Korean J. Chem. Eng., **16**(5), 585-594 (1999)

Deformation and Breakup of a Second-Order Fluid Droplet in an Electric Field

Jong-Wook Ha and Seung-Man Yang†

Department of Chemical Engineering, Korea Advanced Institute of Science and Technology, Taejon 305-701, Korea
(*Received 13 May 1999 • accepted 3 July 1999*)

Abstract–The effects of elastic property on the deformation and breakup of an uncharged drop in a uniform electric field are investigated theoretically using the second-order fluid model as a constitutive equation. Two dimensionless numbers, the electric capillary number (C) and the Deborah number (De), the dimensionless parammeters governing the problem. The asymptotic analytic solution of the nonlinear free boundary problem is determined by utilizing the method of domain perturbation in the limit of small mathcal C and small De. The asymptotic solution provides the limiting point of C above which no steady-state drop shape exists. The linear stability theory shows that the elastic property of fluids give either stabilizing or destabilizing effect on the drop, depending on the deformation mode.

Key words : Electrohydrodynamics, Second-Order Fluid, Drop Deformation, Domain Perturbation

INTRODUCTION

Recently, the dispersion of two immiscible phases has been the subject of intense investigation from both an experimental and a theoretical point of view. Common industrial processes involve dispersion of one fluid phase into another, either to form an emulsion, or to increase the interfacial area between the two phases for more efficient heat and mass transfer. In this paper, we are concerned with one aspect of this general problem; namely, deformation of the interface and linear stability of a single droplet immersed in a continuous phase under the action of a uniform electric field at small Reynolds numbers.

When an uncharged drop is suspended in a dielectric liquid in an external electric field, there is a discontinuity in the stress field at the drop interface. Thus, the interface is deformed from its initial spherical shape due to the mismatch of the normal component of the electric stress [Garton and Krasucki 1964; Taylor 1964; Basaran and Scriven 1989]. In addition, if the conductivities of both phases cannot be neglected, that is, when the two phases are leaky dielectric materials, free charges appear at the drop interface. The action of an electric field on these charges sets the fluids in motion and forming toroidal circulation patterns inside and outside the droplet, which is otherwise quiescent. However, the charge on the two hemispheres of a drop in a uniform electric field is antisymmetric in such a way that the net surface charge is zero [Taylor 1966; Melcher and Taylor 1971; Torza et al., 1971; Arp et al., 1980; Miksis, 1981; Vizika and Saville, 1992; Ha and Yang, 1995; Saville, 1997; Ha and Yang, 1998; Ha and Yang, 1999a, b].

One of the potential technological applications where these effects are prevalent is the processing of a two-phase polymer blend. In this case, the morphology of the dispersed phase, which determines generally the mechanical and other physical properties of the polymer blend, is a crucial factor. A number of studies have considered the underlying physics and processing of polymer blend to elucidate the relationship between morphology and properties of the blend, and at the same time to obtain the desired morphology by applying an external field. Electric field, among other external fields which are used in order to evolve the morphology of the polymer blend, has a few unique advantages including easy manipulation of the field direction and intensity. The dispersed phase of the polymer blend can be easily aligned and stretched to the desired direction by applying the electric field externally.

During the past decade, a few studies concerning the morphology evolution in an immiscible two-phase polymer blend by an external electric field have been reported [see, for example, Moriya et al., 1986; Venugopal and Krause 1992; Xi and Krause 1998]. However, most of these studies utilized theory developed for Newtonian fluids in order to predict the drop deformation. In spite of the non-Newtonian nature of the polymer solutions used, the experimental results did not deviate largely from the predicted theory in the limit of small deformation. However, it has not been confirmed that the stability is not influenced by the viscoelasticity. Unlike the Newtonian fluids, there have been relatively few theoretical investigations relevant to the electrohydrodynamic deformation and stability of non-Newtonian fluids. This is most likely a result of the anticipated uncertainties in selecting of an appropriate constitutive model for non-Newtonian fluids, as well as the obvious difficulty in solving the equations of motion after the choice has been made. In our opinion, however, it is sufficient to consider the influence of small instantaneous departures from Newtonian fluid behavior acting over a large time for this type of problem, at least, from a qualitative point of view.

It is worthwhile to note that the appropriate constitutive model for non-Newtonian fluids which exhibit a slight departure from Newtonian behavior is well-known to be the Rivlin-Ericksen fluid, provided that the motion of fluids are both weak and slow in a rheological sense. This model may be obtained, via the socalled 'retarded-motion' expansion, from almost all of the

†To whom correspondence should be addressed.
E-mail : smyang@cais.kaist.ac.kr

currently popular nonlinear constitutive models [Bird et al., 1987]. Successive terms in the expansion systematically account for the deviation from Newtonianbehavior associated with *elastic effects*. A great deal of phy-sical insight about *elastic effects* can be gained by solving flow problems using ordered-fluid models, even though the ordered-fluids accurately describe neither the dependence of viscosity on the shear rate nor the full range of the time-dependent behavior. Furthermore, the second-order fluid can be prepared experimentally by conventional 'Boger' fluid formation technique [Mackay and Boger, 1987; Tam and Tiu, 1989].

In the present study, we consider theoretically the related problem of the deformation and linear stability of a neutrally buoyant drop in a uniform electric field in otherwise quiescent fluid. The suspending fluid and the fluid inside the drop are assumed to be adequately modeled as second-order fluids. Like most problems, it is impossible to find exact analytical solutions for the deformation and stability of a drop; thus, we turn to a perturbation technique that can be used to develop solutions to flow problems for the retarded-motion expansion at small Deborah numbers. It is noteworthy that since the retarded-motion expansion is itself restricted to a small Deborah number, no significant additional limitations are imposed by the use of the perturbation procedure. However, even when the retarded-motion expansion is used correctly, it is important to note that retaining more terms than the second-order terms in the perturbation solutions often results in series with 'diminishing return'. This clearly indicates that while retention of second-order terms gives both a qualitative and a quantitative description of the deviations from Newtonian behavior, the inclusion of third- and higher-order terms provides only minor improvements to the solution. As a matter of fact, the second-order terms can be determined usually with a moderate analytical effort, but higher-order terms require increasingly tedious and lengthy algebraic developments. Due to these restrictions, the retarded motion expansion is used just through second-order terms for the present analytical investigations on the deviation from Newtonian behavior. The primary thrust of our research is a systematic assessment of the coexisting role of electric field and the elasticity on the drop deformation and stability.

