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Deformation and burst of a liquid droplet freely suspended in a linear shear field

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A theoretical method is presented for predicting the deformation and the conditions for breakup of a liquid droplet freely suspended in a general linear shear field. This is achieved by expanding the solution to the creeping-flow equations in powers of the deformation parameter ϵ and using linear stability theory to determine the onset of bursting. When compared with numerical solutions and with the available experimental data, the theoretical results are generally found to be of acceptable accuracy although, in some cases, the agreement is only qualitative.

1. Introduction

It has become increasingly apparent in recent years that emulsions, whose striking physico-chemical properties have been the subject of numerous investigations for over a century, are also interesting from the fluid-mechanical point of view in that, when in motion, they often exhibit non-Newtonian effects even when the disperse phase is very dilute. In fact, most of the more common rheological characteristics of non-Newtonian fluids, such as viscoelasticity, shear-dependent viscosity, normal stresses in rectilinear flow, etc., are also generally encountered in the flow of emulsions, with the result that the latter have been used sometimes as models for the study of a class of non-Newtonian substances and for the development of their appropriate constitutive equations. In the case of emulsions which are being sheared, these non-Newtonian effects arise of course from the deformation of the individual droplets, and depend, quantitatively, on a number of parameters, a key one being, no doubt, the average size of these drops. Thus, it is important that more precise information regarding drop deformation and burst be obtained, not only because of its potential usefulness to a number of diverse areas in fluid mechanics, but also because such information is essential for the development of a theory that accurately describes the rheological behaviour of flowing emulsions.

To be sure, the problem of determining theoretically the shape of a single droplet freely suspended in an unbounded incompressible liquid undergoing a shearing motion is very complex and no general solution is at present available. However, by considering only the case of a slightly non-spherical particle,

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Taylor (1932, 1934) was able to show that, to a first approximation, the drop should deform into an ellipsoid. Taylor (1934) also investigated this phenomenon experimentally, and observed that his theoretical expression for the drop deformation agreed with the experimental data only for small values of a non-dimensional shear rate. Furthermore, he found that the mode of burst of the particles depended on the type of shear flow generated in his apparatus and on the ratio λ of the viscosity of the disperse phase to that of the suspending medium.

Following Taylor's pioneering work, numerous authors became interested in the subject. Of particular importance is the experimental work of Rumscheidt & Mason (1961), who studied the deformation and breakup of liquid droplets in hyperbolic and simple shear flows, and the theoretical analysis by Chaffey & Brenner (1967), who improved Taylor's result by deriving a better approximation for the drop shape in a steady simple shear flow. Recent developments on the subject are due to Cox (1969), to Torza, Cox & Mason (1972), who studied both theoretically and experimentally the influence of time effects on the deformation and burst, and, finally, to Grace (1971), who conducted a thorough experimental investigation of these phenomena over the record-breaking range of λ's from 10^{-6} to 10^{3}.

Unfortunately, although the more recent theories have extended the validity of Taylor's original analysis to greater ranges of the shear rate, none of them, so far, has yielded a quantitative criterion for a drop breakup in a general shear flow. Indeed, the basic assumption common to all these theories is that the drop is almost spherical, in contrast to all the experimental observations of drops undergoing breakup, which clearly show that the deformation at this point is large. Furthermore, apart from Cox's (1969) equation, all other theoretical expressions for the drop shape were derived assuming a steady state, whereas the phenomenon of burst is inherently transient.

It follows then, from these considerations, that time effects will have to be included in any complete description of this subject. As shown by Cox (1969), this can be achieved in principle by constructing a solution in which the appropriate variables are expanded in powers of ϵ, a small parameter representing the tendency of the drop to deform. Cox then determined theoretically the transient response of a droplet freely suspended in a time-dependent simple shear or hyperbolic flow, but, since his solution had been evaluated only to $O(\epsilon)$, his results failed to indicate the possibility of particle breakup.

It becomes necessary, therefore, to proceed on the basis of the more complicated analysis by Frankel & Acrivos (1970), as extended by Barthès-Biesel (1972), in which the expansion proposed by Cox was carried out to higher order in ϵ. It will be seen that the resulting equation determining the drop shape, plus the use of linear stability theory do indeed lead to predictions for the breakup of droplets freely suspended in a linear shear field which are in relatively good agreement with the experimental findings by Taylor, by Rumscheidt & Mason, and by Grace. Before proceeding with this comparison, however, we wish to outline briefly the main features of our approach.

2. The method of analysis

As is the case with all the previous studies referred to earlier, the Reynolds number of the motion is assumed small enough for inertia effects to be negligible. Then, as shown by Frankel & Acrivos (cf. their equations (2.1) and (2.10)), the equation for the surface of the drop in a system of axes moving with the centre of the particle can be represented by

$$r = 1 + 3\epsilon F_{lm}\frac{x_l x_m}{r^2} + \epsilon^2\left[-\tfrac{6}{5}F_{lm}F_{lm} + 105F_{lmpq}\frac{x_l x_m x_p x_q}{r^4}\right] + O(\epsilon^3), \tag{2.1}$$

where $r = (x_l x_l)^{\frac{1}{2}}$ and where all position co-ordinates have been rendered dimensionless using the radius a of the equivalent spherical drop as the characteristic length. The Cartesian tensor notation and Einstein's summation convention have been adopted. Also, we shall consider here the case where the drop is kept nearly spherical on account of its large surface tension with λ being $O(1)$, hence the small parameter ϵ is defined as

$$\epsilon = \mu_0 Ga/\sigma,$$

where μ_0 is the viscosity of the suspending fluid, G is the magnitude of the shear rate and σ is the surface tension of the drop.

The tensors F_{ij} and F_{ijlm} are chosen to be symmetric with respect to any permutation of their indices and to have zero contraction. As shown by Barthès-Biesel (1972), they obey the two differential equations

$$\begin{aligned}\epsilon\,\partial F_{ij}/\partial t + \tfrac{1}{2}\epsilon\omega_s(\epsilon_{isl}F_{lj} + \epsilon_{jsl}F_{li}) &\\ = a_0 e_{ij} + a_1 F_{ij} + \epsilon[a_2 Sd(e_{il}F_{lj}) + a_3 Sd(F_{il}F_{lj})] &\\ + \epsilon^2\{a_4 e_{ij}(F_{lm}F_{lm}) + F_{ij}[a_5(F_{lm}e_{lm}) + a_6(F_{lm}F_{lm})] &\\ + a_7 Sd(e_{il}F_{lm}F_{mj}) + F_{ijlm}(a_8 e_{lm} + a_9 F_{lm})\} + O(\epsilon^3) &\end{aligned} \tag{2.2}$$

and
$$\epsilon\,\partial F_{ijlm}/\partial t = b_0 F_{ijlm} + b_1 Sd_4(e_{ij}F_{lm}) + b_2 Sd_4(F_{ij}F_{lm}) + O(\epsilon), \tag{2.3}$$

where e_{ij} and ω_i are respectively the rate of strain and the vorticity of the free stream, rendered dimensionless using G, and t is a non-dimensional time. Also, the symmetric deviators of second- and fourth-order tensors are defined as

$$Sd(A_{ij}) = \tfrac{1}{2}(A_{ij} + A_{ji} - \tfrac{2}{3}\delta_{ij}A_{ll})$$

and

$$\begin{aligned}Sd_4(A_{ijab}) = \tfrac{1}{8}\{A_{ijab} + A_{iabj} + 22 \text{ other terms} - \tfrac{2}{7}[\delta_{ab}(A_{ijll} + A_{iljl} + 10 \text{ other terms}) &\\ + 5 \text{ other terms}] + \tfrac{8}{35}(\delta_{ij}\delta_{ab} + \delta_{ia}\delta_{bj} + \delta_{ib}\delta_{ja})(A_{llmm} + A_{lmlm} + A_{lmml})\}, &\end{aligned}$$

where δ_{ij} is the Kronecker delta. The coefficients a_n and b_n are known rational functions of λ and are given in the appendix.

