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Deformation and breakup of slender drops in linear flows

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We study the deformation and breakup of a low-viscosity slender drop in a linear flow, $\bar{v}^{\infty} = \bar{\boldsymbol{L}}\cdot\bar{x}$, assuming that the drop remains axisymmetric. We find that the drop stretches as if it were immersed in an axisymmetric extensional flow with a strength $\bar{\boldsymbol{D}}:\bar{m}\bar{m}$, where $\bar{\boldsymbol{D}} = \frac{1}{2}(\bar{\boldsymbol{L}}+\bar{\boldsymbol{L}}^{\mathrm{T}})$, and $\bar{m}$ is the orientation of the drop, and rotates as if it were a material element in a hypothetical flow $\bar{\boldsymbol{M}} = G\bar{\boldsymbol{D}}+\bar{\boldsymbol{\Omega}}$, where $\bar{\boldsymbol{\Omega}} = \frac{1}{2}(\bar{\boldsymbol{L}}^{\mathrm{T}}-\bar{\boldsymbol{L}})$, and G is a known function of the drop length. The approximations involved in the model are quite good when $\bar{\boldsymbol{M}}$ has only one eigenvalue with a positive real part, and somewhat less precise when $\bar{\boldsymbol{M}}$ has two eigenvalues with positive real parts. In the suitable limits the model reduces to Buckmaster's (1973) model for axisymmetric extensional flow and to the linear-axis version of the more general model proposed by Hinch & Acrivos (1980) for simple shear flow. In establishing a criterion for breakup for all linear flows, we find that the relevant quantity that specifies the flow is the largest positive real part of the eigenvalues of $\bar{\boldsymbol{M}}$, which depends on the drop length and the imposed flow. Our predictions are in reasonable agreement with the recent experimental data of Bentley (1985) for general two-dimensional linear flows and those of Grace (1971) for simple shear and hyperbolic extensional flow. We also present calculations for a class of three-dimensional flows as an illustration of the behaviour of three-dimensional flows in general.

1. Introduction

The study of the deformation and breakup of drops is of both practical and theoretical significance. Its origins can be traced back to the work of G. I. Taylor (1934), whose objective was an analysis of the formation of emulsions in definable flow fields. Much theoretical and experimental work followed since, and an excellent review was given recently by Rallison (1984). An assumption in most of the analyses to date has been that the undisturbed flow around the drop is linear, the most studied being purely extensional and simple shear flows. Though the flow fields encountered in practical situations are complex, an understanding of the deformation and breakup of drops in linear, unsteady flows seems to be at the very core of a theoretical description of the mixing of immiscible fluids. To a first approximation, the velocity field with respect to a moving drop and far from it is

$$\bar{v}^{\infty} = (\nabla\bar{v})\cdot\bar{x},$$

provided that the drop is much smaller than the lengthscale over which the linear approximation holds (Rallison 1984).

In most cases the velocity field in the mixer is unknown; however, studies have revealed some predominant features of complex flows encountered in practice. In the context of their ability to stretch material elements there seem to be three kinds of flow: flows without reorientation, flows with weak reorientation, and flows with strong reorientation. Prototypes of flows without reorientation are all steady curvilineal flows (Chella & Ottino 1985*a*), of a flow with weak reorientation, a rectangular cavity flow (Chella & Ottino 1985*b*), and flows with strong reorientation, all chaotic flows (Aref 1984; Khakhar, Chella & Ottino 1984). Such considerations would be useful for obtaining a qualitative description of drop deformation and breakup in complex flows.

That being our motivation, we study the stretching and rotation of a slender drop of relatively low viscosity μ_i suspended in a fluid of viscosity μ. The flow far from the drop is

$$\bar{v}^{\infty} = \bar{\boldsymbol{L}}\cdot\bar{x}$$

and the Reynolds number with respect to the drop dimension is vanishingly small. In general, $\boldsymbol{L}$ may be a function of time; however, here we consider only slowly varying flows so that time-dependent effects are negligible.

It is well known from experimental studies (Taylor 1934; Torza, Cox & Mason 1972; Grace 1971; Bentley 1985) that when the viscosity ratio is low ($p = \mu_i/\mu \ll 1$) drops deform into slender pointed shapes prior to breakup. A number of researchers have carried out theoretical analyses of this problem for particular flows. Taylor (1964), Buckmaster (1972, 1973), and Acrivos & Lo (1978) studied a symmetrically placed drop in an axisymmetric extensional flow; Hinch & Acrivos (1979) studied the effect of a non-circular cross-section on a drop oriented along the direction of maximum stretching in a two-dimensional extensional flow, and more recently Hinch & Acrivos (1980) analysed the effects of vorticity on a drop oriented almost along the streamlines in a simple shear flow. Inertial effects were studied by Brady & Acrivos (1982) and found to contribute negligibly to the results. In our analysis we generalize some of the previous work to obtain a model that describes the rotation and stretching of an arbitrarily placed drop in a linear flow. The analysis, of course, does not apply to flows that are too 'weak' to deform the drop substantially, so that the possibility of obtaining pointed drops is excluded. Such a case might be encountered when the eigenvalues of ∇v are purely imaginary.

As a simplification we assume the drop to be axisymmetric with a linear axis. The assumption of a circular cross-section for the drop was first used by Hinch & Acrivos (1980) in the analysis of a drop in a simple shear flow. The assumption was justified by the authors by the results of the analysis of a drop in a hyperbolic extensional flow (Hinch & Acrivos 1979), in which the deviation in the critical strain rate was 2% as compared with the value predicted by an analysis assuming a circular cross-section. Their arguments would be valid for all linear flows with a single direction of stretching. In the case of flows with two directions of stretching, which is possible for three-dimensional flows, the deviation from the axisymmetric case would be higher. An extension of the analysis of Hinch & Acrivos (1979) to the case of biaxial extensional flow indicates that the deviation in the length of an inviscid drop is about 18% from the case with a circular cross-section (Khakhar 1986).

It is also well known from experiments on drop deformation in simple shear flow (Torza *et al.* 1972; Grace 1971) that the drop axis bends owing to the flow. In our analysis this bending is manifested as a variation in the rate of rotation along the drop axis. However, the maximum deviation δ_{max} of the drop axis from a line tangent to the axis at the centre of the drop is small. The prediction from the theoretical analysis of Hinch & Acrivos (1980) for a drop at steady state in a simple shear flow, in which the deviation is a maximum among linear flows, is

$$\delta_{max} = 0.72R_0,$$

where R_0 is the radius of the drop at the centre of the drop. The deviation is expected to be even smaller in the case of flows that are more extensional than the simple shear flow. The conjecture is supported by the experiments of Bentley (1985) for drops in mixed flows generated by a four-roller apparatus. Also, one of the limit cases presented by Hinch & Acrivos (1980) for simple shear flow (henceforth referred to as the Linear-Axis (LA) model), which is based on identical assumptions as our analysis, adequately describes deformation and breakup of drops when the rate of rotation is chosen as that at the centre of the drop (though the physics of breakup is somewhat different when the drop axis is allowed to bend). In our analysis we describe the orientation of the drop by a vector tangent to the drop axis at the centre of the drop and neglect any bending of the drop axis that might occur.

The first part of our analysis closely follows that of Hinch & Acrivos (1980) for simple shear flow, the distinguishing feature of the analysis being that it is carried out with respect to a frame that is fixed on the drop axis and rotates with it. The problem is then reduced to the analysis of a stationary drop in a time-varying flow that depends on the rate of rotation of the drop. The resulting model reduces in the suitable limits to the LA model of Hinch & Acrivos (1980) for simple shear flow and that of Buckmaster (1973) for an axisymmetric extensional flow. The conditions for breakup are found by a linear stability analysis of the equation describing the drop length.

The main contribution of this work is to relate the breakup of slender axisymmetric drops to the kinematics of linear three-dimensional flows, leading to flow classification. The basic objective of flow classification is to define parameters that are appropriate functions of the invariants of the flow which reflect its predominant characteristics and provide a basis for comparing flows. Olbricht, Rallison & Leal (1982) were the first to take into account the interaction between a suspended microstructure and the flow in a flow-classification scheme, using an empirical equation to describe the dynamics of the microstructure. The present analysis reveals a simple means of taking into consideration the modification of the flow by the drop and thus allows the systematic study of breakup in any steady linear flow. The predictions of the theory for breakup in two-dimensional flows agree well with the data of Bentley (1985) obtained in a four-roller apparatus for mixed flows and those of Grace (1971) for simple shear and hyperbolic extensional flow. We also present results for a class of three-dimensional flows to illustrate the behaviour that might be possible for three-dimensional flows in general.