THE PROBLEM STATEMENT

We begin by considering the steady deformation and linear stability of a neutrally buoyant drop suspended in an infinite immiscible fluid under a uniform electric field of strength E^{∞}. The two fluids are assumed to be both incompressible and Rivlin-Ericksen fluids, with zero shear viscosities μ_0 for the suspending phase and $\tilde{\mu}_0$ for the fluid inside the drop. Furthermore, a fluid drop is assumed to be a sphere of radius α in the absence of the electric field. The electrical resistivity of the drop phase is denoted as $\tilde{\chi}$, and the permittivity as $\tilde{\varepsilon}$. Corresponding properties of the ambient fluid are χ and ε, respectively, while the interfacial tension between the drop and the continuous phase is γ. As referred to previously, $\tilde{\chi}$ and χ are not infinite even if they may be very large under the leaky

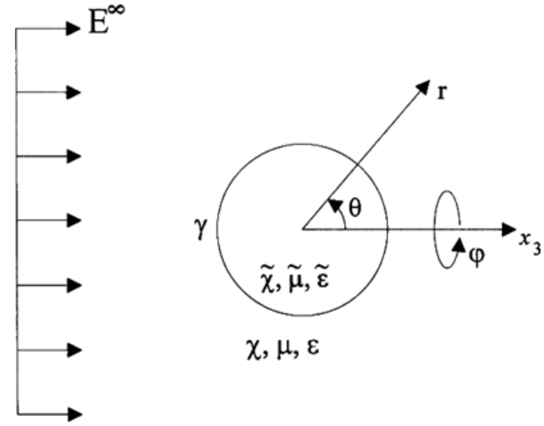

Fig. 1. Spherical co-ordinate system (r, θ, ρ) of the drop in a uniform electric field.

dielectric-fluid assumption. In addition, we adopt a co-ordinate reference system with a fixed origin O at the centre of the drop as illustrated in Fig. 1. In the absence of an electric field, the fluids are quiescent and the drop remains spherical owing to interfacial tension.

The model selected to represent the electric-field part of the problem is deduced from Maxwell's equation by ignoring magnetic effects which are assumed to be insignificant in this study. A further simplification is that the relaxation time for free charges in liquids is short. By ignoring the rates of accumulation and convection of charges and by considering isotropic fluids wherein linear relations prevail between the appropriate vector quantities (e.g. current and electric field), the electric fields can be calculated from a steady-state model of electrostatic phenomena. In this case, the governing equations for the electrostatic potentials $\tilde{V}$ and V inside and outside the drop are the quasi-steady Laplace's equations:

$$\nabla^2 V=0,\ \nabla^2\tilde{V}=0 \ . \tag{1}$$

In addition, the appropriate boundary conditions are as follows:

$$V\to r\cos\theta \text{ as } r\to\infty\ , \tag{2}$$

$$\tilde{V} \text{ is bounded at } r=0\ , \tag{3}$$

$$\mathbf{E}\cdot\mathbf{t}=\tilde{\mathbf{E}}\cdot\mathbf{t} \text{ at } r=1+f, \tag{4}$$

$$\frac{1}{\chi}\mathbf{E}\cdot\mathbf{n}=\frac{1}{\tilde{\chi}}\tilde{\mathbf{E}}\cdot\mathbf{n} \text{ at } r=1+f\ . \tag{5}$$

Here, (2) and (3) describe a uniform electrostatic potential far from the drop and a finite potential at the drop centre, respectively. In the above formulation, the drop interface is defined by r=1+f in which f is the unknown shape function and denotes the departure from sphericity. The continuity of tangential component of the electric fields at the interface and of the conduction current normal to the interface are expressed in (4) and (5). In the latter two equations, $\mathbf{E}$ and $\tilde{\mathbf{E}}$ denote the electric fields developed in both fluid phases and can be related to the electrostatic potentials as $\mathbf{E}=-\nabla V$ and $\tilde{\mathbf{E}}=-\nabla\tilde{V}$. It should be noted that the above equations are nondimensionalized with respect to the characteristic variables such as

$$l_c=a,\ V_c=E^{\infty}a,\ E_c=E^{\infty}\ . \tag{6}$$

Since the electric field is not uniform due to the presence of a fluid drop, electric traction is exerted on the interface. The electric traction can be expressed in terms of the electric stress tensor, which is the so-called Maxwell's stress tensor, defined as

$$\mathbf{T}^{E}=\left(\nabla V \nabla V-\frac{1}{2}\mathbf{I}|\nabla V|^{2}\right). \tag{7}$$

In the above definition, the stress is nondimensionalized by the characteristic value $\varepsilon(E^{\infty})^{2}$. The role of the electric stress in the drop deformation can be seen conveniently by decomposing the electric traction into normal and tangential components to the interface. Due to the normal stress imbalance, the drop cannot remain spherical and must be deformed in order to eliminate this imbalance. Further, the discontinuity of the tangential stresses is responsible for the boundary-driven flow which cancels exactly the tangential stress mismatch produced by the electric field. Since the normal components of the hydrodynamic stresses induced by the boundary-driven flow are also discontinuous at the interface, we must solve the flow problem coupled with the electrostatic one simultaneously to analyze the drop deformation in an electric field.

To formulate the problem for a velocity field generated by the electric stress, the fluid motions are assumed to be dominated by viscous and pressure effects, and the inertial terms in the equations of motion can be neglected entirely so that the fluid motion can be described by the quasi-steady Stokes equation plus the continuity equation. In order to write the governing differential equations and boundary conditions in a nondimensionalized form, we defined the characteristic velocity u_c and characteristic pressure (or stress) p_c as follows

$$u_c=\frac{\varepsilon a(E^{\infty})^{2}}{\mu_0},\ p_c=\varepsilon(E^{\infty})^{2}.$$

The choices of the characteristic variables for the flow field are based on the fact that the flow is generated due to the imbalance of the tangential stress associated with the electric field.

With these conventions and assumptions, the equation of motion and the continuity equation for the suspending fluid can be written in a familiar form

$$\nabla\cdot\mathbf{T}^{H}=0,\ \nabla\cdot u=0. \tag{8}$$

where,

$$\mathbf{T}^{H}=-P\mathbf{I}+\tau_{(1)}+De[\tau_{(1)}\cdot\tau_{(1)}+\phi_1\tau_{(2)}]$$
$$+De^{2}\{\phi_2(\tau_{(1)}:\tau_{(1)})\tau_{(1)}+\phi_3\tau_{(3)}+\phi_4(\tau_{(1)}\cdot\tau_{(2)}+\tau_{(2)}\cdot\tau_{(1)})\}$$
$$+O(De^{3}),$$

and $\tau_{(n)}$ are the Rivlin-Ericksen tensors given by

$$\left.\begin{aligned}
&\tau_{(1)}=(\nabla\mathbf{u})+(\nabla\mathbf{u})^{T};\\
&\tau_{(2)}=\frac{\partial}{\partial t}\tau_{(1)}+\mathbf{u}\cdot\nabla\tau_{(1)}+\tau_{(1)}\cdot(\nabla\mathbf{u})^{T}+\nabla\mathbf{u}\cdot\tau_{(1)};\\
&\tau_{(3)}=\frac{\partial}{\partial t}\tau_{(2)}+\mathbf{u}\cdot\nabla\tau_{(2)}+\tau_{(2)}\cdot(\nabla\mathbf{u})^{T}+\nabla\mathbf{u}\cdot\tau_{(2)}.
\end{aligned}\right\}$$

The Deborah number De and ϕ_1 are dimensionless parameters, defined as $De=\varepsilon(E^{\infty})^{2}\omega_3/\mu_0$ and $\phi_1\ \omega_2/\omega_3$, in which ω_2 and ω_3 denote the dimensional normal stress coefficients, respectively. The Deborah number De is effectively the ratio of an intrinsic relaxation time scale for the fluid to the convective time scale of the fluid motion. As indicated in (7), De is small and the second-order fluid model is applicable to the present case in which non-Newtonian contributions to the fluid motion are assumed to be small. On the other hand, ϕ_1 is of the order unity.