It is evident of course that, if (2.1) is to be consistent with the order of approximation of the perturbation expansion, it should contain the $O(\epsilon^3)$ terms as well, plus, in particular, a sixth-order tensor F_{ijlmpq} in addition to F_{ij} and F_{ijlm}. However, since neither the equation satisfied by this tensor F_{ijlmpq} nor the $O(\epsilon)$ terms in (2.3) could be derived because of their complexity, (2.1) will be assumed from now on to represent exactly the equation for the surface.

In the following, the full equations (2.2) and (2.3) will be termed the $O(\epsilon^2)$ theory or $O(\epsilon^2)$ results, whereas the relations obtained from (2.2) with terms of $O(\epsilon^2)$ neglected (which, incidentally, is the non-dimensional form of equation (3.5) of Frankel & Acrivos (1970)) will be called the $O(\epsilon)$ theory or $O(\epsilon)$ results. In both cases, however, (2.3) will be retained in its entirety as shown.

The approach taken here will consist of assuming that (2.1), (2.2) and (2.3) represent the *exact* solution to the full problem, and that they apply for all values of ϵ, now being thought of as a measure of the shear rate for a given suspension, and for all values of λ. Thus, this model will allow us to treat simultaneously the two extreme cases of high surface tension or high viscosity drops.

We begin by considering the steady-state shape of the drop for a constant shear field, which can be obtained by solving (2.2) and (2.3) with the time derivatives set equal to zero. Noting that (2.3) can be solved immediately for F_{ijlm} in terms of e_{ij} and F_{ij}, we have then, in lieu of (2.2),

$$\begin{aligned}&-\tfrac{1}{2}\epsilon\omega_s(\epsilon_{isl}F_{lj}+\epsilon_{jsl}F_{li})+a_0e_{ij}+a_1F_{ij}+\epsilon[a_2Sd(e_{il}F_{lj})+a_3Sd(F_{il}F_{lj})]\\&\quad+\epsilon^2\{a_4e_{ij}(F_{lm}F_{lm})+F_{ij}[a_5(F_{lm}e_{lm})+a_6(F_{lm}F_{lm})]\\&\quad+a_7Sd(e_{il}F_{lm}F_{mj})-b_0^{-1}[b_1Sd_4(e_{ij}F_{lm})+b_2Sd_4(F_{ij}F_{lm})](a_8e_{lm}+a_9F_{lm})\}=0.\end{aligned}$$

However, from the definition of a fourth-order symmetric deviator, it follows that

$$\begin{aligned}Sd_4(A_{ij}B_{lm})C_{lm}=\tfrac{18}{7}Sd(A_{il}C_{lm}B_{mj})+\tfrac{3}{14}A_{ij}(B_{lm}C_{lm})\\+\tfrac{3}{14}B_{ij}(A_{lm}C_{lm})-\tfrac{6}{35}C_{ij}(A_{lm}B_{lm}),\end{aligned}$$

where use has been made of a tensorial identity established by Rivlin (1955). Thus, the steady-state equation becomes

$$\begin{aligned}&a_0e_{ij}+a_1F_{ij}+\epsilon[a_2Sd(e_{il}F_{lj})+a_3Sd(F_{il}F_{lj})-\tfrac{1}{2}\omega_s(\epsilon_{isl}F_{lj}+\epsilon_{jsl}F_{li})]\\&\quad+\epsilon^2\{e_{ij}[c_1(F_{lm}F_{lm})+c_2(e_{lm}F_{lm})]+F_{ij}[c_3(F_{lm}F_{lm})+c_4(e_{lm}F_{lm})+c_5(e_{lm}e_{lm})]\\&\quad+c_6Sd(e_{il}F_{lm}F_{mj})+c_7Sd(e_{il}e_{lm}F_{mj})\}=0,\end{aligned}\tag{2.4}$$

where

$$c_1=a_4-\frac{39}{35}\frac{a_8b_2}{b_0}-\frac{3}{14}\frac{a_9b_1}{b_0},\quad c_2=-\frac{3}{70}\frac{a_8b_1}{b_0},$$

$$c_3=a_6-\frac{54}{35}\frac{a_9b_2}{b_0},\quad c_4=a_5-3\frac{a_8b_2}{b_0}-\frac{3}{70}\frac{a_9b_1}{b_0},\quad c_5=-\frac{3}{14}\frac{a_8b_1}{b_0},$$

$$c_6=a_7+\frac{36}{7}\frac{a_8b_2}{b_0}-\frac{18}{7}\frac{a_9b_1}{b_0},\quad c_7=-\frac{18}{7}\frac{a_8b_1}{b_0}.$$

The steady-state shape of the drop can then be obtained from (2.4) given λ, ϵ, and time-independent values for e_{ij} and ω_i. Evidently, (2.4), being cubic in F_{ij}, may admit a multiplicity of solutions; indeed, it will be found, in most of the cases considered in this study, that the curves F_{ij} versus ϵ have a turning point similar to that shown on figure 1, thereby implying that the drop could attain at least two steady-state configurations for a given value of the shear rate. Nevertheless, it will be seen in all the subsequent examples that there exists a single solution to

(2.4) which is both stable and physically acceptable in the sense that the resulting expression for r as obtained from (2.1) is everywhere non-negative.

The stability of the steady-state solutions will be systematically investigated by means of a conventional linear analysis in which, with the flow kept constant, small perturbations F'_{ij} and F'_{ijlm} are superimposed upon the steady-state values $\overline{F}_{ij}$ and $\overline{F}_{ijlm}$ of the tensors, and are chosen such that

$$F'_{ij} \ll 1, \quad F'_{ijlm} \ll 1$$

for all i, j, l and m. Equations (2.2) and (2.3) then become, upon linearization,

$$\begin{aligned}\epsilon\partial F'_{ij}/\partial t = {} & a_1 F'_{ij} + \epsilon[-\tfrac{1}{2}\omega_s(\epsilon_{isl}F'_{lj} + \epsilon_{jsl}F'_{li}) + a_2 Sd(e_{il}F'_{lj}) + 2a_3 Sd(\overline{F}_{il}F'_{lj})] \\ & + \epsilon^2\{F'_{lm}[(2a_4 e_{ij} + 2a_6\overline{F}_{ij})\overline{F}_{lm} + a_5\overline{F}_{ij}e_{lm} + a_9\overline{F}_{ijlm}] \\ & + F'_{ij}[a_5(\overline{F}_{lm}e_{lm}) + a_6(\overline{F}_{lm}\overline{F}_{lm})] + a_7 Sd(e_{il}\overline{F}_{lm}F'_{mj}) \\ & + a_7 Sd(e_{il}\overline{F}'_{lm}\overline{F}_{mj}) + F'_{ijlm}(a_8 e_{lm} + a_9\overline{F}_{lm})\}\end{aligned} \qquad (2.5a)$$

and

$$\epsilon\,\partial F'_{ijlm}/\partial t = b_0 F'_{ijlm} + b_1 Sd_4(e_{ij}F'_{lm}) + 2b_2 Sd_4(\overline{F}_{ij}F'_{lm}). \qquad (2.5b)$$

The problem is thus reduced to the solution of a linear system of first-order ordinary differential equations. For the steady state to be stable, the amplitudes of the disturbances F'_{ij} and F'_{ijlm} must decay exponentially with time, which means that all the eigenvalues of the matrix of the system (2.5) must have negative real parts. The stability analysis consists, therefore, of studying the signs of the eigenvalues of a real non-symmetric matrix. Although this is a formidable problem leading, in general, to long computations, for the special flows considered here, the dimensions of the different matrices do not exceed 8; hence, the well-known method of Routh was used with the characteristic polynomial of the matrix being computed by Leverrier's algorithm.