2. Flow around a slender axisymmetric drop

We outline here the analysis of the velocity field around a slender pointed drop that remains axisymmetric, and a result obtain the deformation and rotation of the drop. Further details may be found in Khakhar (1986).

The undisturbed flow far from the drop with respect to a frame that is fixed on the drop axis and rotates with it is given by

$$\boldsymbol{v}^{\infty} = (\dot{\boldsymbol{Q}}\cdot\boldsymbol{Q}^{\mathrm{T}} + \boldsymbol{Q}\cdot\boldsymbol{L}\cdot\boldsymbol{Q}^{\mathrm{T}})\cdot\boldsymbol{x}.$$

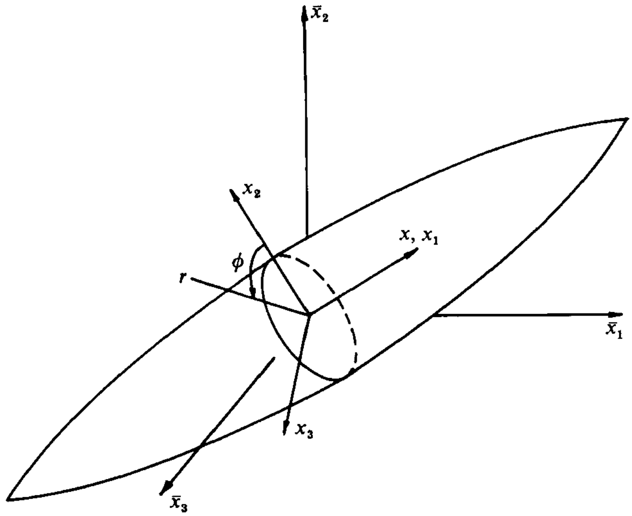


FIGURE 1. Cylindrical coordinate system (r, ϕ, x) fixed on the axis of the slender drop. (x_1, x_2, x_3) and $(\bar{x}_1, \bar{x}_2, \bar{x}_3)$ are the coordinate axes of the rotating and fixed frames respectively.

(the overbar refers to quantities with respect to the fixed frame.) $\boldsymbol{Q} = \boldsymbol{Q}(t)$ is an orthogonal matrix and relates a vector in the two frames by $\boldsymbol{x} = \boldsymbol{Q}\cdot\bar{\boldsymbol{x}}$, and is to be found from the analysis. Based on our assumption of a circular cross-section, we neglect terms in the velocity field containing second-order harmonics of the azimuthal angle (ϕ, see figure 1), which would deform the cross-section and are inconsistent with our assumption. The velocity field then reduces to

$$v_x^\infty = \alpha_1 x + (a_{12}\cos\phi + a_{13}\sin\phi)\, r,$$

$$v_r^\infty = -\tfrac{1}{2}\alpha_1 r + (a_{21}\cos\phi + a_{31}\sin\phi)\, x,$$

$$v_\phi^\infty = (a_{31}\cos\phi - a_{21}\sin\phi)\, x + \tfrac{1}{2}(a_{32} - a_{23})\, r,$$

where

$$a_{ij} = \dot{Q}_{ip} Q_{jp} + (1-\delta_{ij})\, Q_{ip} \bar{L}_{pq} Q_{jq}$$

and

$$\alpha_i = \boldsymbol{D}:\boldsymbol{e}_i\boldsymbol{e}_i \quad \text{(no sum on } i\text{)}.$$

The vector $\boldsymbol{e}_i$ is the unit vector in coordinate direction i, and $\boldsymbol{D}$ is the stretching tensor with respect to the moving frame. In this simplified form the imposed velocity field can thus be seen to be composed of an axisymmetric extensional flow of strength α_1, which depends on the instantaneous orientation of the drop, and a shear flow that is non-axisymmetric.

We construct the disturbance flow generated outside the drop due to the presence of the drop by distributions of sources, source doublets and stresslets arranged along the axis of the drop. Owing to the linearity of the creeping flow equations the external flow is then the sum of the imposed flow and the disturbance flow. The velocity field generated by a line distribution of sources is

$$\boldsymbol{v} = \frac{1}{2}\int_{-1}^{1} \frac{g(s)\,\boldsymbol{x}}{|\boldsymbol{x}|^3}\,\mathrm{d}s,$$

with the constraint

$$\int_{-1}^{1} g(s)\,\mathrm{d}s = 0,$$

and that generated by the source doublets is

$$\boldsymbol{v} = \frac{1}{2}\int_{-1}^{1}\left(\frac{\boldsymbol{h}(s)}{|\boldsymbol{x}|^3} - 3\frac{(\boldsymbol{h}(s).\boldsymbol{x})\,\boldsymbol{x}}{|\boldsymbol{x}|^5}\right)\mathrm{d}s,$$

where $\boldsymbol{x} = (x_1 - s)\,\boldsymbol{e}_1 + x_2\,\boldsymbol{e}_2 + x_3\,\boldsymbol{e}_3$. Distributions of sources and source doublets generate no pressure disturbance. The velocity field due to the stresslets is

$$\boldsymbol{v} = \frac{3}{2}\int_{-1}^{1}\frac{(\boldsymbol{S}(s):\boldsymbol{xx})\,\boldsymbol{x}}{|\boldsymbol{x}|^5}\mathrm{d}s$$

and the corresponding pressure disturbance is

$$P = 3\mu\int_{-1}^{1}\frac{\boldsymbol{S}(s):\boldsymbol{xx}}{|\boldsymbol{x}|^5}\mathrm{d}s.$$

From the form of the imposed velocity field we determine that we require only the components h_2, h_3, S_{12}, S_{13} of $\boldsymbol{h}$ and $\boldsymbol{S}$. An asymptotic evaluation of the above integrals near the drop surface in the limit of slender drops ($R \rightarrow 0$) gives the disturbance velocity as

$$v_x^{\mathrm{d}} \approx S_{12}\frac{\cos\phi}{r} + S_{13}\frac{\sin\phi}{r},$$

$$v_r^{\mathrm{d}} \approx \frac{h_2\cos\phi + h_3\sin\phi}{r^2} - S_{12}'\cos\phi - S_{13}'\sin\phi + \frac{g}{r},$$

$$v_\phi^{\mathrm{d}} \approx -\frac{h_2\sin\phi - h_3\cos\phi}{r^2},$$

where the prime denotes differentiation with respect to x, and $R = R(x,t)$ specifies the drop surface. The associated stress field due to the disturbance is

$$T_{xx}^{\mathrm{d}} \approx \frac{3\mu}{r}(S_{12}'\cos\phi + S_{13}'\sin\phi).$$

$$T_{rx}^{\mathrm{d}} \approx -\frac{\mu}{r^2}(S_{12}\cos\phi + S_{13}\sin\phi),$$

$$T_{rr}^{\mathrm{d}} \approx \frac{2\mu}{r}(S_{12}'\cos\phi + S_{13}'\sin\phi) - \frac{4\mu}{r^3}(h_2\cos\phi + h_3\sin\phi), \\ -\frac{2\mu g}{r^2},$$

where μ is the external viscosity.

The internal flow is dominated by the pressure gradient along the axis of the drop due to the slenderness of the drop and the low internal viscosity, and following the arguments of Hinch & Acrivos (1979, 1980) the axial velocity inside the drop is approximately given by

$$v_x^{\mathrm{i}} \approx -\frac{(R^2 - r^2)(P^{\mathrm{i}})'}{4\mu_{\mathrm{i}}} + v_x^{\mathrm{e}}(R),$$

where $\boldsymbol{v}^{\mathrm{e}} = \boldsymbol{v}^{\infty} + \boldsymbol{v}^{\mathrm{d}}$. The internal pressure is then easily found to be

$$P^{\mathrm{i}} = P_0^{\mathrm{i}}(t) + 8\mu_{\mathrm{i}}\int_0^x\left(\frac{\alpha_1 x}{R^2} + \frac{2}{R^4}\int_0^x R\dot{R}\,\mathrm{d}x\right)\mathrm{d}x,$$

where $P_0^{\mathrm{i}}(t)$ is the pressure at the centre of the drop.

