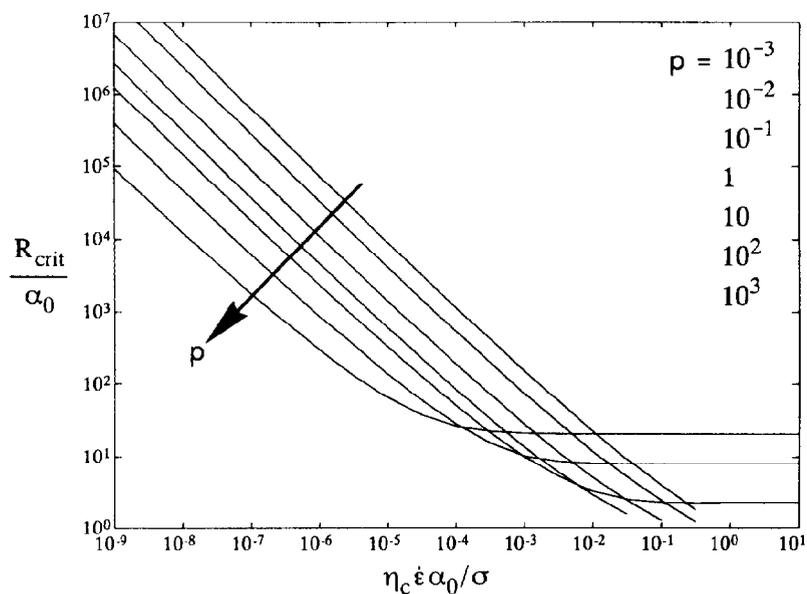
$$t_b^* = t_{crit}^* + t_g^* = \frac{1}{e_f} \left[ 2^{3/2} \ln \left( \frac{R_0}{R_{crit}} \right) + e_f t_g^* \right]. \quad (6)$$

In a flow of constant stretching rate (with known  $\dot{\epsilon}$ ,  $e_f$ , and  $\dot{\gamma}$ ), the breakup time  $t_b$  ( $= t_b^* / \dot{\gamma}$ ) of a thread (with known  $p$ ,  $\eta_c$ ,  $\sigma$ ,  $\alpha_0$ , and  $R_0$ ) can be calculated from Eq. (6), using the theoretical values of  $R_{crit}$  and  $e_f t_g^*$  from Figs. 4 and 6. The resulting dropsizes is obtained from Fig. 5.

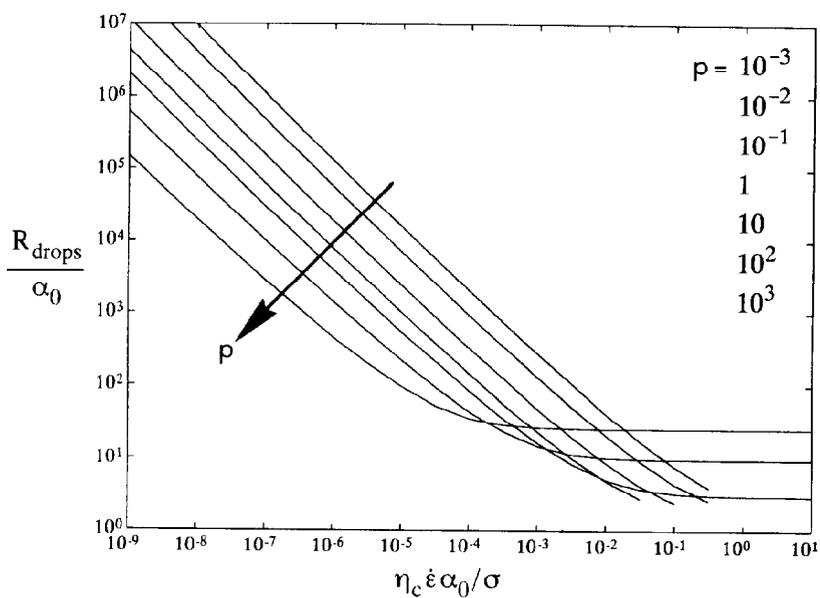
From Figs. 4 and 5 it is evident that upon an increase of the stretching rate the thread is thinned further before breakup occurs producing smaller drops. For low enough values of  $\eta_c \dot{\epsilon} \alpha_0 / \sigma$ , a larger viscosity ratio  $p$  also yields smaller drops because disturbances develop more slowly on a highly viscous thread (thus it has thinned further before breakup occurs). In other situations, the optimum value of  $p$  for obtaining the smallest dropsizes depends on the value of  $\eta_c \dot{\epsilon} \alpha_0 / \sigma$  that is realized in practice. A physical lower limit for the dropsizes arises from the fact that the mean thread radius cannot decrease below  $\alpha_0$ .