The choice of the values of ϵ was guided by the results of the regular perturbation solution, from which it is known that, as $\epsilon \to 0$, there exists a stable steady-state shape given by $F_{ij} = -(a_0/a_1)\,e_{ij}$. It is then natural to examine the existence of solutions to (2.4) by increasing ϵ, starting from $\epsilon = 0$. It will be found, in most cases to be investigated, that, when $|\epsilon|$ exceeds a critical value $|\epsilon|_{\text{crit}}$, no steady-state solution exists and that, consequently, the droplet bursts.

In the following, the solutions for three particular shear flows will be examined: an extensional flow, a hyperbolic flow and a simple shear flow, for which, as will be seen, the basic equations (2.2) and (2.3) simplify considerably.

3. Extensional flow

Here, the undisturbed flow field is given by

$$u_1 = -Gx_1, \quad u_2 = -Gx_2, \quad u_3 = 2Gx_3,$$

or, in dimensionless form,

$$e_{11} = e_{22} = -1, \quad e_{33} = 2, \quad \omega_i = 0.$$

G can be either positive or negative and, correspondingly, ϵ will take both signs. Hence, the drop is pulled either along the axis of revolution or along a ring in the x_1, x_2 plane, depending on whether, respectively, ϵ is positive or negative.

Owing to the symmetry of the flow, it can be shown that the only real solution(s) of (2.4) must also be axisymmetric; consequently

$$F_{11} = F_{22} = -\tfrac{1}{2}F_{33} = F.$$

Thus, for an extensional flow, (2.4) reduces to a single equation:

$$6c_3\epsilon^2F^3 - \epsilon[a_3 + 6\epsilon(c_1 + c_4 + \tfrac{1}{2}c_6)]F^2 + \{a_1 + a_2\epsilon + 6\epsilon^2(c_2 + c_5 + \tfrac{1}{2}c_7)\}F - a_0 = 0. \quad (3.1)$$

Since, in this particular case, the $O(\epsilon)$ equation is quite tractable, it will be studied in detail in order to illustrate the method of analysis.

3.1. *The predictions of the $O(\epsilon)$ theory*

To $O(\epsilon)$, (3.1) becomes

$$a_3\epsilon F^2 - (a_1 + a_2\epsilon)F + a_0 = 0, \quad (3.2)$$

where a_0 and a_3 are always positive and a_1 always negative. This equation will have real solutions provided that its discriminant is positive or zero, i.e.

$$(a_1 + a_2\epsilon)^2 - 4a_0a_3\epsilon \geqslant 0,$$

from which it follows that

$$\epsilon \leqslant \{2a_0a_3 - a_1a_2 - [4a_0a_3(a_0a_3 - a_1a_2)]^{\frac{1}{2}}\}/a_2^2, \quad (3.3)$$

or

$$\epsilon \geqslant \{2a_0a_3 - a_1a_2 + [4a_0a_3(a_0a_3 - a_1a_2)]^{\frac{1}{2}}\}/a_2^2. \quad (3.4)$$

For values of λ ranging from 0 to ∞, it was found that the limiting ϵ's were positive and of order 0·1 for (3.3), and 3·0 for (3.4). However, (3.4) will be ignored from now on because it gives rise to an unrealistic shape for the drop in which r, given by (2.1), attains negative values. Besides, under normal experimental conditions, the initial state corresponds to $\epsilon = 0$, and, as will be shown in the following, the drop bursts before the limit indicated by (3.4) can ever be reached.

A critical value of ϵ past which no steady state exists has therefore been predicted:

$$\epsilon_{\text{crit}}^{+} = \{2a_0a_3 - a_1a_2 - [4a_0a_3(a_0a_3 - a_1a_2)]^{\frac{1}{2}}\}/a_2^2.$$

In addition, when (3.3) is satisfied, two values of F are obtained for a given ϵ:

$$\overline{F}_{\pm} = \{a_1 + a_2\epsilon \pm [(a_1 + a_2\epsilon)^2 - 4a_0a_3\epsilon]^{\frac{1}{2}}\}/2a_3\epsilon,$$

of which the physically realistic one is selected on the basis of the linear stability analysis described earlier. Specifically, in view of (3.1), the latter yields in this case three independent differential equations

$$\epsilon\, dF'/dt = (a_1 + \epsilon a_2 - 2\epsilon a_3\overline{F})F',$$
$$\epsilon\, dF'_{12}/dt = (a_1 - \epsilon a_2 + 2\epsilon a_3\overline{F})F'_{12},$$
$$\epsilon\, dF'_{13}/dt = (a_1 + \tfrac{1}{2}\epsilon a_2 - \epsilon a_3\overline{F})F'_{13},$$

the solutions to which will decay only if

$$a_1 + \epsilon a_2 - 2\epsilon a_3\overline{F} < 0, \quad (3.5a)$$

$$a_1 - \epsilon a_2 + 2\epsilon a_3\overline{F} < 0, \quad (3.5b)$$

$$a_1 + \tfrac{1}{2}\epsilon a_2 - \epsilon a_3\overline{F} < 0. \quad (3.5c)$$

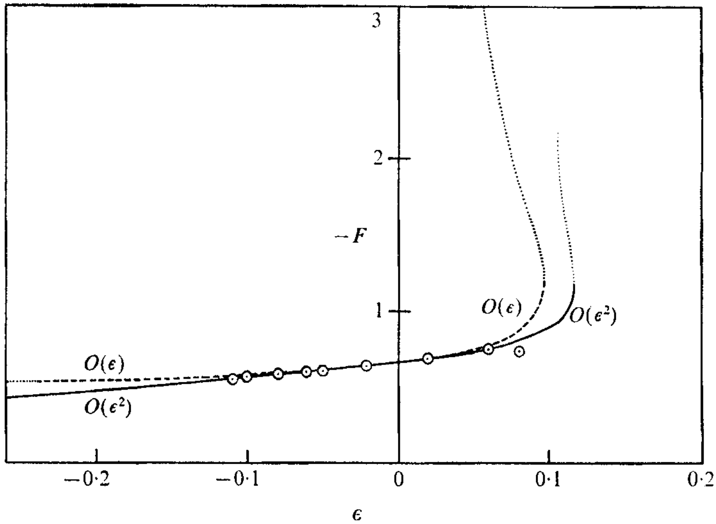


FIGURE 1. Extensional flow: steady-state values of F for $\lambda = 0$. ⊙, exact numerical solution; $\cdots$, unstable state.

Obviously $\overline{F}_-$ never satisfies (3.5*a*) and can be immediately discarded, so that the solution is unique. Similarly, $\overline{F}_+$ always satisfies (3.5*a*) and (3.5*c*) since a_1 is negative. In contrast, $\overline{F}_+$ satisfies (3.5*b*) only if ϵ exceeds a critical value

$$\epsilon_{\text{crit}}^- = \{2a_0a_3 - a_1a_2 - [(2a_0a_3 - a_1a_2)^2 + 3a_1^2a_2^2]^{\frac{1}{2}}\}/a_2^2,$$

which is real and negative for all values of λ. Evidently, when $\epsilon < \epsilon_{\text{crit}}^-$, the instability results from the F'_{12} disturbance. A curve of $\overline{F}_+$ versus ϵ is plotted in figure 1 for $\lambda = 0$.

It has been shown then that, according to the $O(\epsilon)$ theory, a stable physically realistic droplet shape cannot be attained in an extensional flow when the shear rate exceeds a certain critical value and that, consequently, the drop bursts. It is also of interest to remark at this point that one burst criterion provided by this analysis, namely $\epsilon < \epsilon_{\text{crit}}^+$, qualitatively corresponds to that given by Taylor (1932), who postulated that breakup would take place when the surface tension forces, tending to keep the drop in one piece, could no longer balance the viscous forces, tending to disrupt the particle. Specifically we note that, since the differential equations (2.2) and (2.3) as obtained from the perturbation method correspond to a normal stress balance at the interface, the left-hand side of (3.1) or (3.2) represents the difference between the viscous force and surface tension force at the point where the surface of the drop intersects the x_3 axis. From very simple algebraic considerations, it then follows that this quantity will always be positive when ϵ is greater than ϵ_{crit}^+, which confirms Taylor's hypothesis and shows that the drop will break when, at the point of maximum extension, viscous forces exceed those arising from surface tension.