Similarly for the fluid inside the drop, we obtain

$$\nabla\cdot\tilde{\mathbf{T}}^{H}=0,\ \nabla\cdot\tilde{\mathbf{u}}=0, \tag{9}$$

where,

$$\tilde{\mathbf{T}}^{H}=-\tilde{P}\mathbf{I}+\tilde{\tau}_{(1)}+\tilde{De}[\tilde{\tau}_{(1)}\cdot\tilde{\tau}_{(1)}+\phi_1\tilde{\tau}_{(2)}]$$
$$+\tilde{De}^{2}[\tilde{\phi}_2(\tilde{\tau}_{(1)};\tilde{\tau}_{(1)})\tilde{\tau}_{(1)}+\tilde{\phi}_{(3)}\tilde{\tau}_{(3)}+\tilde{\phi}_4(\tilde{\tau}_{(1)}\cdot\tilde{\tau}_{(2)}+\tilde{\tau}_{(2)}\cdot\tilde{\tau}_{(1)})]+O(\tilde{De}^{3}),$$

with $\tilde{\tau}_{(n)}$ defined analogously to $\tau_{(n)}$, but using $\tilde{u}$ instead of $\mathbf{u}$. In this case, $\tilde{De}$ and $\tilde{\phi}_1$ are defined using the same quantities pertaining to the drop phase. The exact relationship between De and $\tilde{De}$ in this situation will be considered in the subsequent sections; for now, we shall simply assume that they are of the same order of magnitude.

Let us then consider the boundary conditions for the flow fields induced by the electric field. For the present case, the continuity of tangential velocities and the kinematic condition on the surface of deformed drop are

$$\mathbf{u}\cdot\mathbf{t}=\tilde{\mathbf{u}}\cdot\mathbf{t}, \tag{10}$$

$$\mathbf{u}\cdot\mathbf{n}=\tilde{\mathbf{u}}\cdot\mathbf{n}=0. \tag{11}$$

In addition, the tangential and normal stress balances at the interface are, respectively,

$$(\mathbf{T}^{E}-S\tilde{\mathbf{T}}^{E}):\mathbf{nt}+(\mathbf{T}^{H}-\lambda\tilde{\mathbf{T}}^{H}):\mathbf{nt}=0, \tag{12}$$

$$(\mathbf{T}^{E}-S\tilde{\mathbf{T}}^{E}):\mathbf{nn}+(\mathbf{T}^{H}-\lambda\tilde{\mathbf{T}}^{H}):\mathbf{nn}=\frac{1}{C}(\nabla\cdot\mathbf{n}). \tag{13}$$

Here λ is a zero-shear-rate viscosity ratio whereas S is the permittivity ratio of drop to continuous phase. In addition, the superscript E and H stand for the electric stress and hydrodynamic stress, respectively. The electric capillary number C is a dimensionless ratio between electric forces and restoring interfacial tension and is given by $C=\varepsilon a(E^{\infty})^{2}/\gamma$. Therefore, De can be related to C as follows; $De=\delta C$, where $\delta=\omega_3\gamma/\alpha\mu_0$. In fact, De is linearly proportional to C.

In general, the problem formulated above is nonlinear, in spite of the fact that the governing equations are linear. The nonlinearity comes solely from the boundary conditions. Another difficulty inherent in this problem arises from the fact that the interface location where the boundary conditions are applied is *a priori* unknown and must be determined as a part of the solution. Thus, for arbitrary C, where the deformation may be quite significant, the problem can only be solved numerically. In the present work, instead, we restrict our attention to the case of small deformations from the spherical shape, with the spheroidal shape being preserved by interfacial tension.

Hence we can employ a purely analytical approach by considering the asymptotic limit C<<1. As a result, the magnitude of deviation from sphericity is expected to be O(C) and approximate analytic solution can be obtained. The basic idea is that the drop shape is only slightly nonspherical, and the boundary conditions at the drop interface can be linearlized about the boundary conditions for an exactly spherical drop. This approach is an example of a general technique known as the method of domain perturbation. In the perturbation expansion which follows, we trace the general procedures outlined by Leal [1992], in which the velocity, kinematic and shear stress conditions are satisfied at each order of perturbation, and the deformation of the drop is then calculated using the normal stress balance.

We now proceed formally to the solution of our problem, via a double asymptotic expansion in C and De. Thus,

$$1 \gg C,\ De \gg C^2,\ CDe,\ De^2 \cdots.$$

We may also write down formal expansions for the velocity, pressure and stress fields. For the suspending phase, these are

$$\left.\begin{aligned} &\mathbf{u}=\mathbf{u}^{(0)}+De\,\mathbf{u}^{(De)}+C\mathbf{u}^{(C)}\cdots; \\ &P=P^{(0)}+DeP^{(De)}+CP^{(C)}\cdots; \\ &\mathbf{T}=\mathbf{T}^{(0)}+De\mathbf{T}^{(De)}+C\mathbf{T}^{(C)}\cdots. \end{aligned}\right\} \tag{14}$$

Here, $\mathbf{u}^{(0)}$ is the velocity of a Newtonian, spherical drop in a Newtonian fluid under the action of electric field, whereas $\mathbf{u}^{(De)}$ represents the non-Newtonian contribution to the velocity of a spherical drop, and so on. We can obtain similar expressions for the fluid inside of the drop,

$$\left.\begin{aligned} &\tilde{\mathbf{u}}=\tilde{\mathbf{u}}^{(0)}+De\,\tilde{\mathbf{u}}^{(De)}+C\tilde{\mathbf{u}}^{(C)}\cdots; \\ &\tilde{P}=\tilde{P}^{(0)}+De\tilde{P}^{(De)}+C\tilde{P}^{(C)}\cdots; \\ &\tilde{\mathbf{T}}=\tilde{\mathbf{T}}^{(0)}+De\tilde{\mathbf{T}}^{(De)}+C\tilde{\mathbf{T}}^{(C)}\cdots. \end{aligned}\right\} \tag{15}$$

The drop shape should also be considered in the context of the expansions for the velocity, pressure and stress fields for the suspending phase and the fluid inside of the drop. Since the Newtonian velocity field alone is sufficient to cause deformation of a Newtonian drop at O(C), it is obvious that the O(De) non-Newtonian velocity field will cause deformation at O(CDe), and so on. Hence, on the drop surface,

$$\begin{aligned} F&=r-1-f^{(N)}-f^{(NN)} \\ &=r-1-Cf^{(C)}-CDef^{(CDe)}-C^2f^{(C^2)}-\cdots=0\,, \end{aligned} \tag{16}$$

where $f^{(C)}$, $f^{(CDe)}$, and $f^{(C^2)}$ denote the deformations at O(C), O(CDe), and O(C^2), respectively. The deviation from sphericity is contained in the shape function f(θ). The outer unit normal **n** and the principal radii of curvature $\nabla\cdot n$ are now easily expressed in terms of the shape functions as

$$\begin{aligned} \mathbf{n}&=\nabla F/|\nabla F|=\mathbf{e}_r-C\nabla f^{(C)} \\ &-CDe\nabla f^{(CDe)}-C^2\left[\nabla f^{(C^2)}+\frac{1}{2}(\nabla f^{(C)}\cdot\nabla f^{(C)})e_r\right]-\cdots\,, \end{aligned} \tag{17}$$

and thus, the mean curvature of the drop surface is,

$$\begin{aligned} \nabla\cdot\mathbf{n}&=2-C([2f^{(C)}+\nabla^2 f^{(C)}]-CDe[2f^{(CDe)}+\nabla^2 f^{(CDe)}]) \\ &-C^2[2f^{(C^2)}-2f^{(C)}f^{(C)}+\nabla^2 f^{(C^2)}]-\cdots. \end{aligned} \tag{18}$$

This completes the formulation of problem for a second-order drop subjected to a uniform electric field. In the next section, the governing equations and boundary conditions for both the electric fields and the flow fields at each order are solved by collecting the terms with the same order in C and De.