We now determine the unknown functions (g, h_1, h_2, S_{12}, S_{13}, R) by imposing the boundary conditions on the surface of the drop. Neglecting the curvature in the axial direction, the normal-stress balance on the surface of the drop is given by

$$(\boldsymbol{T}^{\mathrm{e}}-\boldsymbol{T}^{\mathrm{i}}):\boldsymbol{nn} \approx \sigma/R,$$

where $\boldsymbol{n}$ is the outward unit normal and σ is the surface tension. On substituting the expressions for internal and external stresses we obtain

$$\frac{2\mu}{R}(S_{12}' \cos\phi + S_{13}' \sin\phi) - \frac{4\mu}{R^3}(h_2 \cos\phi + h_3 \sin\phi) - \mu\alpha_1 + \frac{2\mu R'}{R^2}(S_{12}\cos\phi + S_{13}\sin\phi)$$
$$-2\mu R'(a_{12}+a_{21})\cos\phi + 2\mu R'(a_{13}+a_{31})\sin\phi = \frac{\sigma}{R} - P_{\mathrm{i}},$$

where the internal viscous stresses are assumed to be negligibly small compared with the pressure P_{i}. The normal-stress boundary condition is satisfied if the strengths of the distributions of the singular solutions are

$$S_{12} = (a_{12}+a_{21})R^2, \quad S_{13} = (a_{13}+a_{31})R^2$$

defining the strength of the stresslets,

$$h_2 = (a_{12}+a_{21})R'R^3, \quad h_3 = (a_{13}+a_{31})R'R^3$$

the strength of the source doublets, and

$$g = \frac{P_{\mathrm{i}}R^2}{2\mu} - \tfrac{1}{2}\alpha_1 R^2 - \frac{\sigma R}{2\mu}$$

the strength of the sources. The shear stress on the surface of the drop is vanishingly small, and the shear-stress boundary conditions are also approximately satisfied by the above equations.

The kinematic condition on the surface of the drop describing the change of radius with time is given by

$$\dot{R} = v_{\mathrm{r}}^{\mathrm{e}}(R) - R' v_x^{\mathrm{e}}(R),$$

and, on substituting for the velocity, we find

$$\dot{R} = (a_{21}\cos\phi + \alpha_{31}\sin\phi)\,x - 2RR'(a_{12}+a_{21})\cos\phi$$
$$-2RR'(a_{13}+a_{31})\sin\phi - RR'(a_{12}\cos\phi + a_{13}\sin\phi)$$
$$+\frac{P_{\mathrm{i}}R}{2\mu} - \alpha_1 R - \frac{\sigma}{2\mu} - \alpha_1 R'x,$$

where we have used the expressions found earlier for the strengths of distributions. The above condition holds if the following relations are satisfied:

$$\dot{R} = -\alpha_1(R+R'x) + \frac{P_{\mathrm{i}}R}{2\mu} - \frac{\sigma}{2\mu},$$
$$a_{21}\left(1-\frac{2RR'}{x}\right) = \frac{3RR'}{x}a_{12},$$
$$a_{31}\left(1-\frac{2RR'}{x}\right) = \frac{3RR'}{x}a_{13}.$$

The first equation describes the change of the drop shape with time, while the latter two describe completely the rotation of the drop since one degree of freedom, of the original three, is lost owing to the axisymmetry of the drop.

The equation for the evolution of the drop surface is identical in form with the one derived for a slender drop in an axisymmetric flow by Buckmaster (1973) and admits the following stable solution as shown by Acrivos & Lo (1976):

$$R = R_0(1-x^2/l^2).$$

The evolution of the length of the drop is given by

$$\frac{\dot{l}}{l} = \alpha_1 - \frac{\sigma}{2\sqrt{5}\,\mu a}\frac{(l/a)^{\frac{1}{2}}}{1+0.8p(l/a)^3}.$$

The complications of the matrices $\boldsymbol{Q}$ and $\dot{\boldsymbol{Q}}$ are avoided by describing the rotation of the drop by the unit vector $\bar{\boldsymbol{m}}$ oriented along the drop axis. On substituting $\bar{\boldsymbol{m}} = \boldsymbol{Q}^{\mathrm{T}}\cdot\boldsymbol{e}_1$ in the equations for rotation found above and simplifying, we obtain

$$\dot{\boldsymbol{m}} = (G\bar{\boldsymbol{D}}+\bar{\boldsymbol{\Omega}})\cdot\bar{\boldsymbol{m}} - (G\bar{\boldsymbol{D}}:\bar{\boldsymbol{m}}\bar{\boldsymbol{m}})\,\bar{\boldsymbol{m}}$$

where $G = (1-5RR'/x)/(1+RR'/x)$, and $\bar{\boldsymbol{D}}$ and $\bar{\boldsymbol{\Omega}}$ are the symmetric and antisymmetric parts of the velocity gradient tensor. Since G is a function of x, the rate of rotation depends on the axial distance along the drop, which physically can be interpreted as a tendency for the drop axis to bend. Based on our earlier justification, we calculate G at the centre of the drop to be

$$G = \frac{1+12.5a^3/l^3}{1-2.5a^3/l^3}.$$

In the case of a simple shear flow, and when the drop axis is almost aligned with the streamlines, the above equations reduce to those of Hinch & Acrivos (1980) for the LA model, and for a drop oriented along the direction of stretching in an axisymmetric extensional flow to that of Buckmaster (1973).

If we define a hypothetical flow with a velocity gradient tensor given by

$$\nabla\bar{\boldsymbol{v}}^* = G\bar{\boldsymbol{D}}+\bar{\boldsymbol{\Omega}}$$

the equation for rotation can be written as

$$\dot{\bar{\boldsymbol{m}}} = (\nabla\bar{\boldsymbol{v}}^*)\cdot\bar{\boldsymbol{m}} - (\bar{\boldsymbol{D}}^*:\bar{\boldsymbol{m}}\bar{\boldsymbol{m}})\,\boldsymbol{m},$$

which is the equation for the rotation for a material element. Since G is slightly greater than 1, the drop rotates as a material element in a flow that appears to be slightly more extensional in character than the imposed flow. The internal circulation within the drop seems to 'take up' some of the vorticity of the imposed flow, making the flow seem more extensional in character.

3. Non-dimensionalization of the model equations

The characteristic length and time chosen for rendering the equations dimensionless are $a/p^{\frac{1}{3}}$ and $1/\dot{\gamma}$ respectively, where $\dot{\gamma} = (\bar{\boldsymbol{D}}:\bar{\boldsymbol{D}})^{\frac{1}{2}}$. Based on the quantities defined above, the model equations reduce to

$$\frac{\dot{l}}{l} = e - \frac{1}{2\sqrt{5}\,E}\frac{l^{\frac{1}{2}}}{1+0.8\,l^3},$$

$$\dot{\bar{\boldsymbol{m}}} = G(\bar{\boldsymbol{M}}\cdot\bar{\boldsymbol{m}} - e\bar{\boldsymbol{m}}),$$

where $G = (1+12.5p/l^3)/(1-2.5p/l^3)$, $E = (\dot{\gamma}\mu a/\sigma)p^{\frac{1}{6}}$ is the dimensionless strain rate, $e = \bar{\boldsymbol{D}}:\bar{\boldsymbol{m}}\bar{\boldsymbol{m}}/\dot{\gamma}$ is the stretching efficiency (Chella & Ottino 1985*a*), and

$$\bar{\boldsymbol{M}} = (G\bar{\boldsymbol{D}}+\bar{\boldsymbol{\Omega}})/G\dot{\gamma}$$

is the normalized velocity gradient tensor, which defines the hypothetical flow according to which the drop rotates. The characteristic time is chosen so that the results obtained for the breakup of drops are frame-indifferent. The maximum efficiency corresponds to a material element oriented along the direction of stretching in an axisymmetric extensional flow ($e = \sqrt{\frac{2}{3}}$), and this dimensionless form clearly shows the upper bound for the rate of stretching of a drop in any linear flow given by

$$\dot{l}/l \leqq \sqrt{\tfrac{2}{3}}.$$

We also note that in the limit of very long drops ($l \rightarrow \infty$) the parameter G goes to 1 and the resistance to stretching becomes negligible so that the drop stretches and rotates as a material element in the flow. Next we study the breakup of drops in linear flows as predicted by the above equations.

4. Breakup in steady linear flows

In the context of the model, a drop is said to break when it undergoes unbounded extension, and this is a consequence of the surface-tension forces being unable to balance the viscous stresses due to the imposed flow. The drop might actually break by mechanisms that are beyond the scope of this model. Here we derive criteria for the breakup of drops in a linear flow which is constant with time.