3.2. *The predictions of the $O(\epsilon^2)$ theory*

The same procedure as that illustrated in § 3.1 was followed here, the difference being that the various equations were studied numerically since no simple analytical solution is available for a cubic equation. In particular (3.1) was solved by the Newton–Raphson method for various values of ϵ and λ. Again, the curves F versus ϵ obtained for positive ϵ showed clearly the existence of a critical ϵ, which differed somewhat, but not significantly, from that predicted by the $O(\epsilon)$ theory. In contrast, however, the trend of these curves for negative ϵ was quite different in the sense that, whereas the $O(\epsilon)$ values of F increased steadily with ϵ, the $O(\epsilon^2)$ curves were observed to have a turning point for some λ's similar to that for positive ϵ.

It was found numerically from the stability analysis that the upper parts of the curves F versus ϵ, for positive and negative values of ϵ, are unstable and that again, when ϵ is negative, instability is first caused by the F'_{12} disturbance. Thus, as with the $O(\epsilon)$ theory, breakup in this latter case will be due to any small three-dimensional disturbance.

3.3. *Comparison with a numerical solution of the creeping-flow equations*

At present there exist no experimental results regarding the deformation and burst of liquid droplets immersed in an elongational flow, owing to the technical problems encountered in the design of an apparatus to produce such a flow field. However, extensional flow does occur in nature, for example, when a liquid thread is pulled. At any rate, it appears that the only results with which the predictions of the theory can be compared are those obtained by Frankel & Acrivos (1970) from a numerical solution of the creeping-flow equations, at steady state, for $\lambda = 0$.† Unfortunately, the numerical scheme used by these authors failed for values of ϵ less than $-0{\cdot}11$ and greater than $0{\cdot}08$ probably owing to the fact that the elongated drop shape could no longer be represented by a converging series of surface spherical harmonics as required by their technique. Whether this failure indicates drop breakup is not clear at this point and further analysis is required. However, one can still make use of these numerical results for values of ϵ ranging from $-0{\cdot}11$ to $0{\cdot}08$, in order to assess the accuracy of the perturbation analysis developed in the previous sections. In particular, it is seen in figure 1, where the numerical values of F have also been plotted, that good agreement exists between the theoretical and the exact results. Thus it is to be expected that the drop shapes predicted by the theory (whether it is the $O(\epsilon)$ or the $O(\epsilon^2)$, since the two do not differ appreciably in this range of values of ϵ) will be very close to those computed from the numerical solution.

† Recently, Buckmaster (1972, 1973) also considered this problem for a range of λ's, and determined the droplet shape using slender-body theory. He showed that his solution is non-unique, which is in agreement with our findings, but otherwise his results differ quantitatively from ours. In particular, according to his solution, a steady shape, not necessary stable, exists for all positive ϵ, provided that $\lambda = 0$. Evidently, this matter deserves further study.

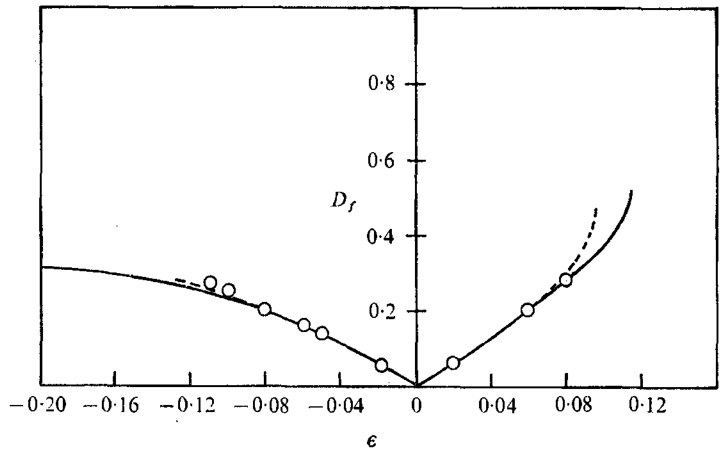


FIGURE 2. Extensional flow: comparison between the analytical and numerical values of the deformation for $\lambda = 0$. ○, numerical; – – –, $O(\epsilon)$; ——, $O(\epsilon^2)$.

The deformation D_f of the particles, a quantity often measured by experimentalists, is, therefore, also studied here. As is customary, D_f is defined by

$$D_f = (L-B)/(L+B),$$

where L and B are respectively the lengths of the major and minor axes of the deformed drop. In the present case, the equation for the surface of the particle stems readily from (2.1) and becomes in the x_1, x_3 plane

$$r = 1 - \tfrac{3}{2}\epsilon F + \epsilon^2\left[-\frac{729}{8b_0}F(-b_1+b_2F) - \tfrac{36}{5}F^2\right] + \epsilon\left[-\tfrac{9}{2}F - \frac{399}{2b_0}\epsilon F(-b_1+b_2F)\right]\cos 2\phi - \frac{2835}{8b_0}\epsilon^2 F(-b_1+b_2F)\cos 4\phi,$$

where ϕ is defined by

$$x_3 = r\cos\phi, \quad x_1 = r\sin\phi.$$

The analytic expression for D_f,

$$D_f = \left|\frac{\epsilon\left[-\tfrac{9}{2}F - \dfrac{399}{2b_0}\epsilon F(-b_1+b_2F)\right]}{1 - \dfrac{3\epsilon}{2}(F + \tfrac{24}{5}\epsilon F^2) - \dfrac{891}{2b_0}\epsilon^2 F(-b_1+b_2F)}\right|, \tag{3.6}$$

thus follows easily.

A comparison between the above values of D_f and the corresponding numerical results is shown on figure 2. The agreement is again good, thereby tending to indicate that the shape of the drop is not very sensitive to small relative variations in ϵ, provided of course that ϵ is less than ϵ^+_{crit} or larger than ϵ^-_{crit}. Also, it is interesting to note that the $O(\epsilon)$ results are slightly better than those of the $O(\epsilon^2)$ theory for negative ϵ. The difference is, however, not significant and may be due to the fact that, as explained earlier following (2.3), the $O(\epsilon^2)$ expression for the shape does not contain all the relevant terms.

λ	$\epsilon^{+}_{\text{crit}}$	$\epsilon^{-}_{\text{crit}}$
0·0	0·115	none found
0·2	0·074	−0·27
0·5	0·058	−0·15
1·0	0·052	−0·13
2·0	0·045	−0·13
4·0	0·044	−0·13
7·0	0·043	−0·13
15·0	0·043	−0·13

TABLE 1. Extensional flow. Critical ϵ for various values of λ (results from the $O(\epsilon^2)$ theory).

The limited comparison between the theoretical and numerical results is therefore very encouraging, and indicates that the theory gives entirely satisfactory predictions in the range of values of ϵ for which a stable drop shape is attained. Hence, the results obtained from the perturbation analysis for other values of λ are also given here (in table 1), because they might be of some use in estimating the maximum drop size that can be attained by viscous droplets freely suspended in this particular flow field.