The value of  $\alpha_0$ , the initial disturbance, was estimated by Kuhn (1953). Based on thermal fluctuations of the interface, he derived

$$\alpha_0 = \sqrt{\frac{21kT}{8\pi^{3/2}\sigma}}, \quad (7)$$



**FIG. 4.** Dimensionless critical thread radius, indicating the start of growth of the disturbance that leads to breakup of a constantly stretching thread, as a function of the dimensionless stretching rate; parameter is the viscosity ratio.



**FIG. 5.** Dimensionless drop radius resulting from thread breakup during extension at a constant rate  $\dot{\epsilon}$  as a function of the dimensionless stretching rate; parameter is the viscosity ratio.

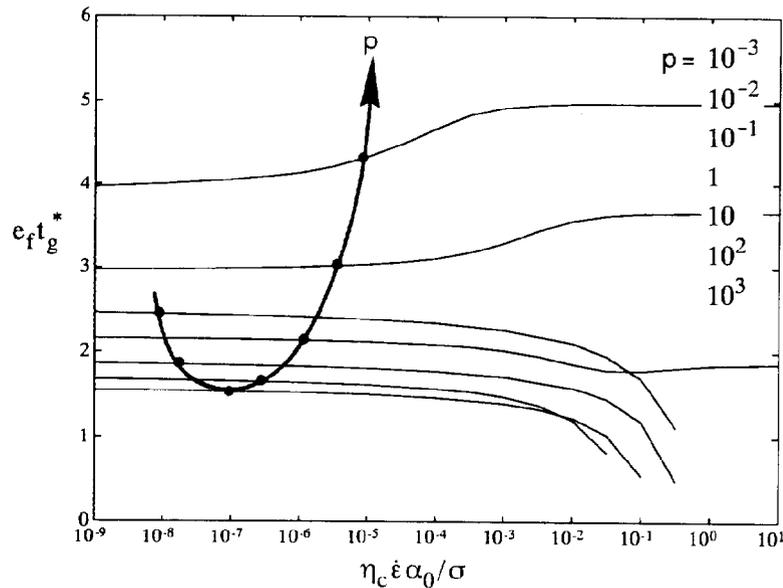


FIG. 6. Flow type independent growth time of the disturbance that leads to breakup of a constantly stretching thread as a function of the dimensionless stretching rate; parameter is the viscosity ratio.

where  $k$  is the Boltzmann constant and  $T$  the absolute temperature. Kuhn's prediction yields values of the order  $10^{-9}$  m. Although Mikami *et al.* (1975) suggest  $10^{-8}$  to  $10^{-7}$  m, Kuhn's estimate appears to be satisfactory for the experiments in the next paragraph. Because  $\alpha_0$  is included in both the horizontal and the vertical axis of Figs. 4 and 5, the value of  $\alpha_0$  is not as important as it might seem: Changing its value shifts  $R_{\text{crit}}$  ( $\eta_c \dot{\epsilon} / \sigma$ ) and  $R_{\text{drops}}$  ( $\eta_c \dot{\epsilon} / \sigma$ ) equally in both horizontal and vertical direction resulting in a shift along the  $-45^\circ$  diagonal (on a logarithmic scale). This is of minor influence on the linearly descending parts of the curves (slope  $-0.9$ ); only the apparent bottoms for large  $p$  change with  $\alpha_0$ . For  $\alpha_0 = 10^{-9}$  m, Tjahjadi and Ottino (1991) approximated calculated  $R_{\text{crit}}$  data by a power-law equation, yielding straight lines in our plot.

In steady simple shear flow, the results are more complicated since  $\dot{\epsilon}$  and hence  $e_f$  decay (as known functions) with time. Khakhar and Ottino (1987) elaborated this case but the results cannot be condensed as in Figs. 4, 5, and 6, since the initial thread radius and orientation appear as extra independent variables. Moreover, in simple shear flow it has to be checked whether the influence of the shear component on the disturbance is negligible.