SMALL DEFORMATION THEORY

The asymptotic expansion procedure for the deformation of a Newtonian fluid drop in an electric field is already known, and thus, it is not necessary to repeat the procedure here [Ajayi, 1978; Ha and Yang, 1995].

The velocity fields, which is correct up to O(C), are given by in terms of stream functions inside and outside the drop, that is,

$$\begin{aligned} \Psi^{(N)}&=(A_2^{(0)}-A_2^{(0)}r^{-2})Q_2(\eta)+C[(A_2^{(C)}+B_2^{(C)}r^{-2})Q_2(\eta) \\ &+(A_4^{(C)}r^{-2}+B_4^{(C)}r^{-4})Q_4(\eta)]+O(C^2)\,, \end{aligned} \tag{19}$$

$$\begin{aligned} \tilde{\Psi}^{(N)}&=(A_2^{(0)}r^5-A_2^{(0)}r^3)Q_2(\eta)+C[\tilde{A}_2^{(C)}r^5+\tilde{B}_2^{(C)}r^3)Q_2(\eta) \\ &+(\tilde{A}_4^{(C)}r^7+\tilde{B}_4^{(C)}r^5)Q_4(\eta)]+O(C^2)\,, \end{aligned} \tag{20}$$

Here, the set of constants A_n, B_n, $\tilde{A}_n$ and $\tilde{B}_n$ must be determined from the appropriate boundary conditions. In (18) and (19), $Q_n(\eta)$, stands for the Gegenbauer polynomials defined by

$$Q_n(\eta)=\int_{-1}^{\eta}P_n(\eta)d\eta\,,$$

in terms of the Legendre polynomial $P_n(\eta)$ of order n with η=cosθ.

After obtaining the electrostatic fields, and then, applying boundary conditions (9), (10), and (11), the following non-zero coefficients which specify the stream functions can be found.

$$A_2^{(0)}=-\frac{9R(1-SR)}{5(2R+1)^2(1+\lambda)}. \tag{21}$$

$$\left.\begin{aligned} &A_2^{(C)}=6\left(\frac{2}{5}\frac{1-R}{1+2R}+\frac{1}{35}\frac{1-\lambda}{1+\lambda}\right)F_2^{(C)}A_2^{(0)};\quad \tilde{A}_2^{(C)}=A_2^{(C)}-\frac{5}{7}F_2^{(C)}A_2^{(0)}; \\ &B_2^{(C)}=-A_2^{(C)}-\frac{46}{21}F_2^{(C)}A_2^{(0)};\qquad \tilde{B}_2^{(C)}=-A_2^{(C)}+\frac{3}{7}F_2^{(C)}A_2^{(0)}. \end{aligned}\right\}$$

$$\left.\begin{aligned} &A_4^{(C)}=\frac{46}{21}F_2^{(C)}A_2^{(0)};\qquad \tilde{A}_4^{(C)}=\frac{4}{3}F_2^{(C)}A_2^{(0)}; \\ &B_4^{(C)}=-\frac{82}{21}F_2^{(C)}A_2^{(0)};\qquad \tilde{B}_4^{(C)}=-\frac{64}{21}F_2^{(C)}A_2^{(0)}. \end{aligned}\right\} \tag{22}$$

Finally, by applying the normal stress balance at the interface, the correction for the shape function which is accurate up to O(C^2) is obtained. The result is

$$f^{(N)}=CF_2^{(C)}P_2(\eta)+C^2[F_0^{(C^2)}+F_2^{(C^2)}P_2(\eta)+F_4^{(C^2)}P_4(\eta)] \tag{23}$$

where

$$F_2^{(C)}=\frac{3}{4(2R+1)^2}\left[(1+R^2-2SR^2)+R(1-SR)\frac{3(2+3\lambda)}{5(1+\lambda)}\right].$$

$$F_0^{(C^2)}=\frac{1}{5}(F_2^{(C)})^2,$$

$$F_0^{(C^2)}=\frac{1}{4}\left[\frac{9F_2^{(C)}}{(2R+1)^2}(1+R^2-2SR^2)\left(\frac{4}{5}\frac{1-R}{1+2R}-\frac{2}{7}\right)+\frac{20}{7}(F_2^{(C)})^2\right.$$

$$-A_2^{(C)}(2+3\lambda)-\frac{1}{7}F_2^{(C)}A_2^{(0)}(-8+13\lambda)\Big],$$

$$F_0^{(C^2)}=\frac{1}{18}\Big[\frac{24}{35}\frac{9F_2^{(C)}}{(2R+1)^2}(1+R^2-2SR^2)+\frac{36}{7}-(F_2^{(C)})^2$$

$$-\frac{2}{35}F_2^{(C)}A_2^{(0)}(88+113\lambda)\Big].$$

The constant $F_0^{(C^2)}$ is included in (22) to ensure that the drop volume remains constant through the deformation.

We consider in this subsection the O(De) problem which represents a first viscoelasticity correction to a drop suspended in a uniform electric field. The electrostatic potentials are also governed by Laplace's equations subject to the boundary conditions (2)-(5). It can be easily seen from (1) and boundary conditions that the electrostatic potentials outside and inside the drop are zero in O(De). This is due to the fact that the electrostatic potentials are obviously independent of the rheological properties of the fluids including the viscoelasticity. Hence, the O(De) problem is reduced to a purely hydrodynamic one induced by an electric field.

For the suspending phase, the equations of motion at O(De) plus the continuity equation are

$$\nabla\cdot\mathbf{T}^{H(De)}=0,\ \nabla\cdot\mathbf{u}^{(De)}=0, \tag{24}$$

in which

$$\mathbf{T}^{H(De)}=-P^{(De)}\mathbf{I}+\tau_{(1)}^{(De)}+\mathbf{T}^{extra},$$

and

$$\mathbf{T}^{extra}=[\tau_{(1)}^{(0)}\cdot\tau_{(1)}^{(0)}+\phi_1\tau_{(2)}^{(0)}].$$