4. Hyperbolic flow

This undisturbed flow is given here by

$$u_1 = Gx_1, \quad u_2 = -Gx_2, \quad u_3 = 0,$$

or, in dimensionless form, by

$$e_{11} = -e_{22} = 1, \quad \omega_i = 0,$$

all other components of e_{ij} being zero. It then follows from (2.4) that the non-zero components of F_{ij} and F_{ijlm} are

$$F_{11}, F_{22}, F_{33}, \quad F_{1111}, F_{1122}, F_{2222}, F_{1133}, F_{2233}, F_{3333},$$

of which only five are independent on account of the requirement that the tensors have zero contraction. For reasons of symmetry, these are chosen to be

$$S = F_{11} + F_{22}, \quad D = F_{11} - F_{22}, \quad F_{1122},$$

$$S_{1111} = F_{1111} + F_{2222}, \quad D_{1111} = F_{1111} - F_{2222}.$$

Thus, (2.4) reduces to a system of two nonlinear equations in the unknowns S and D,

$$a_1 S + \tfrac{1}{3}\epsilon[a_2 D - \tfrac{1}{2}a_3(3S^2 - D^2)] + \epsilon^2 S[\tfrac{1}{2}c_3(3S^2 + D^2) + D(c_4 + \tfrac{1}{3}c_6) + \tfrac{1}{3}c_7 + 2c_5] = 0, \tag{4.1a}$$

$$a_1 D + 2a_0 + \epsilon S(a_2 + a_3 D) + \epsilon^2[\tfrac{1}{2}(2c_1 + c_3 D)(3S^2 + D^2) + c_4 D^2 + \tfrac{1}{2}c_6(S^2 + D^2) + D(2c_2 + c_7 + 2c_5)] = 0, \tag{4.1b}$$

which was solved numerically for various values of ϵ. Also, the stability equations (2.5) decompose here into a 5×5 matrix $\mathbf{P}$, corresponding to the non-zero steady-state elements of the tensors $\mathbf{F}$, and three 3×3 matrices, corresponding to the other elements.

4.1. *The predictions of the theory*

The results from both the $O(\epsilon)$ and $O(\epsilon^2)$ analysis will be summarized here, since they are qualitatively similar. Specifically, it was found numerically for values of the viscosity ratio ranging from 0 to 20† that a critical ϵ past which the steady-state equations (4.1) had no physically acceptable solution existed in each case, thereby implying that the drop bursts. Also, the graphs F_{11} and F_{22} versus ϵ were very similar to those shown on figure 1 for positive ϵ, their lower branches being stable to all small disturbances and their upper branches unstable on account solely of the matrix $\mathbf{P}$, which has some eigenvalues with a positive real part.

4.2. *Comparison with experimental data*

Three experimental studies of the deformation and burst of a liquid drop in a hyperbolic flow have been conducted, respectively, by Taylor (1934), by Rumscheidt & Mason (1961) and by Grace (1971), who observed that breakup of the particle would generally occur when the rate of strain exceeded a certain limit.

A comparison between the theoretical values of the deformation for $\lambda = 0$, 0·91 and 20 and Taylor's experimental data reveals that, except for $\lambda = 0$, the $O(\epsilon)$ curves are in better agreement with experiment than those computed from the $O(\epsilon^2)$ theory. In contrast, as is apparent from figure 3, the $O(\epsilon^2)$ results follow more closely Rumscheidt & Mason's experimental curve. Since there are some reasons for believing, however, that Taylor's observations were not very accurate and that Rumscheidt & Mason's data are more reliable, it would appear that the $O(\epsilon^2)$ theory is somewhat superior to the $O(\epsilon)$ analysis. As shown in table 2, this latter conclusion is also supported by a comparison between the analytical values of ϵ_{crit} and those given by Rumscheidt & Mason for $\lambda = 1$ and $\lambda = 6$. The experimental value of ϵ_{crit} for $\lambda = 0$, as obtained from Grace's work, suggests, however, that the present theory becomes inaccurate when λ is small. Similarly, for $\lambda > 6$, Grace's results indicate that ϵ_{crit} increases slowly with λ (e.g. $\epsilon_{\text{crit}} = 0{\cdot}17$ for $\lambda = 10^2$), whereas, according to our analysis ϵ_{crit} becomes approximately 0·08 for $\lambda \geqslant 20$. Thus, it would appear that the theory is satisfactory when λ is $O(1)$, i.e. when the true value of ϵ_{crit} is still small.

The drop profiles, as calculated from the $O(\epsilon^2)$ theory for values of ϵ less than, but near ϵ_{crit}, were found to be very similar to those reported by Rumscheidt & Mason for $\lambda = O(1)$ or larger, but unlike the pointed shapes which are observed experimentally for inviscid drops. This last fact should not be too surprising, though, since pointed ends can hardly be described by (2.1), in which only two harmonics have been retained.

† For all practical purposes, a viscosity ratio of 20 can be assumed infinite since the various coefficients a_i and c_i have effectively reached their asymptotic values corresponding to $\lambda \to \infty$.

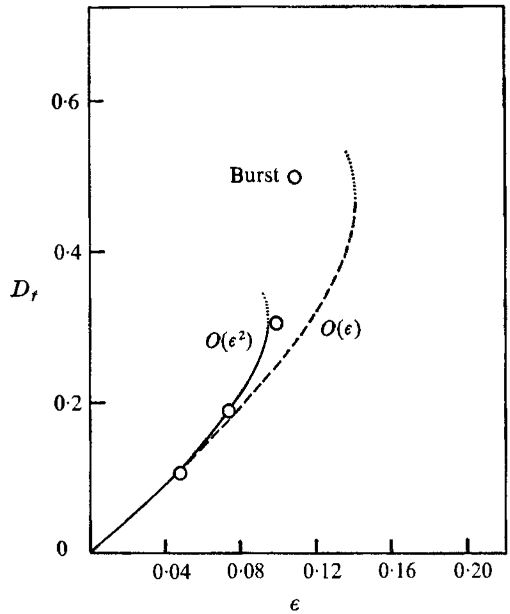


Figure 3. Hyperbolic flow: deformation of a drop. $\lambda = 6$. $\cdots$, unstable state; $\odot$, experiment, Rumscheidt & Mason (1961).

	ϵ_{crit}		
λ	$O(\epsilon)$	$O(\epsilon^2)$	Experimental
0	0·195	0·253	0·5
1·0	0·15	0·104	0·1
6·0	0·14	0·096	0·11

Table 2. Hyperbolic flow: burst criterion.

5. Simple shear flow

A simple shear flow in the x_1, x_2 plane will now be considered. Here

$$u_1 = 2Gx_2, \quad u_2 = u_3 = 0,$$

consequently, the non-dimensional rate of strain and vorticity become

$$e_{12} = 1, \quad \omega_3 = -2,$$

all other components being zero. From (2.4) it follows that the only non-zero components of the tensors F_{ij} and F_{ijlm} are

$$F_{12}, F_{11}, F_{22}, F_{33}, \quad F_{1111}, F_{1222}, F_{1112}, F_{2211}, F_{1133}, F_{2233}, F_{1233}, F_{3333},$$

of which only eight are independent, owing to the requirement that both F_{ij} and F_{ijlm} should have zero contraction. Hence, the solution was developed using the eight independent variables

$$F_{12},\quad S = F_{11}+F_{22},\quad D = F_{11}-F_{22},\quad F_{1122},$$
$$S_{1111} = F_{1111}+F_{2222},\quad D_{1111} = F_{1111}-F_{2222},$$
$$S_{1112} = F_{1112}+F_{1222},\quad D_{1112} = F_{1112}-F_{1222},$$

in terms of which (2.4) reduces to the system of three equations

$$a_1 S + \tfrac{1}{3}\epsilon[2a_2 F_{12} + a_3\{2F_{12}^2 - \tfrac{1}{2}(3S^2 - D^2)\}] + \epsilon^2 S[2c_3\{F_{12}^2 + \tfrac{1}{4}(3S^2 + D^2)\} + 2F_{12}(c_4 + \tfrac{1}{3}c_6) + \tfrac{1}{6}c_7 + 2c_5] = 0, \tag{5.1a}$$

$$a_1 D + \epsilon(4F_{12} + a_3 SD) + \epsilon^2 D[2c_3\{F_{12}^2 + \tfrac{1}{4}(3S^2 + D^2)\} + 2c_4 F_{12} + c_7 + 2c_5] = 0, \tag{5.1b}$$

$$a_1 F_{12} + a_0 + \epsilon[S(\tfrac{1}{2}a_2 + a_3 F_{12}) - D] + \epsilon^2[2\{F_{12}^2 + \tfrac{1}{4}(3S^2 + D^2)\}(c_1 + c_3 F_{12}) + F_{12}^2(2c_4 + c_6) + \tfrac{1}{4}c_6(S^2 + D^2) + F_{12}(2c_2 + c_7 + 2c_5)] = 0. \tag{5.1c}$$

The above were solved numerically by a Newton–Raphson method extended to nonlinear systems, for various values of ϵ increasing from zero. Also, the stability analysis of those steady-state solutions obtained from (5.1) was found to give rise to an 8×8 matrix **P**, corresponding to the non-zero elements of F_{ij} and F_{ijlm}, and to a 6×6 matrix **S**, corresponding to the elements of the tensor **F** which are zero at steady state.