## EXPERIMENTAL

In contrast to drop breakup under quasiequilibrium conditions (Fig. 1), the breakup of an extending thread has been rarely studied experimentally. Therefore, the results from the above analysis were verified using model liquids in an opposed jets device. This device generates a plane hyperbolic flow and has been described in Janssen *et al.* (1993). Castor oil ( $\eta_d = 0.74$  Pa s) was used as drop phase while the continuous phase consisted of different grades of silicon oil; the interfacial tension equals  $4 \times 10^{-3}$  N/m. A drop of a few mm diameter is subjected instantaneously to a steady

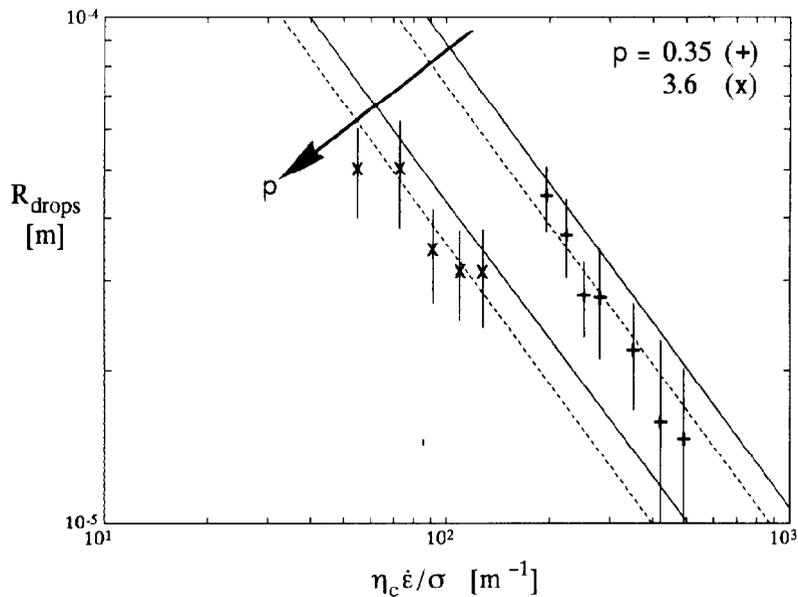


FIG. 7. Experimental (symbols) and calculated (curves) drop sizes as a function of the flow parameter  $\eta_c \dot{\epsilon} / \sigma$  in steady plane hyperbolic flow; in the calculations  $\alpha_0 = 10^{-9}$  m (full curves) or  $10^{-10}$  (dashed curves); parameter is the viscosity ratio.

elongation rate sufficiently high to deform it affinely ( $Ca/Ca_{crit} > 5$ ). Disintegration of the extending thread formed is studied using a microscope and videorecordings. The original drop is extended sufficiently far to consider the thread infinite and neglect end effects.

While Mikami *et al.* (1975) compared various experimental data, except for the final drop size, to calculated values, Fig. 7 compares experimental drop sizes to calculated ones derived from Fig. 5. The scatter in the experimental data is due to nonuniformity of the breakup process and to satellite droplets emerging between the main drops (Tjahjadi *et al.*, 1992). The relevant drop size was averaged over many experiments. The influence of  $\eta_c \dot{\epsilon} / \sigma$  and  $p$  is predicted properly by the theory. The value of  $10^{-9}$  m for  $\alpha_0$  (Kuhn's estimate) gives a reasonable agreement which is remarkable since the theory is based on an extrapolation of a linear (small amplitude) analysis. A better fit is obtained using  $10^{-10}$  m, however, such small disturbances are physically unrealistic.

Apart from the final drop sizes, the experimental breakup times  $t_b$  were also compared to calculated values (Fig. 8). The separate contributions of  $t_{crit}$  and  $t_g$  cannot be measured because it is experimentally impossible to detect the critical point where the fatal disturbance starts growing (see Fig. 3). Therefore, only the total breakup times are compared. The measured breakup time reaches from an initial thread radius  $R_0$  to the point of breakup. The calculated breakup time is obtained from Eq. (6) using the experimental value of  $R_0$  and the theoretical values of  $R_{crit}$  and  $e_f t_g^*$  (from Figs. 4 and 6). So in fact, comparison of measured and calculated breakup times verifies the theoretical plots of  $R_{crit}/\alpha_0$  and  $e_f t_g^*$  (the assumption of affine stretching is certainly valid in the experiments). With  $\alpha_0 = 10^{-9}$  m, the agreement is again satisfactory while a better fit is obtained, here, by increasing  $\alpha_0$  to  $10^{-8}$  m (rather than a decrease