The corresponding equations for the fluid inside the drop are, of course, completely analogous to the expressions (23) for the outside phase, and can be simply expressed by adding the tilde mark to the variables. The continuity of the tangential velocity and the kinematic condition at r=1 are

$$u_\theta^{(De)}=\tilde{u}_\theta^{(De)}, \tag{25}$$

$$u_r^{(De)}=\tilde{u}_r^{(De)}=0. \tag{26}$$

The stress balances can be expressed in terms of the electric and hydrodynamic stressest at O(De) as shown in O(1) problem. The balances for the normal and tangential stresses are

$$T_{r\theta}^{E(De)}-S\tilde{T}_{r\theta}^{E(De)}+\tau_{(1),r\theta}^{(De)}-\lambda\tilde{\tau}_{(1),r\theta}^{(De)}+(\tau_{(1)}^{(De)}\cdot\tau_{(1)}^{(0)})_{r\theta}+\phi_1\tau_{(2),r\theta}^{(0)}$$
$$-\beta[(\tilde{\tau}_{(1)}^{(0)}\cdot\tilde{\tau}_{(1)}^{(0)})_{r\theta}+\tilde{\phi}_1\tilde{\tau}_{(2),r\theta}^{(0)}]=0, \tag{27}$$

$$T_{rr}^{E(De)}-S\tilde{T}_{rr}^{E(De)}-P^{(De)}+\lambda\tilde{P}^{(De)}+\tau_{(1),rr}^{(De)}-\lambda\tilde{\tau}_{(1),rr}^{(De)}+(\tau_{(1)}^{(0)}\cdot\tau_{(1)}^{(0)})_{rr}+\phi_1\tau_{(2),rr}^{(0)}$$
$$-\beta[(\tilde{\tau}_{(1)}^{(0)}\cdot\tilde{\tau}_{(1)}^{(0)})_{rr}+\tilde{\phi}_1\tilde{\tau}_{(2),rr}^{(0)}]=-2f^{(De)}-\nabla^2-f^{(De)}. \tag{28}$$

The parameter β which appears in (26) and (27) represents the ratio of Deborah numbers of the two fluids, i.e.,

$$\beta\equiv(\tilde{D}e/De)\lambda=\tilde{\omega}_3/\omega_3,$$

and is thus independent of the zero-shear-rate viscosity ratio λ. Consequently, for moderate values of λ, both the fluid motions inside and outside the drop contribute to the drop deformation at O(De) if β is of O(1). If β approaches zero or infinity, one of the fluids may be considered Newtonian, and therefore produces no direct contribution at O(De) to the drop deformation.

In the case of the Newtonian fluid, the momentum equation for the creeping flows can be reduced to a homogeneous differential equation for the stream functions as noted from (18) and (19) of the O(1) problem. However, in the presence of the non-Newtonian contribution of the extra stress tensor, the stream functions of O(De) satisfy an inhomogeneous equation of the form

$$E^4\Psi_p^{(De)}=Z(r,\theta), \tag{29}$$

in which the function Z(r, θ) can be easily shown to be

$$Z(r,\theta)=\Big[\frac{\partial}{\partial\theta}(\nabla\cdot\mathbf{T}^{extra})_r-\frac{\partial}{\partial r}[r(\nabla\cdot\mathbf{T}^{extra})_\theta]\Big]\sin\theta,$$

and a similar expression for the stream function $\tilde{\Psi}^{(De)}$ for the drop phase can be obtained in terms of the non-Newtonian extra stress tensor.

The particular solutions of the above equations can be determined with the aids of the O(1) solutions and the results are

$$\Psi_p^{(De)}=\frac{1}{7}(-4r^{-3}+2r^{-5})(1+\phi_1)(A_2^{(0)})^2P_2(\eta)+\frac{1}{7}(18r^{-3}+12r^{-5})$$
$$(1+\phi_1)(A_2^{(0)})^2P_4(\eta),$$
$$\tilde{\Psi}_p^{(De)}=0. \tag{30}$$

The solutions of the homogeneous parts

$$E^4\Psi_h^{(De)}=0,\ E^4\tilde{\Psi}_h^{(De)}=0 \tag{31}$$

have the same forms as those of the Newtonian fluid case which appear in (18) and (19). The unknown coefficients contained in the O(De) stream function can be determined from (24), (25), and (26), with the leading order results. The stream functions $\Psi^{(De)}$ and $\tilde{\Psi}^{(De)}$ can be expressed as (18) and (19) with the nonzero coefficients $A_n^{(De)}$, $B_n^{(De)}$, $\tilde{A}_n^{(De)}$ and $\tilde{B}_n^{(De)}$ (n=2, 4), which are given below :

$$A_2^{(De)}=\tilde{A}_2^{(De)}+\frac{1}{7}(1+\phi_1)(A_2^{(0)})^2,$$

$$B_2^{(De)}=-\tilde{A}_2^{(De)}+\frac{1}{7}(1+\phi_1)(A_2^{(0)})^2,$$

$$\tilde{B}_2^{(De)}=-\tilde{A}_2^{(De)},$$

$$A_2^{(De)}=\tilde{A}_4^{(De)}-\frac{3}{7}(1+\phi_1)(A_2^{(0)})^2,$$

$$B_4^{(De)}=-\tilde{A}_4^{(De)}-\frac{27}{7}(1+\phi_1)(A_2^{(0)})^2,$$

$$\tilde{B}_4^{(De)}=-\tilde{A}_4^{(De)},$$

$$\tilde{A}_2^{(De)}=\frac{(A_2^{(0)})^2}{35(1+\lambda)}[39(1+\phi_1)+30\beta(1+\tilde{\phi}_1)],$$

$$\tilde{A}_4^{(De)}=\frac{(A_2^{(0)})^2}{63(1+\lambda)}[9(1+\phi_1)+40(1+\beta)-100(\phi_1+\beta\tilde{\phi}_1)]. \tag{32}$$

From the normal stress balance (27), we can determine the correction for the drop shape induced by non-Newtonian contributions. The result is

$$F^{(NN)}=CDef^{(CDe)}=CDe[F_2^{(CDe)}P_2(\eta)+F_4^{(CDe)}P_4(\eta),] \tag{33}$$

where

$$F_2^{(CDe)}=-\frac{2+3\lambda}{4}\tilde{A}_2^{(De)}+\frac{1}{21}\Big[\frac{73}{2}(1-\beta)+55(\phi_1-\beta\tilde{\phi}_1)\Big](A_2^{(0)})^2,$$

$$F_2^{(CDe)} = -\frac{4+5\lambda}{60}\tilde{A}_4^{(De)} + \frac{2}{315}[22(1-\beta) - 52(\phi_1 - \beta\tilde{\phi}_1)]$$
$$(A_2^{(0)})^2 + \frac{(A_2^{(0)})^2}{210}(1+\phi_1).$$

LINEAR STABILITY OF THE STEADY-STATE SHAPE

In the preceding sections, we have determined the steady-state shape function f up to $O(C^2)$:

$$f = f^{(N)} + f^{(NN)} = Cf^{(C)} + CDef^{(CDe)} + C^2 f^{(C^2)}. \tag{34}$$

The most important feature of the present solution is that it exhibits multiple steady-state solutions for electric capillary numbers below a critical value, but no steady-state solution beyond the critical value. Kang and Leal [1988] and Yang et al. [1993], and more recently, Ha and Yang [1995] have shown that in the small deformation limit, the critical electric capillary number which separates the stable and unstable steady-state solution branches, can be determined without solving the unsteady disturbance problem. According to their perturbation theory, an estimation of the critical electric capillary number can be carried out by transforming the C-perturbation into a P_2-perturbation, in which the small parameter is the magnitude of the $P_2(\eta)$ mode of deformation

$$\zeta \equiv \int_{-1}^{1} fP_2(\eta)d\eta, \tag{35}$$

instead of C. Then, the electric capillary number as a function of ζ is given by

$$C = c_1\zeta + c_2\zeta^2, \tag{36}$$

in which

$$c_1 = \frac{5}{2F_2^{(C)}},\quad c_2 = -\frac{25[F_2^{(C^2)} + \delta F_2^{(CDe)}]}{4(Fe^{(C)})^3}.$$