The dynamics of stretching of the drop are qualitatively illustrated in figure 2, where we have plotted the resistance to stretching offered by the drop versus the drop length for different strain rates E. The dashed line shows the asymptotic value of the efficiency. For strain rates smaller than the critical strain rate E_c there are two steady states, one stable (l_s) and one unstable (l_s^*). For strain rates higher than the critical strain rate there are no steady states, and the drop extends indefinitely. At the critical strain rate the steady state (l_{sc}) is incipiently stable and the two lines are tangent to one another.

Olbricht *et al.* (1982) studied the equation of rotation for the case of G being an arbitrary constant. In our case G is a function of the length of the drop, and the equations for stretch and rotation are coupled. At steady state, however, the following relations hold:

$$e_\infty = \frac{1}{2\sqrt{5}\,E}\,\frac{l_s^{\frac{1}{2}}}{1+0.8\,l_s^3},$$

$$\bar{\boldsymbol{M}}\cdot\bar{\boldsymbol{m}}_i = \lambda_i\,\bar{\boldsymbol{m}}_i, \quad e_\infty = \max_i \mathrm{Re}(\lambda_i),$$

where the subscript s denotes the steady state quantities. On physical grounds, a steady-state length exists only if the asymptotic efficiency e_∞ is positive, where e_∞ is the largest positive real part of the eigenvalues of $\bar{\boldsymbol{M}}$. However, a steady-state orientation for the drop exists only if the eigenvalue containing the largest positive part is purely real. If the eigenvalue is complex a steady-state length exists, but the drop rotates in a plane orthogonal to the direction of maximum compression.

We now study the stability of the length of the drop by linearizing the equation for stretching of the drop about the steady-state length l_s. The stability of the

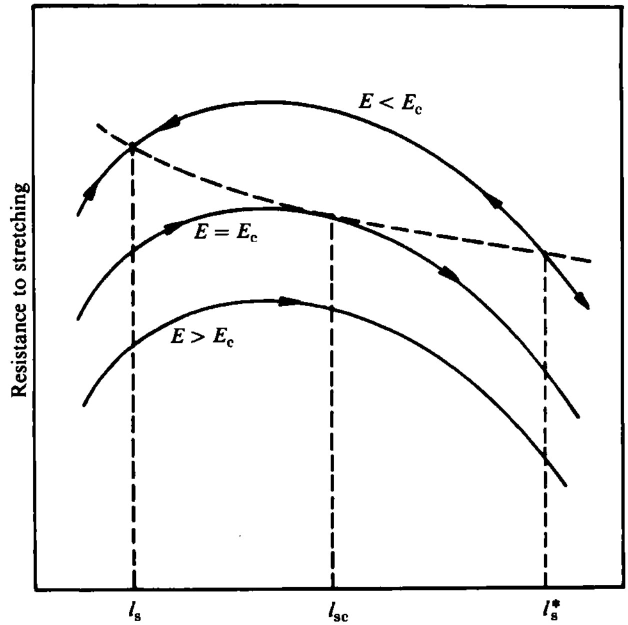


FIGURE 2. Qualitative dynamics of stretching of a slender axisymmetric drop: ——, resistance to stretching versus the drop length l for different strain rates E; ---, asymptotic imposed strain.

orientation is assumed to be unaffected by small changes in the length due to the weak coupling between the equations. Hence

$$\dot{\Delta}/\Delta = l_s f'(l_s),$$

where $\Delta = l - l_s$ and $\dot{l} = lf(l)$. At incipient stability $f'(l_{sc}) = 0$, where l_{sc} is the critical drop length, since $f'(l_s) > 0$ implies instability and $f'(l_s) < 0$ implies stability. Given p and $\boldsymbol{L}$, we can calculate the largest drop (l_s) that can survive in the flow from the condition for incipient stability as

$$\left.\frac{\partial e_\infty}{\partial l_s}\right|_{l\mathrm{sc}} = \frac{1-4l_{\mathrm{sc}}^3}{2l_{\mathrm{sc}}(1+0.8l_{\mathrm{sc}}^3)},$$

and subsequently the critical strain rate from

$$E_c = \frac{l_{\mathrm{sc}}^{\frac{1}{2}}}{2\sqrt{5}\, e_\infty(1+0.8l_{\mathrm{sc}}^3)}.$$

Hence the criteria for breakup of drops in any linear flow can be found from the equations above. Flows in which e_∞ does not exist are not described by this model since they would not deform a drop into a slender pointed shape.

Since e_∞ is frame-indifferent (Chella & Ottino 1985*a*), the criteria derived here are also frame-indifferent. The methods of flow classification provide a general framework for describing the kinematics of flows, and in the next section we examine the criteria for breakup found above in terms of the kinematics of all steady linear flows.

5. Flow classification with respect to breakup of slender drops

The above analysis indicates that the relevant flow classification parameter in the context of the breakup of slender axisymmetric drops is the asymptotic efficiency e_∞.

We emphasize that e_∞ is the asymptotic efficiency of the hypothetical flow $\bar{\boldsymbol{M}}$, which depends on the drop length, and takes into account the interaction between the drop and the imposed flow. We consider below the behaviour of the asymptotic efficiency in terms of the invariants of the hypothetical flow $\bar{\boldsymbol{M}}$.

The two invariants of the hypothetical flow, $\operatorname{tr}(\bar{\boldsymbol{M}}^2)$ and $\det(\bar{\boldsymbol{M}})$, which specify the asymptotic efficiency, may be expressed in terms of the invariants of the imposed flow as

$$\operatorname{tr}(\bar{\boldsymbol{M}}^2) = 1-\frac{2\,|\bar{\boldsymbol{\omega}}|^2}{\dot{\gamma}^2 G^2},$$

$$\det(\bar{\boldsymbol{M}}) = \frac{\det(\bar{\boldsymbol{D}})}{\dot{\gamma}^3}+\frac{e_{\bar{\omega}}\,|\bar{\boldsymbol{\omega}}|^2}{\dot{\gamma}^2 G^2},$$

where $\bar{\boldsymbol{\omega}}$ is the spin vector, which is related to the vorticity tensor by $\bar{\boldsymbol{\Omega}}\cdot\boldsymbol{b} = \bar{\boldsymbol{\omega}}\times\boldsymbol{b}$ for any arbitrary vector $\boldsymbol{b}$, and $e_{\bar{\omega}}$ is the efficiency, which is defined as

$$e_{\bar{\omega}} = \frac{\bar{\boldsymbol{D}}:\boldsymbol{m}_{\bar{\omega}}\boldsymbol{m}_{\bar{\omega}}}{\dot{\gamma}}, \quad \boldsymbol{m}_{\bar{\omega}} = \frac{\bar{\boldsymbol{\omega}}}{|\bar{\boldsymbol{\omega}}|}.$$

$|\bar{\boldsymbol{\omega}}|/\dot{\gamma}$ is simply the relative magnitude of the vorticity, $\det(\bar{\boldsymbol{D}})/\dot{\gamma}^3$ specifies the distribution of the extensional component, and $e_{\bar{\omega}}$ depends on the orientation of the spin vector relative to the principal directions of strain. Since $|\bar{\boldsymbol{\omega}}|/\dot{\gamma}$ is not frame-indifferent, neither $\operatorname{tr}(\bar{\boldsymbol{M}}^2)$ nor $\det(\bar{\boldsymbol{M}})$ are frame-indifferent. However, the asymptotic efficiency, which is a function of the two, is, as is obvious from its definition.