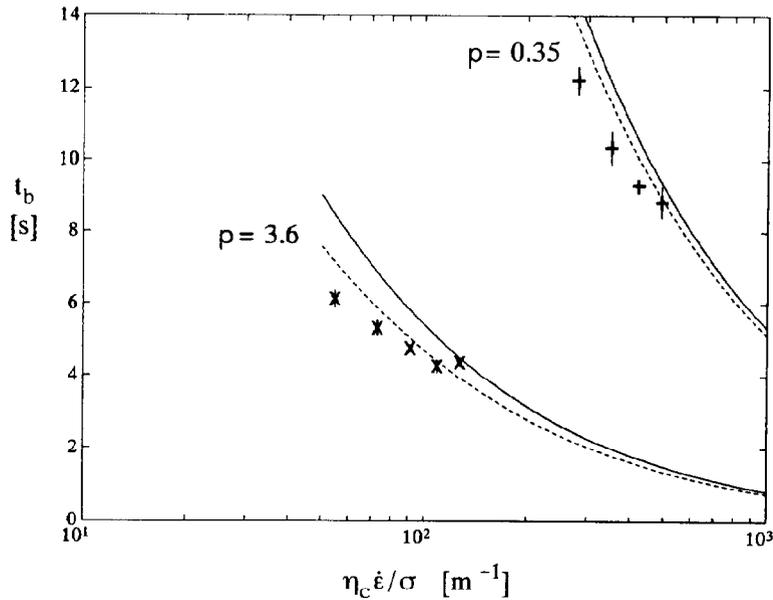


FIG. 8. Experimental (symbols) and calculated (curves) breakup times as a function of the flow parameter  $\eta_c \dot{\epsilon} / \sigma$  in steady plane hyperbolic flow;  $R_0 = 5 \times 10^{-4}$  m; in the calculations  $\alpha_0 = 10^{-9}$  m (full curves) or  $10^{-8}$  (dashed curves); parameter is the viscosity ratio.

of  $\alpha_0$  like in Fig. 7). In the experiments  $t_g$  typically is about 2 s. If  $R_0 \gg R_{crit}$ , as is the case in most practical situations,  $t_g$  becomes negligible with respect to  $t_{crit}$  and the total breakup time can be approximated by affine deformation only [the first term of Eq. (6) with  $R_{crit}$  taken from Fig. 4].

### COMPARISON OF DISPERSION MECHANISMS

Now that the breakup of an extending liquid thread has been reviewed and confirmed experimentally, it is possible to compare the dropsizes resulting from the two dispersion mechanisms of Fig. 2. For plane hyperbolic flow this is illustrated in Fig. 9. The full curves are deduced from Fig. 5: the transient process with constant stretching rate; and the dashed lines from Fig. 1: the stepwise equilibrium mechanism. Generally, the transient mechanism yields smaller drops and it is evident that  $p = 1$  is not the optimum viscosity ratio. Rather than that, a higher viscosity ratio yields a finer dispersion, as mentioned by Tjahjadi and Ottino (1991). If drops are formed by the transient mechanism, while the equilibrium mechanism would result in smaller droplets (typically at high values of  $\eta_c \dot{\epsilon} / \sigma$ ), they just break once more.

In a practical situation, a point to consider is whether the time available for each of the idealized processes to occur is sufficient. For the transient mechanism the breakup time is given by Eq. (6). For the stepwise equilibrium route the breakup time is generally longer. The time for one breakup step at  $Ca_{crit}$  was measured experimentally by Grace (1971). (In fact, Grace's experiments must have been performed slightly above  $Ca_{crit}$ , since exactly at  $Ca_{crit}$  the breakup time is infinite.)