The expansion in terms of ζ is equivalent to interchanging the dependent and independent variables from ζ and C, respectively, to C and ζ. By the transformation, the limit point, which appears as a singular point on the stable solution branch at a critical electric capillary number C_c, is converted to a regular point. Thus, we can determine the electric capillary number C as a function of ζ for both the unstable and stable branches of the solution curve, as shown in Fig. 2. The critical electric capillary number, which occurs when $\partial C/\partial\zeta = 0$, can thus be estimated as

$$C_c = \frac{F_2^{(C)}}{4[F_2^{(C^2)} + \delta F_2^{(CDe)}]} \text{ at } \zeta_c = \frac{(F_2^{(C)})^2}{5[F_2^{(C^2)} + \delta F_2^{(CDe)}]}. \tag{37}$$

It can be easily shown that the stability of the solution branch is exchanged at the critical point estimated above.

In a rigorous linear stability analysis, we must consider an arbitrarily small three-dimensional disturbance to a steady-state shape and examine whether the drop will return to the steady-state shape or continue to either deform or break up, by solving the corresponding unsteady problem for the disturbance. The steady-state drop shape can be always expressed in terms of spherical surface harmonics. However, for an axisymmetric case, the spherical surface harmonics can be related to the Legendre polynomials, $P_n(\eta)$. Thus, the present solution for the steady-state drop shape is given by (22), and (32) in terms of $P_n(\eta)$. The unsteady problem in quasi-steady Stokes flow for the three-dimensional disturbance of O(C) to the steady shape can be constructed by considering the kinematic condition

$$u_i n_i = \tilde{u}_i n_i = Cq_i q_m \frac{\partial f_{im}}{\partial t}, \tag{38}$$

in which $q_i = x_i/r$ $(r = (x_i x_i)^{1/2})$. The corresponding unsteady problem was formulated by Barthes-Biesel and Acrivos [1973]. For the three-dimensional disturbances of O(C), $f_{12} = f_{21}$, $f_{11} = f_{22} = -1/2\, f_{33}$, and $f_{13} = f_{31} = f_{32} = f_{23}$, due to the axisymmetry of the problem about the x_3-axis. Thus, we have to consider three simultaneous unsteady problems for f_{33}, f_{12}, and f_{13}:

$$C\frac{\partial f_{ij}}{\partial t} = h_{ij}(C,\ De,\ F_2^{(C)}, F_2^{(CDe)}, F_4^{(CDe)}, F_0^{(C^2)}, F_2^{(C^2)}, F_4^{(C^2)})f_{ij}, \tag{39}$$

in which the detailed formula for the parameter F_n at each order is given in the previous section. The unsteady solutions will decay and the steady drop shape is stable only if all of the coefficient functions h_{33}, h_{12}, and h_{13} are always negative. It can be shown straightforwardly that the stability condition is satisfied only when

$$C < C_c = \frac{F_2^{(C)}}{4[F_2^{(C^2)} + \delta F_2^{(CDe)}]}, \tag{40}$$

which is identical to the critical electric capillary number for the stability of $P_2(\eta)$ mode. Consequently, up to the O(C) disturbance to the steady-state shape which is correct to $O(C^2)$, the stability condition can be determined by estimating the limit point for the existence of the steady state in P_2-perturbations.

Although instability may be manifested by the amplification of nonaxisymmetric as well as axisymmetric deformations, there is a region in which only the axisymmetric instability modes are observed in a uniform electric field [Saville, 1970]. Thus, the present study can afford insights into the phenomena of electrohydrodynamic stability in the region of validity. For example, experiments reported by Taylor [1969] showed that the stability and instability phenomena in an axisymmetric mode have been observed with field strengths in the range 0-6 kVcm^{-1}

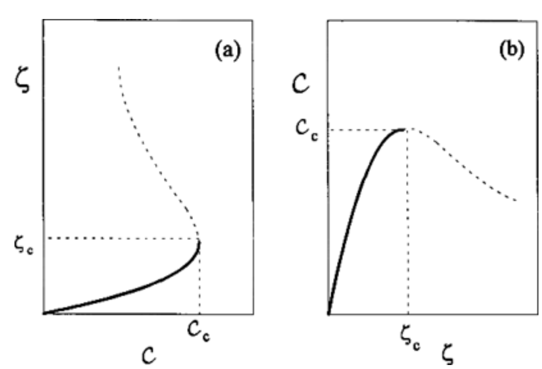

Fig. 2. Representation of stability exchange.
(a) C in terms of ζ; (b) ζ in terms of C

while at somewhat higher field strengths instability is manifested in a nonaxisymmetric form. Of course, the result outlined above also may not be valid for a large deformation problem in which there are significant nonlinear interactions.

DISCUSSION

In the previous sections, we obtained the asymptotic solution correct up to $O(C^2)$ for the steady-state shape of a drop subjected to a uniform electric field. In addition, we also considered the linear stability of the drop by transforming the C-perturbation into the P_2-perturbation. Since the continuous and dispersed fluids are assumed to be non-Newtonian, specifically, second-order fluids, it is expected that the drop behaviors would be different from those of a Newtonian fluid. In this section, we will discuss the results given by the small deformation theory and linear stability analysis. The main purpose of this section is to determine whether the elasticity of the continuous and drop phases stabilizes or destabilizes the drop. To do so, among various parameters we will concentrate on the role of β, the ratio of the normal stress coefficients of two phases, and the Deborah number of the continuous phase.

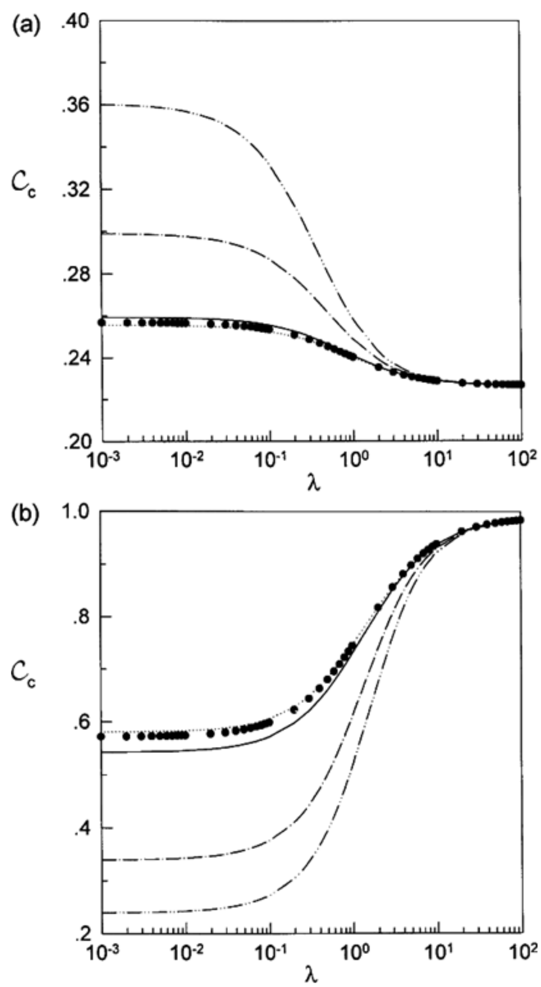

Fig. 3. Critical electric capillary number C_c as a function of the zero-shear-rate viscosity ratio.