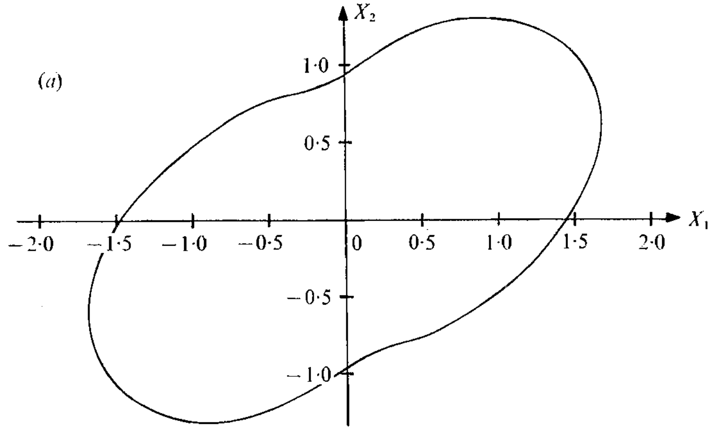
5.1. *The predictions of the $O(\epsilon)$ theory*

The $O(\epsilon)$ theory has already been considered by Chaffey & Brenner (1967) from a point of view slightly different from the one adopted here, in that they used a method of successive approximations to solve the various equations. However, their results are not very different from those of the present approach. In short, according to the $O(\epsilon)$ theory the major axis of deformation of the drop will be first oriented at 45° relative to the x_1 axis and then will tend to rotate towards the x_1 axis as the rate of shear is increased. Also, for all values of the viscosity ratio, the deformation of the particle is a monotonically increasing function of ϵ whose curvature is negative (as will be seen later, this is in disagreement with the experimental evidence, which shows a positive curvature for some values of λ). Furthermore, the $O(\epsilon)$ theory predicts that viscous drops ($\lambda > 3{\cdot}6$) attain a stable limiting shape in which the major axis of the particle is aligned with the x_1 axis.

It is interesting to note that, in contrast to the experimental observations, no critical ϵ is found, i.e. the $O(\epsilon)$ theory predicts that the steady states of the droplet are stable to all small disturbances. This fact seems to be due to the stabilizing effect of the free-stream vorticity.

5.2. *The predictions of the $O(\epsilon^2)$ analysis*

According to this analysis, drops of low viscosity ($\lambda < 0{\cdot}1$) or of high viscosity ($\lambda > 3{\cdot}6$) will behave essentially as predicted by the $O(\epsilon)$ theory and their shapes will be stable to all small disturbances. In contrast, for particles of medium viscosity ($0{\cdot}1 < \lambda < 3{\cdot}6$), a critical value of ϵ past which (5.1) had no solution was found, thus indicating breakup of the drop. Also, the curves F_{11}, F_{22} and F_{12} versus



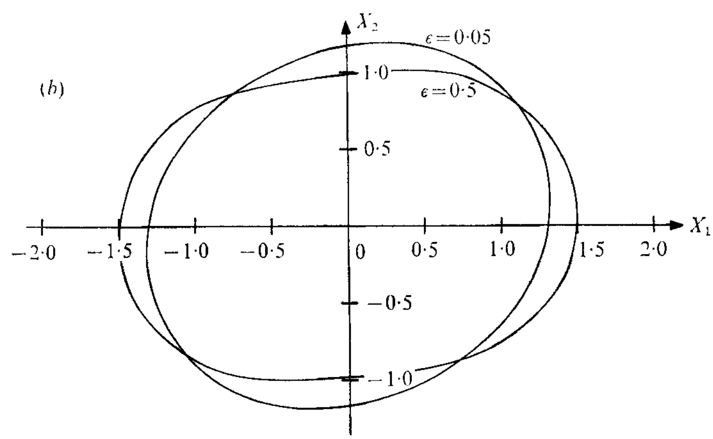


FIGURE 4. Simple shear: limiting shape of drop, as predicted by the $O(\epsilon^2)$ theory. (*a*) $\lambda = 1$, $\epsilon = 0{\cdot}13$. (*b*) $\lambda = 6{\cdot}4$, $\epsilon = 0{\cdot}05$ and $0{\cdot}5$.

ϵ have here a turning point which again, as in the case of extensional or hyperbolic flow, demarcates the stable from the unstable part of the curve, the instability being due only to the matrix **P**.

As should be expected, the $O(\epsilon^2)$ theory also indicates that, with increasing rate of shear, the droplet, which was first oriented at 45°, will tend to rotate towards the x_1 axis and to align itself with this axis when λ is large. Some steady-state profiles of drops in the x_1, x_2 plane are shown in figures 4(*a*) and (*b*). Except for high values of λ, these shapes have a 'bump' which does not seem realistic and which probably would have been removed if higher order tensors and a better approximation for F_{ijlm} had been used in the computation of r.

5.3. *Comparison with experimental data*

Since a simple shear flow is easy to reproduce in a laboratory, some experimental observations of the deformation and burst of liquid droplets are now available. In particular, the results obtained by Rumscheidt & Mason (1961), by Torza

λ	Limiting D_f Experimental	Theoretical
3·8	0·38	0·31
6·4	0·28	0·20
15·0	0·085	0·077

TABLE 3. Simple shear. Limiting values of the deformation for high λ.

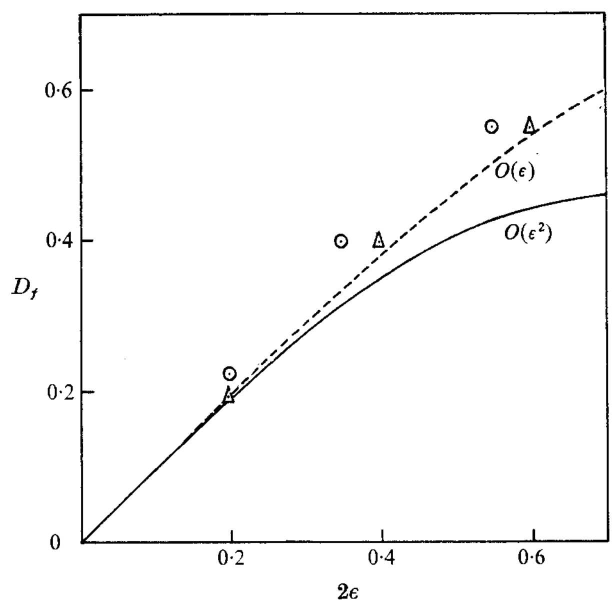


FIGURE 5. Simple shear: deformation of a drop. $\lambda = 0$. Experiment: △, Rumscheidt & Mason; ⊙, Taylor.

et al. (1972) and by Grace (1971) will be used here to assess the validity of the theoretical model.

It has been noted already by Chaffey & Brenner (1967) that the $O(\epsilon)$ theory, when applied to very viscous droplets, is in close agreement with the experimental observations. This is equally true for the $O(\epsilon^2)$ results, which will be compared below with the experimental findings and which, in this case, do not differ substantially from those obtained from the $O(\epsilon)$ analysis.

Shown in table 3 are the limiting deformations reported by Rumscheidt & Mason (1961), as well as those computed from our model, which were found to be in slightly better agreement with experiment than those derived by Taylor (1934). Similarly, there is good agreement between the theoretically and experimentally determined orientations of the particle for $\lambda = 6$. However, this is not the case for $\lambda = 3{\cdot}8$ for reasons which, at present, cannot be explained.