The *accessible flow domain*, defined by Olbricht *et al.* (1982), is a region in the $(\operatorname{tr}(\bar{\boldsymbol{M}}^2), \det(\bar{\boldsymbol{M}}))$-space which encompasses all possible flows for a given G. Following Olbricht *et al.* and Chella & Ottino (1985*a*), we obtain explicit expressions for e_∞ by considering its character in the domain. The range of values accessible by $\operatorname{tr}(\bar{\boldsymbol{M}}^2)$ is

$$1 \geqq \operatorname{tr}(\bar{\boldsymbol{M}}^2) \geqq -\infty.$$

and for each value of $\operatorname{tr}(\bar{\boldsymbol{M}}^2)$ the range of accessible values for $\det(\bar{\boldsymbol{M}})$

$$(4-3\operatorname{tr}(\bar{\boldsymbol{M}}^2))/3\surd 6 \geqq \det(\bar{\boldsymbol{M}}) \geqq -(4-3\operatorname{tr}(\bar{\boldsymbol{M}}^2))/3\surd 6.$$

The above limits are easily obtained by considering the limits of each of the individual invariants of the imposed flow given above. (Our results are slightly different from those of Olbricht *et al.* owing to our choice of the characteristic time.) In region BOAB′ (figure 3) the largest positive part of the eigenvalues occurs in the root of the cubic equation that is purely real, and the asymptotic efficiency is given by

$$e_\infty^3-\tfrac{1}{2}\operatorname{tr}(\bar{\boldsymbol{M}}^2)\,e_\infty-\det(\bar{\boldsymbol{M}}) = 0.$$

The dotted lines in figure 3 correspond to $(\det(\bar{\boldsymbol{M}}))^2-(\operatorname{tr}(\bar{\boldsymbol{M}}^2))^3/54 = 0$. In the region BOCB″ (figure 3) the largest positive part of the eigenvalues occurs in the complex-conjugate roots, and hence the asymptotic efficiency is given by

$$-8e_\infty^3+\operatorname{tr}(\bar{\boldsymbol{M}}^2)\,e_\infty-\det(\bar{\boldsymbol{M}}) = 0.$$

Hence in both regions the relationship between $\operatorname{tr}(\bar{\boldsymbol{M}}^2)$ and $\det(\bar{\boldsymbol{M}})$ is linear for a fixed asymptotic efficiency. From the above considerations we see that flows to the right of line DB in the accessible flow domain (figure 3) have a single direction of stretching, and thus would be the ones for which the assumption of a circular cross-section is most accurate.

The derivative of the asymptotic efficiency with respect to the steady-state length

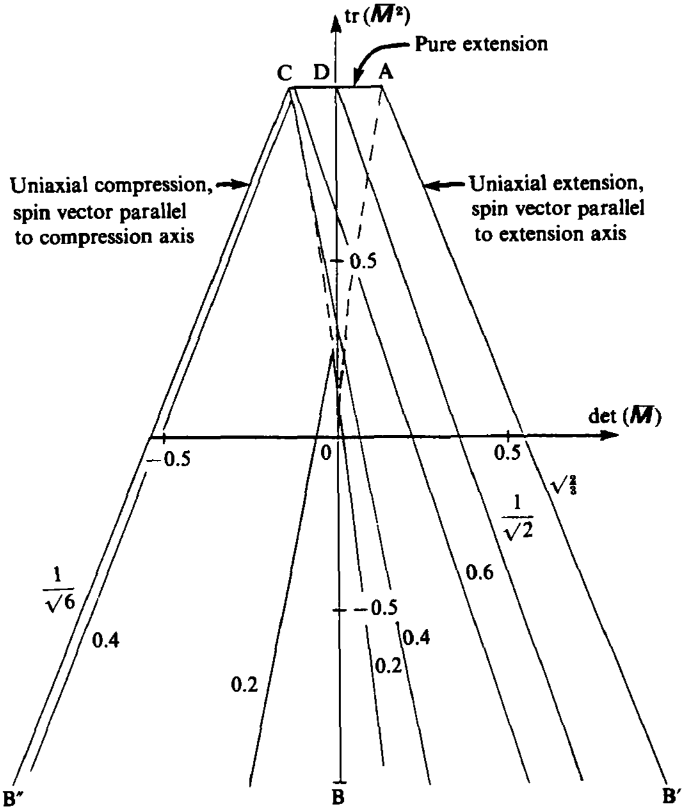


FIGURE 3. Accessible flow domain with lines of constant asymptotic efficiency e_∞.

in each region may now be found by implicitly differentiating the appropriate equation, and the equations for obtaining the critical length reduce to

$$\frac{4l_{sc}^3-1}{1+0.8l_{sc}^3} = A,$$

where
$$A = \frac{2(e_\infty - e_{\bar{\omega}})(|\bar{\omega}|/\dot{\gamma})^2(G-1)(G+5)}{(3e_\infty^2 - \frac{1}{2}\mathrm{tr}(\boldsymbol{M}^2))\,G^3 e_\infty}$$

in region BOAB′, and

$$A = \frac{2(2e_\infty + e_{\bar{\omega}})(|\bar{\omega}|/\dot{\gamma})^2(G-1)(G+5)}{(24e_\infty^2 - \mathrm{tr}(\boldsymbol{M}^2))\,G^3 e_\infty}$$

in region BOCB″. Using one of the above equations, depending on the location within the accessible flow domain, we can calculate l_{sc}^3 by solving the corresponding nonlinear algebraic equation. The critical strain rate is easily obtained once the critical drop length is known.

In the limit of purely extensional flows ($|\bar{\omega}|/\dot{\gamma} = 0$) we find $A = 0$, hence $l_{sc}^3 = \frac{1}{4}$ and

$$E_c = 0.148/e_\infty.$$

Identical results are obtained when the spin vector is aligned with the direction of maximum stretching ($e_\infty = e_{\bar{\omega}}$) in region BOAB′ or if it is aligned with the direction of maximum compression ($e_\infty = -\frac{1}{2}e_{\bar{\omega}}$) in region BOCB″.

The results obtained above for general three-dimensional flows are difficult to present in a concise form; we therefore calculate criteria for breakup for the following two particular classes of flows:

(1) two-dimensional flows: here we compare our predictions with experimental results and compare our model in the appropriate limits with previous theoretical results;

(2) a class of three-dimensional flows with an axisymmetric strain component to study the effect of changing the orientation of the spin vector relative to the strain axes.

The main objective of studying flows of type (2) is to elucidate the physics of breakup of drops in three-dimensional flows.

6. Comparison with experimental results

The breakup of drops has been studied experimentally for two-dimensional flows by a number of researchers. Until the recent work of Bentley (1985), experiments had been carried out for simple shear (Karam & Bellinger 1968; Torza *et al.* 1972; Grace 1971) and two-dimensional extensional flow (Taylor 1934; Grace 1971). Bentley (1985) studied the breakup of drops in planar flows generated by a computer-controlled four-roller apparatus. By varying the relative speeds of the rollers, a broad spectrum of planar flows could be obtained ranging from pure extension to pure rotation. Here we compare the predictions of our model with the experimental data of Bentley (1985) and Grace (1971).

All linear two-dimensional flows can be represented by the following velocity field:

$$v_x = Sy, \quad v_y = \psi Sx.$$

$\psi = 1$ corresponds to the case of pure extensional flow, $\psi = 0$ to simple shear and $\psi = -1$ to pure rotation. The data of Bentley (1985) were taken for flows with ψ ranging from 0.2 to 1. The criteria for breakup in two-dimensional flows are given by

$$e_\infty = \frac{1}{\sqrt{2}}\left[1-\left(\frac{(1-\psi)}{G(1+\psi)}\right)^2\right]^{\frac{1}{2}},$$

$$\left.\frac{\partial e_\infty}{\partial l_s}\right|_{l_{sc}} = -\frac{(G-1)(G+5)(1-\psi)^2}{4l_{sc}\, G^3 e_\infty (1+\psi)^2}.$$

In this case $\det(\boldsymbol{M}) = 0$. Numerically calculated values of the critical strain rate are compared with the data of Bentley (1985) and Grace (1971) in figure 4. For ease of comparison with the experimental data we have plotted the more familiar dimensionless strain rate defined as

$$Ca = S\mu a/\sigma$$

instead of E_c. In figure 5 we compare our predictions with the experimental data for scalar deformation defined as

$$D_{fc} = \frac{l_{sc} - R_{0sc}}{l_{sc} + R_{0sc}},$$

where R_{0sc} is the radius corresponding to the critical length l_{sc}. Except for the extensional flow data of Grace (1971), the agreement between the two seems to be