Filled circles denote C_c of a Newtonian drop and continuous phases with the same electrical properties. - - - -, β=0.01; ——, β=1, —·—, β=10; —··—, β=20. (a) S=1, R=0.1, δ=1, and $\phi_1=\tilde{\phi}_1=-0.50$. (b) S=1, R=100, δ=0.1 and $\phi_1=\tilde{\phi}_1=-0.50$.

Prior to analysis of the deformation and stability of the drop, we have to estimate the material parameters ϕ_1 and $\tilde{\phi}_1$ of the two fluids. The second normal stress coefficient is not nearly as well studied experimentally as the shear viscosity and first normal stress coefficient. However, the most important point to note about the second normal stress coefficient is that its magnitude is much smaller than that of the first normal stress coefficient. Although there are some disputes on the magnitude of the second normal stress coefficient and even on the sign of its value, it is generally accepted that the magnitude of the second normal stress coefficient ranges from 1% and 20% of that of the first normal stress coefficient. Therefore, in general, it is believed that ϕ_1 and $\tilde{\phi}_1$ should always lie between −0.5 and −0.6, as verified experimentally by Leal [1975]. It is also known that although our knowledge about the second normal stress coefficient is still incomplete, the first normal stress coefficient is sufficient to provide general behaviors of a non-Newtonian drop, especially when the fluids are weakly non-Newtonian. In the present study, we thus fixed both values of ϕ_1 and $\tilde{\phi}_1$ as −0.50 to avoid complexity caused by the nondeterministic parameter.

The estimated critical electric capillary number, obtained from (36) as a function of the zero-shear-rate viscosity ratio λ, is represented in Fig. 3(a) and 3(b) for a prolate and for an oblate spheroid, respectively. Also displayed in these figures is that the critical electric capillary number influenced by the ratio of the normal stress differences β is compared to that of a Newtonian pair in which other parameters, such as the permittivity ratio S and the resistivity ratio R are the same. It can be noted that the effect of non-Newtonian elasticity is diminished as the viscosity ratio increases. However, when both the phases are non-Newtonian, elasticity of the drop phase make the drop either stable or not, depending upon the type of deformation. For example, as the ratio of the normal stress difference ratio β increases the drop becomes more stable for the prolate-type deformation, whereas the trend is reversed for the oblate-type deformation. In addition, for the prolate-type deformation, comparison with the Newtonian pair shows that a non-Newtonian fluid drop in a non-Newtonian fluid is slightly less stable when β is less than unity, and becomes more stable when β increases above unity. In the polymer blending technology, it is a well-known rule of thumb that the drop phase elasticity is expected to reduce the deformation and increase the critical shear rate for the drop breakup, while the matrix elasticity should increase the deformation and decrease the critical shear rates [Elmendorp and Maalcke, 1985].

These somewhat complicated results can be understood by simply considering $F_2^{(CDe)}$ which is the largest, first contribution from the non-Newtonian property and appears in (32). For a moment, let us restrict our attention to the stability of a prolate spheroid. When $F_2^{(CDe)}$ is positive, the drop deforms into a prolate spheroid at O(CDe) causing the critical electric capillary number C_c to decrease. Since we have fixed the value of ϕ_1 and

$\tilde{\phi}_1$ in the present analysis, the sign of $F_2^{(CDe)}$ is determined by the viscosity ratio and β. However, the sign is independent of the electrical properties of the fluids. The velocity field, and consequently the deformation of the drop are considerably influenced by the electrical properties at O(1). However, at O(CDe), the behavior of drop is determined solely by the non-Newtonian rheological properties of the fluids. In Fig. 4, the effect of λ and β on the sign of $F_2^{(CDe)}$ is reproduced. From this figure, it can be seen why the drop is stabilized as β increases and less stable when β is smaller than unity. It can be also noticed from this figure that the non-Newtonian contribution is vanished, and thus, no deformation occurs at O(CDe) for a certain combination of β and λ. In this special case, the drop behaves like a Newtonian drop although the drop possesses non-Newtonian properties, i.e., non-zero normal stress difference. This stability diagram is depicted in Fig. 5, in which the contours of $F_2^{(CDe)}=0$ are plotted for various combinations of ϕ_1 and $\tilde{\phi}_1$. Above a given contour line, $F_2^{(CDe)}<0$ the non-Newtonian contribution makes the drop stable. On the other hand, below the contour line, the viscoelasticity acts in the opposite way and the drop becomes less stable.

The Deborah number De of a continuous phase is also an important parameter for the stability of a non-Newtonian drop. As discussed previously, De is linearly proportional to the electric capillary number and the proportionality constant δ is determined by the rheological properties of the fluid. As δ (or equivalently, De) increases, the drop becomes more stable in the prolate-type deformation, which is illustrated in Fig. 6. The stability of an oblate spheroid can be explained by a similar consideration of $F_2^{(CDe)}$ in terms of β and De.

Finally, the effect of the resistivity ratio R on the stability of a drop is shown in Fig. 7 in which both the viscosity and permittivity ratios are fixed at 0.001 and at unity, respectively. Due to the fact that the permittivity ratio S is unity, the drop remains always stable if the resistivity ratio R is unity. The effect of

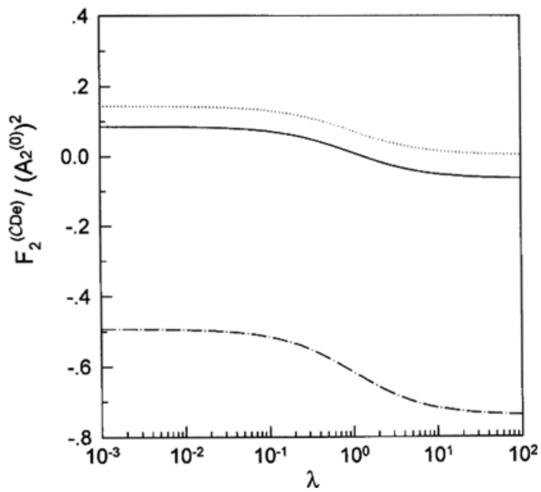

Fig. 4. $F_2^{(CDe)}/(A_2^{(0)})^2$ in terms of the zero-shear-rate viscosity ratio, λ.
$\phi_1=\tilde{\phi}_1=-0.50$. - - - -, β=0.01; ——, β=0.1; —·—, β=1.