Shown in figure 5 is a corresponding comparison for the case $\lambda = 0$, from which it is clear that the $O(\epsilon)$ predictions are somewhat superior to those arising from

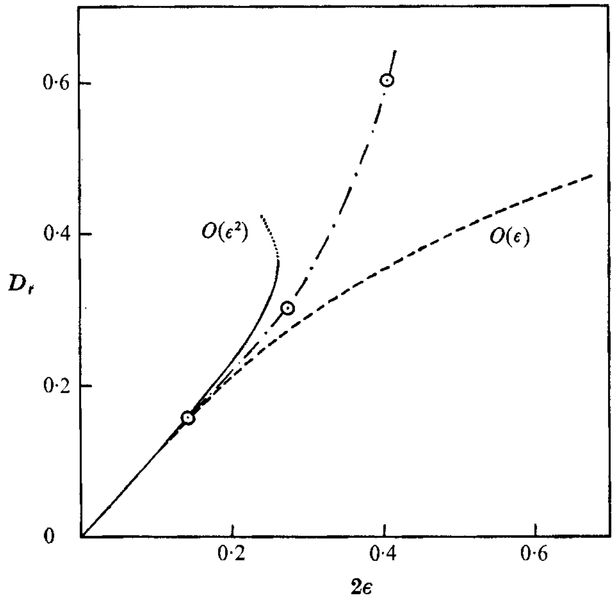


FIGURE 6. Simple shear: deformation of a drop. $\lambda = 1$. ⊙, experiment, Rumscheidt & Mason.

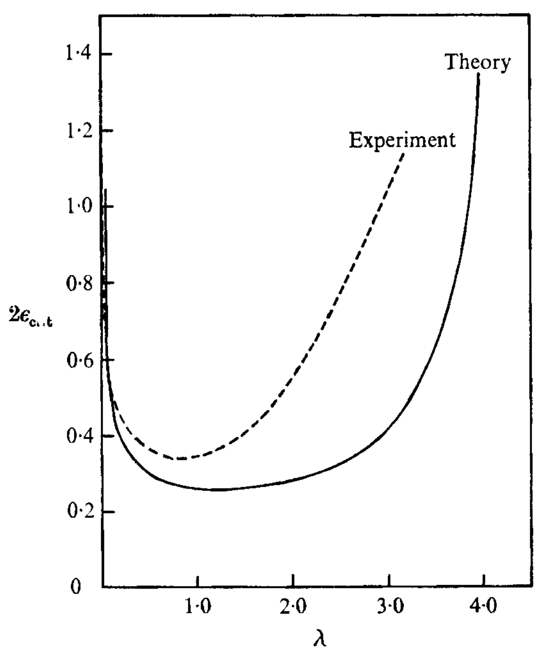


FIGURE 7. Simple shear: variations of ϵ_{crit} with λ. Comparison with experiments of Torza *et al.* (1972).

the $O(\epsilon^2)$ theory. Of more interest, however, are the results for values of λ between 0·1 and 3·6, since, as was said earlier, only the $O(\epsilon^2)$ theory predicts the experimentally observed drop breakup. Such a comparison between the theoretical and the experimental curves for deformation is shown in figure 5 for $\lambda = 1$, from which it is clear that, in describing the trend of the curve, the $O(\epsilon^2)$ theory represents a considerable improvement over the $O(\epsilon)$ analysis, although it indicates bursting of the drop at a lower value of ϵ than is found experimentally. This is one of the major drawbacks of the $O(\epsilon^2)$ results, namely that they are not in very good quantitative agreement with experimental data, although it is felt that they give a good qualitative picture of the phenomenon.

Finally, a plot was made of ϵ_{crit} versus λ and is compared in figure 7 with the corresponding graph given by Torza *et al.* Again, agreement is achieved but only in a qualitative sense. Grace gives a similar graph, but with many more experimental points for small values of λ. From those results, it is possible to conclude that ϵ_{crit} strongly depends on λ when $\lambda < 0{\cdot}1$ and that

$$\epsilon_{\text{crit}} \propto \lambda^{-1\cdot3}.$$

Thus, as λ goes to zero, ϵ_{crit} increases rapidly and attains large values (e.g. $\epsilon_{\text{crit}} \simeq 10$ for $\lambda = 4 \times 10^{-4}$). In contrast, the present theory indicates that no critical value of ϵ exists where λ is smaller than 0·1. Of course, this discrepancy between the theoretical predictions and the experimental evidence should not be too surprising, because the coefficients a_i, b_i or c_i, appearing in the theoretical expressions, are all well-behaved rational functions of λ which tend to a finite limits as $\lambda \rightarrow 0$. Thus, the experimentally observed relation between ϵ_{crit} and λ cannot be predicted analytically. Furthermore, since the theory results from the truncation on an infinite power series, in ϵ, it should be hardly expected to predict reliable values of ϵ larger than unity, such as those found experimentally for $\lambda < 0{\cdot}1$.

6. Discussion

It should be noted here that, since the surface of the drop is assumed to be represented exactly by (2.1), the volume of the drop remains constant to $O(\epsilon^2)$ only. Thus, a simple, but not very sensitive test of the validity of the theory presented earlier consists of evaluating the volume V of the particle, defined by

$$V = \frac{1}{4\pi}\int_S r^3 d\Omega,$$

where r is given by (2.1), $d\Omega$ is an element of a solid angle and S is the surface of the drop.

Shown in tables 4–6 are the computed values of V very near the critical point for, respectively, simple shear, hyperbolic and extensional flows. As is apparent, droplets of low viscosity (i.e. $\lambda < 0{\cdot}3$) can expand by as much as 11 % in the worst case (extensional flow, $\lambda = 0{\cdot}2$, $\epsilon = -0{\cdot}26$), while the change in volume of viscous drops is quite small and at most of order 4 %. It has been shown, however, by

λ	ϵ	V
0	0·15	1·0055
0	0·3	1·051
0	0·5	1·118
0·14	0·24	1·064
1·0	0·13	1·012
2·2	0·15	1·012
4·0	0·5	1·038
17·0	0·5	1·003

TABLE 4. Simple shear: volume of the drop at the point of breakup. $O(\epsilon^2)$ theory.

λ	ϵ	V
0	0·25	1·103
1·0	0·1	1·004
6·0	0·09	1·003
20·0	0·08	1·002

TABLE 5. Hyperbolic flow: volume of the drop at the point of breakup. $O(\epsilon^2)$ theory.

λ	ϵ	V
0	0·115	1·086
	−0·24	1·067
0·2	0·0725	1·037
	−0·26	1·114
0·5	0·0575	1·007
	−0·14	1·017
1·0	0·05	1·005
	−0·12	1·011
4·0	0·04	1·002
	−0·12	1·012
15·0	0·04	1·002
	−0·12	1·012

TABLE 6. Extensional flow: volume of the drop at the point of breakup. $O(\epsilon^2)$ theory.

Barthès-Biesel (1972) that there exists, at steady state, a one-to-one correspondence between the incompressible and compressible cases with the ratio of the appropriate ϵ's being $V^{\frac{1}{3}}$. Thus, since for all the examples considered $V^{\frac{1}{3}}$ was found to lie between 1·0 and 1·03, it follows that the value of ϵ_{crit} is not significantly affected by this change of volume, and that the calculated burst criterion is, approximately, still within the range of validity of the present theory.