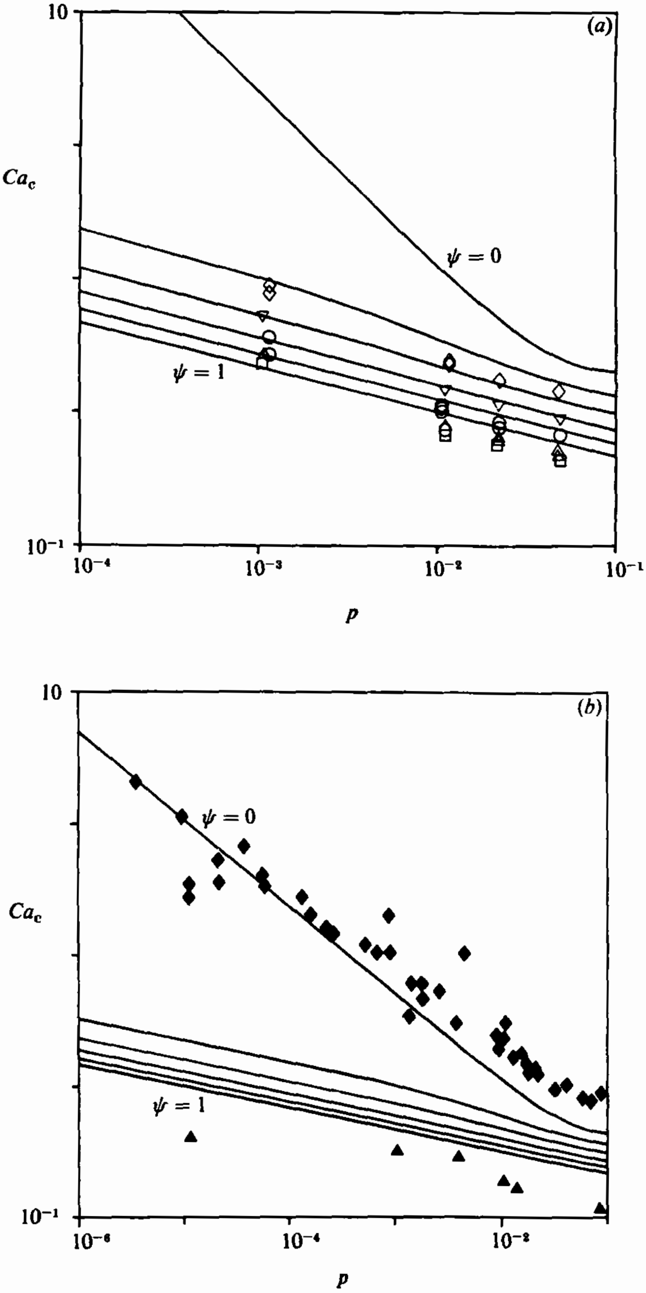


FIGURE 4. Critical capillary number Ca_c versus the viscosity ratio p for two-dimensional flows specified by the parameter ψ. Lines are predictions of the theory for $\psi = 0$, 0.2, 0.4, 0.6, 0.8 and 1.0 in (*a*) and (*b*). (*a*) Data of Bentley (1985) for $\psi = 0.2$ ($\Diamond$), 0.4 (∇), 0.6 ($\bigcirc$), 0.8 ($\triangle$) and 1.0 ($\square$). (*b*) Data of Grace (1971) for $\psi = 0$ ($\blacklozenge$) and 1.0 ($\blacktriangle$).

reasonably good. According to Bentley (1985), the deviation in Grace's (1971) data may be due to the great difficulty in performing experiments in an extensional flow manually, since the drop moves away from the stagnation point at an exponential rate for the smallest deviations in position.

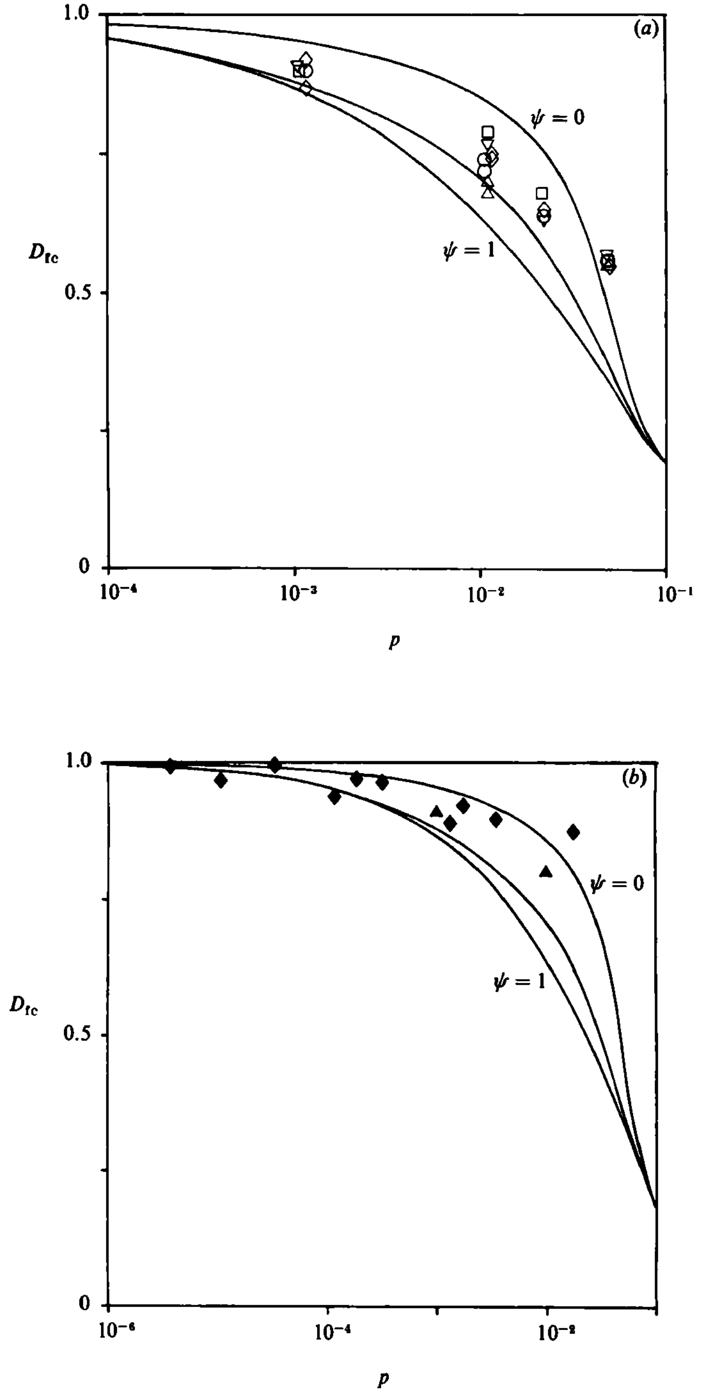


FIGURE 5. Scalar deformation at the point of breakup, D_{fc}, versus the viscosity ratio. Lines are predictions of the theory for $\psi = 0$, 0.2, and 1.0 in (*a*) and (*b*). (*a*) Data of Bentley (1985) for $\psi = 0.2$ (◇), 0.4 (▽), 0.6 (○), 0.8 (△) and 1.0 (□). (*b*) Data of Grace (1971) for $\psi = 0$ (◆) and 1.0 (▲).

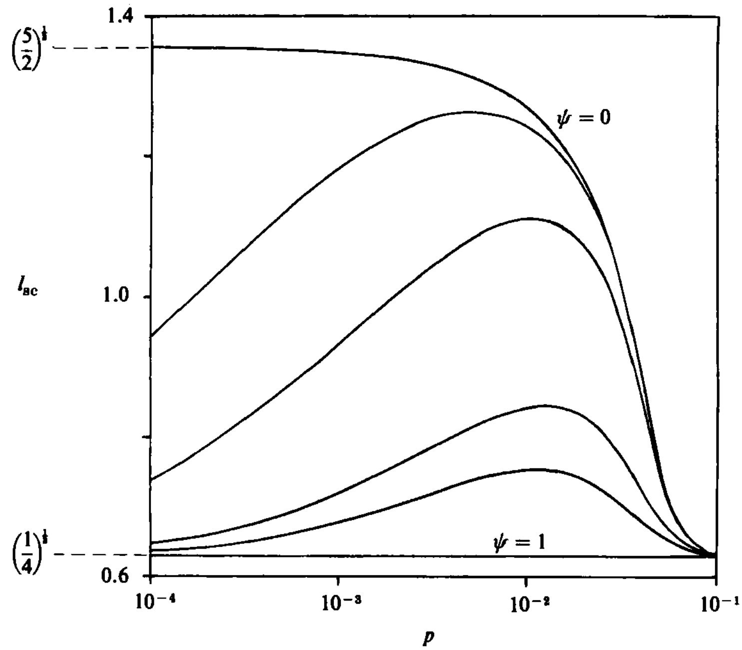


FIGURE 6. Dimensionless half-length of the drop at breakup l_{sc} versus the viscosity ratio p for two-dimensional flows specified by ψ. Lines are predictions of theory for $\psi = 1.0, 0.2, 0.1, 0.01, 0.001$ and 0.