Note that the dashed lines in Fig. 9 continue down to unrealistic small length scales. Of course, a lower limit exists due to the length scale below which continuum me-

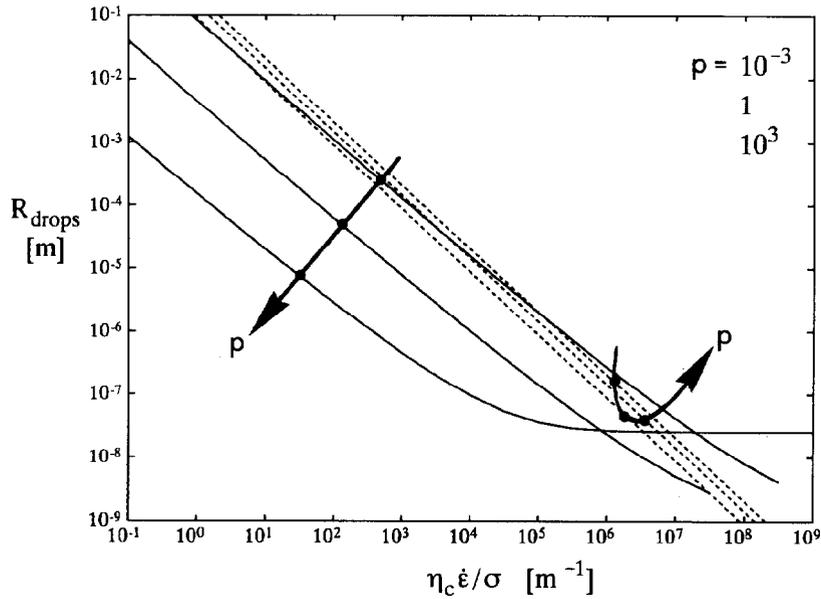


FIG. 9. Minimum attainable dropsize in plane hyperbolic flow resulting from (i) the stepwise equilibrium mechanism of drop breakup at  $Ca_{crit}$  (dashed lines) and (ii) the transient mechanism of thread breakup during extension with  $\alpha_0 = 10^{-9}$  m (full curves); parameter is the viscosity ratio.

chanics cannot be applied (order of magnitude  $10^{-9}$  m). To deduce the full curves,  $\alpha_0$  was chosen to be  $10^{-9}$  m. As explained before, changing the arbitrary value  $\alpha_0$  is of minor influence on the dropsize. The value of the flow parameter  $\eta_c \dot{\epsilon} / \sigma$  typically ranges from 1 to  $10^5$   $m^{-1}$  for model liquids in an experimental device (e.g., opposed jets or Couette) and from  $10^4$  to  $10^8$   $m^{-1}$  for polymer melts in the typical shear rate range of processing equipment.

For simple shear flow, the dropsize resulting from the stepwise equilibrium mechanism is, again, obtained from Fig. 1. Dropsizes resulting from transient thread breakup in steady simple shear were deduced from Fig. 8 of Khakhar and Ottino (1987); both the initial radius and orientation of the thread have to be chosen. In Fig. 10 the comparison is presented for two initial radii and viscosity ratios, with  $\alpha_0 = 10^{-9}$  m and the initial angle between thread axis and streamlines  $\varphi_0 = 27^\circ$  (corresponding to  $[c = 2 \tan \varphi_0] = 1$ , an upper bound). Upon decreasing  $\varphi_0$  to  $3^\circ$  ( $c = 0.1$ , a lower bound) the full curves shift 1 decade to the right. Note that along the horizontal axis the (constant) shear rate  $\dot{\gamma}$  is used rather than the time-dependent stretching rate  $\dot{\epsilon}$ ; for comparison with plane hyperbolic flow,  $\dot{\epsilon}$  in Fig. 9 should be substituted by  $\dot{\gamma}/2$ .

The dropsize resulting from the transient mechanism is, in simple shear flow, proportional to  $\dot{\gamma}^{-0.3}$  (instead of  $\dot{\gamma}^{-0.9}$  in steady elongation) and is larger than in an elongational flow of equal shear rate. Again, the transient mechanism may yield smaller drops than the stepwise equilibrium route, but the difference is much less pronounced than in elongational flow. The influence of the viscosity ratio is extreme in the equilibrium process (see also Fig 1): at  $p > 4$  the equilibrium mechanism is not able to break drops since  $Ca_{crit}$  goes to infinity. For the transient mechanism an increasing  $p$  keeps reducing the dropsize. Beyond  $p = 4$ , the stretching of the original