Finally, in order to ensure that when $|\epsilon|$ exceeds $|\epsilon_{\text{crit}}|$ our theory would predict drop breakup, the transient deformations of the particle were also studied in the case $|\epsilon| > |\epsilon_{\text{crit}}|$ when, initially, F_{ij} and F_{ijlm} were given values corresponding to those of a known steady shape which exists when $|\epsilon|$ is near but

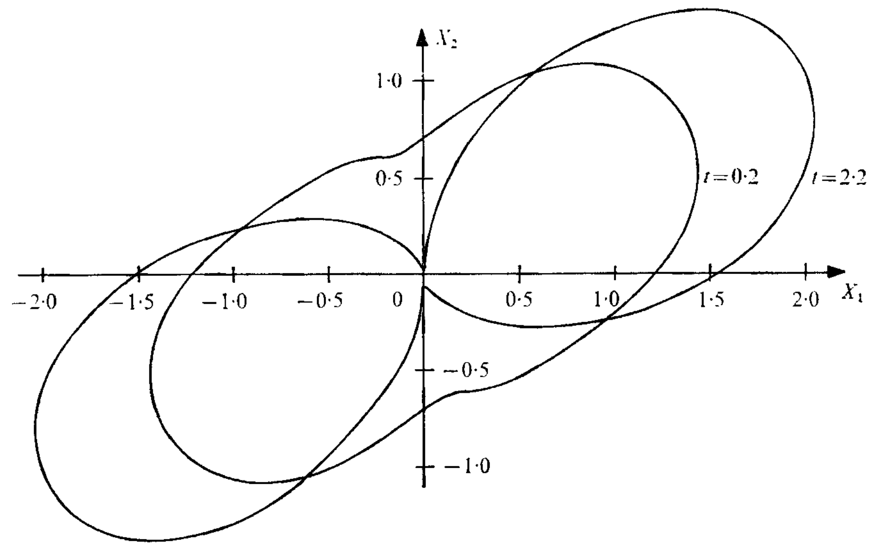


FIGURE 8. Simple shear: burst of a drop, as predicted by the $O(\epsilon^2)$ theory. $\lambda = 1$, $\epsilon = 0·15$.

smaller than $|\epsilon_{crit}|$. Thus, (2.2) and (2.3) were solved as an initial-value problem, using the Kutta–Merson single-step technique.

Unfortunately, when the shear rate exceeded its critical value, the results of the transient analysis indicated that, as a result of the truncation error introduced in (2.1), the increase in the volume of the drop could become significant. For example, the value of V at bursting varied from 1·088 (hyperbolic flow, $\lambda = 6$, $\epsilon = 0·11$) to 1·99 (simple shear, $\lambda = 2·2$, $\epsilon = 0·2$). Consequently, the transient behaviour of the particle, as predicted by this theory, is at best only of qualitative value.

We shall now summarize the results that were obtained in the course of this study from both the steady-state and transient solutions.

For a simple shear flow, the $O(\epsilon^2)$ relations describe correctly the behaviour of very viscous drops ($\lambda > 3·6$). However, the $O(\epsilon)$ equations fail to predict the experimentally observed breakup of the drop for λ less than 3·6. In contrast, the $O(\epsilon^2)$ theory gives a good qualitative description of this phenomenon, since for λ ranging between 0·1 and 3·6, it yields a burst criterion, although it does not predict the bursting of drops of low viscosity which is found to occur at high values of ϵ. Unfortunately, the theoretically computed critical rates of shear and the corresponding deformations are systematically smaller than those observed experimentally.

For values of the rate of shear in excess of the critical ϵ, a transient analysis of the drop shape indicates that the deformation of the particle accelerates with time and that bursting is preceded by a pinching of the middle of the drop as shown in figure 8. This is in agreement with the latest experimental observations of Torza *et al.*, although the experimentally attained deformations of the particle were larger than those found here theoretically.

For a plane hyperbolic flow, a limiting value of the rate of shear, past which no steady state exists, is predicted by both the $O(\epsilon)$ and the $O(\epsilon^2)$ equations for all values of λ. This theoretical burst criterion is in good quantitative agreement with that observed experimentally by Rumscheidt & Mason for viscous drops, the $O(\epsilon^2)$ results being slightly better than the $O(\epsilon)$ ones. However, the mode of burst reported by Taylor or by Rumscheidt & Mason differs from that predicted by the transient theoretical analysis. Specifically, experimental evidence shows that the drop is pulled into a thread or develops pointed ends, whereas the theory indicates that the particle necks in the middle. This discrepancy can be attributed to the use of (2.1) to represent the shape of the drop.

In the case of an elongational flow, a critical rate of shear is again provided by either the $O(\epsilon)$ or the $O(\epsilon^2)$ theories, from which it appears that the viscous drops are the easiest to break. Moreover, a comparison with some numerical results for $\lambda = 0$ shows that the theory gives a good quantitative picture of the phenomenon. However, it was not found possible to compare the theoretical mode of burst, in which the particle necks in the middle, with the actual one, since the scheme for numerically solving the full creeping-flow equations was developed for steady state, and failed before a clearly defined critical shear rate could be reached. In view of the results obtained for hyperbolic flow, however, it might be expected that the theoretical mode of burst might not be very realistic for elongational flow either.

It is interesting to note that, according to the present theory, the drop will burst on account of three-dimensional disturbances when pulled along a ring ($G < 0$).

To conclude, then, we have shown that our analysis predicts the existence of a maximum shear rate beyond which a drop of known size, freely suspended in a linear shear field, is expected to burst. Of course, the agreement between the theoretical predictions and the experimental observations was found, at times, to be only qualitative, yet, considering all the rather drastic simplifications that went into the development of the theory, even this limited agreement is gratifying. Thus, it would appear that the analysis described in this paper could be used in the future with some confidence to estimate the critical value of the non-dimensional shear rate ϵ, and thereby infer some key properties of flowing emulsions (e.g. the maximum size of the droplets), when the viscosity ratio of the two phases is $O(1)$, for the case of general shear flows where no experimental studies of droplet breakup are at present available.

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Appendix

The coefficients a_i and b_i are all rational functions of λ and, as shown by Barthès-Biesel (1972), are given by

$$a_0 = 5/3(2\lambda+3),$$

$$a_1 = -40(\lambda+1)/(2\lambda+3)(19\lambda+16),$$

$$a_2 = 10(4\lambda-9)/7(2\lambda+3)^2,$$

$$a_3 = 288(137\lambda^3+624\lambda^2+741\lambda+248)/7(2\lambda+3)^2(19\lambda+16)^2,$$

$$a_4 = -\frac{2(11\,172\lambda^4+18\,336\lambda^3+17\,440\lambda^2+3499\lambda-7572)}{49(2\lambda+3)^3(19\lambda+16)},$$

$$a_5 = -\frac{2(\lambda-1)(22\,344\lambda^3+52\,768\lambda^2+45\,532\lambda+19\,356)}{49(2\lambda+3)^3(19\lambda+16)},$$

$$a_6 = -48P(\lambda)/49(2\lambda+3)^3(19\lambda+16)^3(10\lambda+11)(17\lambda+16),$$

$$a_7 = 48(\lambda-1)(2793\lambda^3+7961\lambda^2+8474\lambda+3522)/49(2\lambda+3)^3(19\lambda+16),$$

$$a_8 = -400(43\lambda^2+79\lambda+53)/3(2\lambda+3)^2(19\lambda+16),$$

$$a_9 = 80Q(\lambda)/(2\lambda+3)^2(19\lambda+16)^2(10\lambda+11)(17\lambda+16),$$

where

$$P(\lambda) = 2\,127\,976\lambda^7-16\,341\,920\lambda^6-38\,494\,964\lambda^5+122\,942\,551\lambda^4+474\,068\,311\lambda^3 +591\,515\,680\lambda^2+332\,123\,136\lambda+71\,700\,480,$$

$$Q(\lambda) = 405\,260\lambda^5+2\,366\,960\lambda^4+9\,142\,173\lambda^3+8\,595\,967\lambda^2+3\,334\,160\lambda +693\,760.$$

Also

$$b_0 = -360(\lambda+1)/(17\lambda+16)(10\lambda+11),$$

$$b_1 = 1/7(2\lambda+3),$$

$$b_2 = \frac{16(-14\lambda^3+207\lambda^2+431\lambda+192)}{21(2\lambda+3)(19\lambda+16)(17\lambda+16)(10\lambda+11)}.$$

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