In the limit of low viscosity ratios, approximate expressions may be obtained for the breakup criteria. Two cases are possible.

(1) When the flow is mostly extensional in character ($\psi \approx 1$) we find $\partial e_\infty / \partial l_s|_{l_{sc}} \approx 0$, hence $l_{sc}^3 \approx \frac{1}{4}$ and

$$Ca_c \approx 0.148 \psi^{-\frac{1}{2}} p^{-\frac{1}{6}}.$$

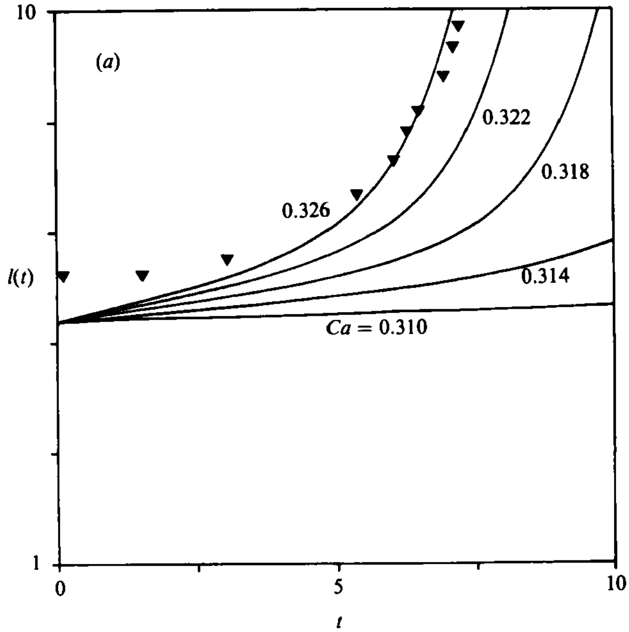
A similar expression (the constant was 0.145) was suggested by Bentley (1985), based on physical arguments. When $\psi = 1$ our prediction is the same as that of Taylor (1964) and Acrivos & Lo (1976) for an axisymmetric flow, hence the model does not distinguish between two-dimensional and axisymmetric extensional flows. This was expected, since we do not permit the drop to become non-axisymmetric in our analysis.

(2) When the flow is simple shear ($\psi = 0$) we find $\partial e_\infty / \partial l_s|_{l_{sc}} \approx -\frac{3}{2} l_{sc} e_\infty$, hence $l_{sc}^3 = \frac{5}{2}$ and

$$Ca_c = 0.0501 p^{-\frac{2}{3}},$$

which are the results obtained by Hinch & Acrivos (1980) for the LA model for simple shear flow.

In figure 6 we plot the exactly calculated critical length versus the viscosity ratio for different flow types. The graph indicates that the above approximations are reasonably accurate for most cases of interest.The graph also illustrates the great difficulty in obtaining experimental data for breakup of drops in simple shear; even the slightest deviations ($\psi = 0.001$) from simple shear ($\psi = 0$) result in drastic changes in the critical length and consequently the critical strain rate for low p. Given



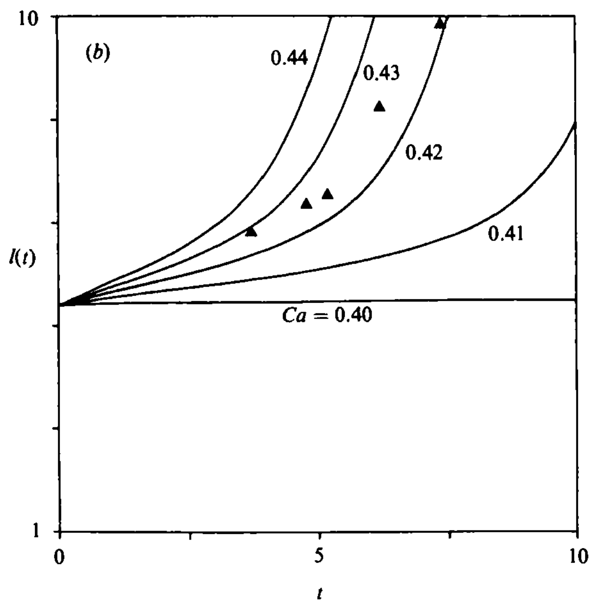


FIGURE 7. Drop length l versus time t for a fixed viscosity ratio $p = 0.012$. Lines are predictions of theory for different strain rates Ca for $\psi = 1.0$ (a) and $\psi = 0.6$ (b). Symbols are data of Bentley (1985) for $\psi = 1.0$ (▼) and 0.6 (▲).

these considerations, the agreement between theory and experiment seems satisfactory.

Bentley (1985) also obtained transient deformation data for strain rates slightly exceeding the critical strain rate. Theoretical prediction of the drop length versus time obtained by numerically integrating the equations for deformation and rotation, with

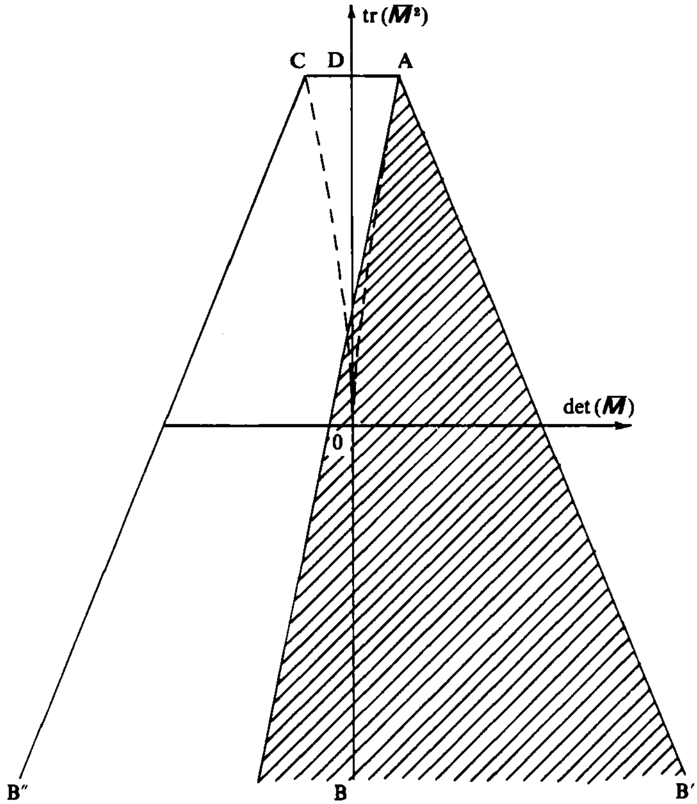


FIGURE 8. Shaded area shows the fraction of the accessible flow domain occupied by the class of flows with an axisymmetric stretching component.

the drop initially at its critical length and orientation, for a number of different strain rates close to the critical strain rate are compared with the data in figure 7. (The dimensionless length and time in this case correspond to those chosen by Bentley (1985) and are $1/a$ and $\psi^{\frac{1}{2}}S_c t$ respectively.) There seems to be qualitative agreement between the two, though the theory under predicts the critical length.

7. Breakup in linear flows with an axisymmetric strain component

In general, a flow can be completely specified by a diagonal rate-of-strain tensor corresponding to an extensional flow and a spin vector corresponding to a solid-body rotation. In the case of two-dimensional flows considered earlier, the spin vector is orthogonal to the principal axes of strain and the flow is completely specified by the relative magnitude of the vorticity ($|\bar{\omega}|/\dot{\gamma}$). Here we study the effect of changing the orientation of the spin vector for a constant relative magnitude of the vorticity. In order to reduce the number of parameters, we consider the class of flows that have an axisymmetric strain component, so that the relative orientation of the spin vector can be specified by a single parameter. The rate-of-strain tensor in this case is given by

$$\overline{D} = \operatorname{diag}(\alpha, -\tfrac{1}{2}\alpha, -\tfrac{1}{2}\alpha)$$