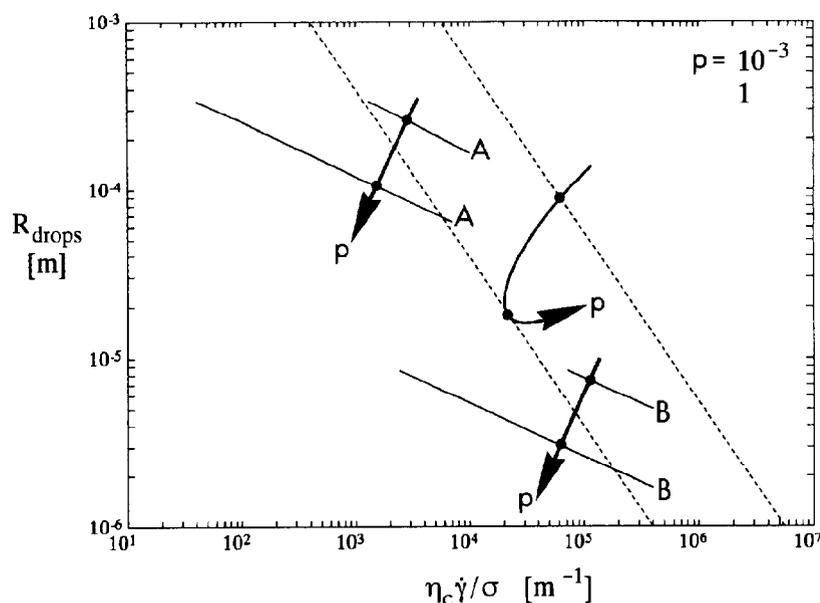


FIG. 10. Minimum attainable dropsize in simple shear flow resulting from (i) the stepwise equilibrium mechanism of drop breakup at  $Ca_{crit}$  (dashed lines) and (ii) the transient mechanism of thread breakup during extension with  $\alpha_0 = 10^{-9}$  m,  $\varphi = 27^\circ$ , and  $R_0 = 2 \times 10^{-4}$  m (A) or  $5 \times 10^{-6}$  m (B) (full curves); parameter is the viscosity ratio.

drop into a thread is impossible, however, it is sufficient that it takes place only once, e.g., in a local region of elongational flow.

In simple shear flow, reorientation of the thread out of the streamline direction temporarily increases the local stretching rate  $\dot{\epsilon}$  favoring the suppression of disturbances and reducing the dropsize. Hence, periodic reorientation (via stretching and folding; see, e.g., Ottino, 1989) improves not only the distributive mixing, but also the dispersive mixing (except in the nonstretching regions of the folds).

Practical flow situations bring arbitrary histories of both shear and stretching rate. The analysis of thread breakup during extension should accordingly be performed using a locally given function  $\dot{\epsilon}(t)$ ; results of general validity are hard to obtain. If, however,  $\dot{\epsilon}$  is fairly constant within the growth time of a disturbance ( $t_g$ ), the presented results are valid momentarily. A number of complications arise due to the presence of many drops and threads. As found in a number of our experiments, collision of an extending thread with a drop or another thread locally imposes a large disturbance amplitude resulting in a scission, followed by endpinching (Stone and Leal, 1989). The drop sizes from Fig. 5, actually, represent a lower limit. Also, coalescence of the drops formed (see, e.g., Chesters, 1991) coarsens blend morphologies and has to be taken into account in order to simulate the development of a morphology in a practical blending situation. This is the subject of the present research in our laboratory.

## CONCLUSION

For Newtonian liquids it has been quantitatively shown that the more realistic transient mechanism of thread breakup during extension generally results in smaller

drops than the stepwise equilibrium mechanism of repeated drop breakup at  $Ca_{crit}$ . The transient mechanism predicts that a viscosity ratio of unity is not optimum for obtaining the finest dispersion. Rather than that, a larger viscosity ratio (thread/matrix) yields a finer dispersion.

For constant stretching rate, the results of the classic analysis of thread breakup during extension are independent of the initial radius and the initial disturbance amplitude. Hence, it is possible to condense them in a few graphs. The analysis has been verified experimentally and the agreement is satisfactory.

In fact, the two dispersion mechanisms could already have been compared in the 30's (Taylor, 1934; Tomotika, 1936). Only the complexity of the transient analysis and the lack of computational facilities, apparently, delayed the complete interpretation of this work.

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