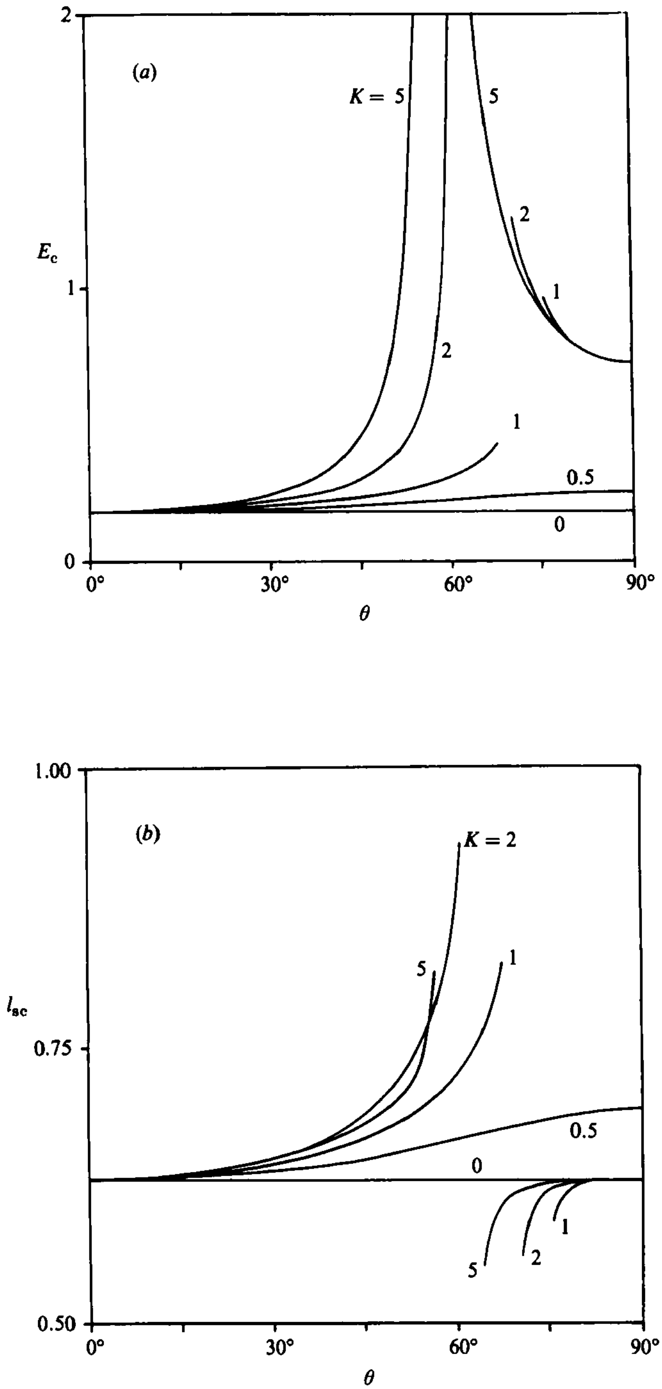


FIGURE 9. Critical dimensionless strain rate E_c (a) and the corresponding critical dimensionless critical half-length l_{sc} (b), versus the orientation of the spin vector θ for fixed viscosity ratio $p = 10^{-3}$ and different amounts of vorticity relative to strain K.

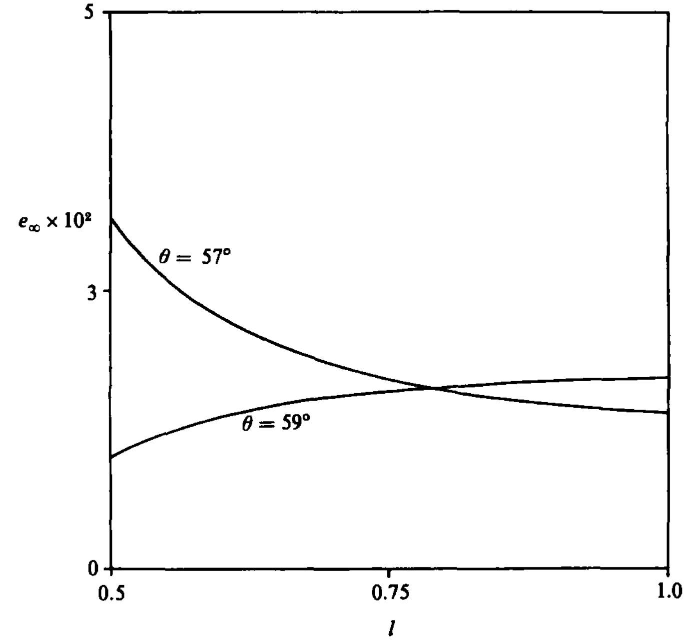


FIGURE 10. Asymptotic efficiency e_∞ versus drop length l for fixed viscosity ratio $p = 10^{-3}$ and relative magnitude of vorticity $K = 5$ for different orientations of the spin vector θ.

and the invariants of the flow by

$$\operatorname{tr}(\boldsymbol{M}^2) = 1 - \frac{K}{G^2}$$

$$\det(\boldsymbol{M}) = \frac{\operatorname{sign}(\alpha)}{3\sqrt{6}}\left\{1 + \frac{3K(3\cos^2\theta - 1)}{2G^2}\right\},$$

where $K = 4|\bar{\omega}|^2/3\alpha^2$ is the relative magnitude of the vorticity, and θ is angle between the spin vector and the axis of stretching (or compression if α is negative). Here we consider only positive α, hence for a fixed value of $\operatorname{tr}(\boldsymbol{M}^2)$ the range of $\det(\boldsymbol{M})$ is

$$\frac{1}{3\sqrt{6}}\left(1 + \frac{9K}{2G^2}\right) \geqq \det(\boldsymbol{M}) \geqq \frac{1}{3\sqrt{6}}\left(1 - \frac{3K}{2G^2}\right),$$

The upper limit corresponding to the boundary of the accessible flow domain. Hence the class of flows under consideration occupy only part of the accessible flow domain, as shown in figure 8.

We consider the breakup of drops in the class of flows defined above. From the equations derived earlier, we calculate the critical drop length and the critical strain rate, for different orientations of the spin vector holding the relative magnitude of the vorticity and the viscosity ratio constant. Figure 9(*a*) shows the critical strain rate versus the angle θ for several values of K. When $K = 0$ the orientation has no effect, as expected, and we obtain the result for an axisymmetric extensional flow. For small values of K ($K = 0.5$) we see an increase in the critical strain rate with increasing angle of orientation of the spin vector. At higher values of K we see that there exists a critical orientation, which depends on the value of K, at which the critical strain rate goes to infinity, implying that the drop cannot be broken in the flow.This value corresponds to the orientation at which the efficiency is undefined ($\mathrm{tr}(\bar{\boldsymbol{M}}^2) < 0$, $\det(\bar{\boldsymbol{M}}) = 0$) and the eigenvalues are purely imaginary. The flow in this case has closed elliptic streamlines in planes parallel to one another, resulting in periodic stretching and compression of material lines (Chella & Ottino 1985*a*), and hence it is unable to cause breakup.

The behaviour of the corresponding critical drop length is shown in figure 9(*b*). Again we see a discontinuity in the graph at the critical angle, but in this case the drop length at breakup is smaller than that for purely extensional flows as we approach the singular point from higher angles, and the reverse is true when we approach it from lower angles. The graph can be explained in terms of the behaviour of the asymptotic efficiency in the different regions of the accessible domain. Figure 10 shows the asymptotic efficiency as a function of the steady-state length for fixed viscosity ratio K and different θ close to the critical orientation. When the flow is located to the left of the line OB (figure 8) in the accessible flow domain the slope of the asymptotic efficiency curve is positive ($\theta = 57°$ in figure 10) and when the flow is to the right of OB the slope is negative ($\theta = 59°$ in figure 10). Recall that at the point of incipient stability the following equation holds:

$$\left.\frac{\partial e_{\infty}}{\partial l_{\mathrm{s}}}\right|_{l_{\mathrm{sc}}} = \frac{1-4l_{\mathrm{sc}}^3}{2l_{\mathrm{sc}}(1+0.8l_{\mathrm{sc}}^3)}.$$

From the above equation we see that if the slope of the asymptotic efficiency curve is positive $l_{\mathrm{sc}}^3 < \frac{1}{4}$, and if it is negative then $l_{\mathrm{sc}}^3 > \frac{1}{4}$, thus justifying the behaviour displayed in figure 9(*b*). (For purely extensional flows ($K = 0$) $l_{\mathrm{sc}}^3 = \frac{1}{4}$.) Since one of the assumptions in our analysis is that the drop is long and slender, the above considerations indicate that the model is not well suited for studying breakup in flows located close to the line OB in the accessible flow domain (figure 3) in region $BOCB''$ where critical drop length is small.

Computations in this section reveal some of the typical behaviour that might be observed in three-dimensional flows. Such behaviour is not possible for two-dimensional flows.

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