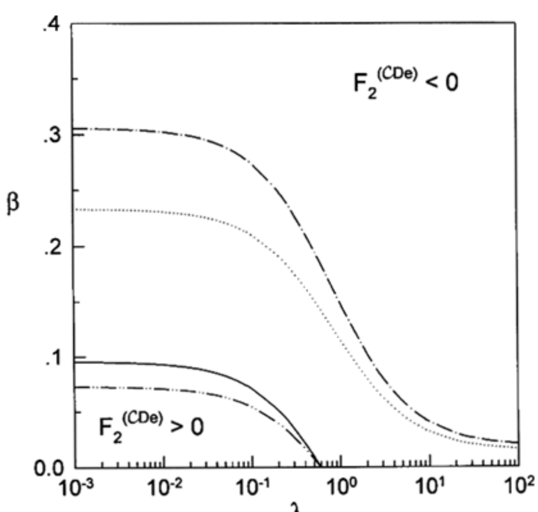

Fig. 5. Contours of $F^{(CDe)}=0$ representing the non-Newtonian contribution to the drop stability.
- - - -, $\phi_1=\tilde{\phi}_1=-0.5$; ——, $\phi_1=\tilde{\phi}_1=-0.55$; —·—, $\phi_1=-0.5$, and $\tilde{\phi}_1=-0.55$, —··—, $\phi_1=-0.55$ and $\tilde{\phi}_1=-0.50$.

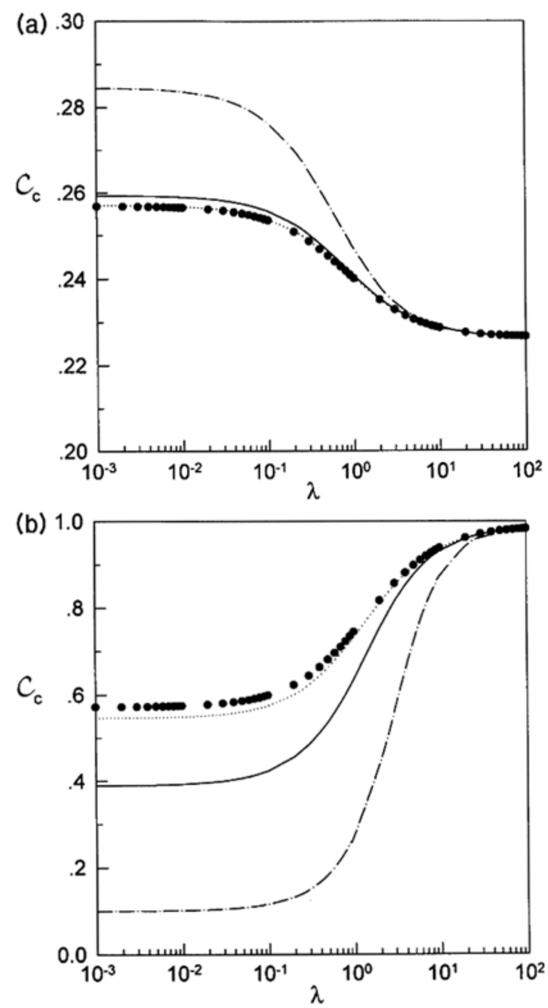

Fig. 6. Effect of Deborah number on C_c.
(a) S=1, R=0.1, β=1 and $\phi_1=\tilde{\phi}_1=-0.50$, (b) S=1, R=100, β=1 and $\phi_1=\tilde{\phi}_1=-0.50$: - - - -, δ=0.1; ——, δ=1, —·—, δ=10.

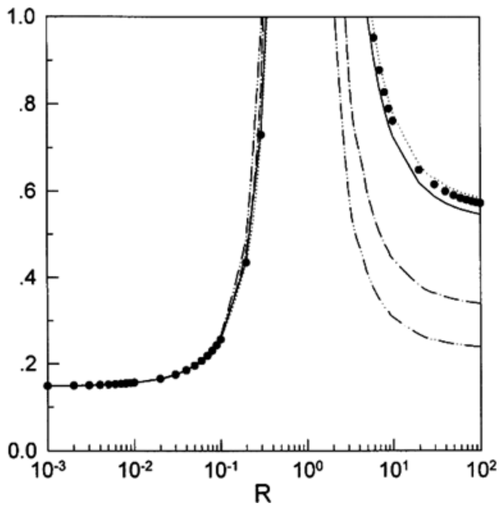

Fig. 7. Estimation of C_c in terms of the resistivity ratio R.
S=1, λ=0.001, δ=0.1 and $\phi_1=\tilde{\phi}_1=-0.50$. - - - -, β=0.01; —, β=1; –-–, β=10; –·–, β=20.

elasticity is more appreciable for an oblate spheroid compared to the case of a prolate spheroid for which the influence of elasticity is small. According to the results of linear stability analysis carried out in the present study, C_c is very weak function of R provided that the resistivities of two phases are quite different.

CONCLUSIONS

In the present study, we investigated one of the general problems concerned with liquid-liquid dispersion; more specifically, we studied the effect of non-Newtonian properties on the deformation and breakup of a droplet in a uniform electric field from theoretical point of view. In order to obtain an analytic solution for the drop shape, the fluids were simply assumed as second-order fluids. The inherent nonlinearity of the free boundary problem was removed by the method of domain perturbation and a double asymptotic expansion in terms of C and De. The stability of the steady-state drop shape was studied by transforming the C-perturbation into a P_2-pertubation, and thus, the critical electric capillary number, which separates the stable and unstable steady-state solution branches could be determined.

The theoretical approach suggested that the non-Newtonian contributions made the drop either more stable or unstable. As the normal stress coefficient of the drop phase increased, the stability of the drop was enhanced for a prolate spheroid. However, for an oblate drop, the drop stability was deteriorated significantly as the normal stress difference coefficient of the drop phase increased. Non-Newtonian contributions to the drop deformation became small as the viscosity ratio increased. The small deformation theory showed that only rheological properties of the fluids were involved in determining the type (i.e., prolate or oblate) of deformations at O(CDe).

REFERENCES

Ajayi, O. O., "A Note on Taylor's Electrohydrodynamic Theory," *Proc. R. Soc. Lond. A*, **364**, 499 (1978).

Arp, P. A., Foister, R. T. and Mason, S. G., "Some Electrohydrodynamic Effects in Fluid Dispersions," *Adv. Colloid Interface Sci.*, **12**, 295 (1980).

Barthes-Biesel, D. and Acrivos, A., "Deformation and Burst of a Liquid Droplet Freely Suspended in a Linear Shear Field," *J. Fluid Mech.*, **61**, 1 (1973).

Basaran, O. A. and Scriven, L. E., "Axisymmetric Shapes and Stability of Charged Drops in an External Electric Field," *Phys. Fluids A1*, 799 (1989).

Bird, R. B., Armstrong, R. C. and Hassager, O., "Dynamics of Polymeric Liquids," 2nd ed., John Wiley, New York (1987).

Elmendorp, J. J. and Maalcke, R. J., "A Study on Polymer Blending Microrheology : Part I," *Polym. Eng. Sci.*, **25**, 1041 (1985).

Garton, C. G. and Krasucki, Z., "Bubbles in Insulating Liquids : Stability in an Electric Field," *Proc. R. Soc. Lond.*, **A280**, 211 (1964).

Ha, J.-W. and Yang, S.-M., "Effects of Surfactant on the Deformation and Stability of a Drop in a Viscous Fluid in an Electric Field," *J. Colloid Interface Sci.*, **175**, 369 (1995).

Ha, J.-W. and Yang, S.-M., "Effect of Nonionic Surfactant on the Deformation and Breakup of a Drop in an Electric Field," *J. Colloid Interface Sci.*, **206**, 195 (1998).

Ha, J.-W. and Yang, S.-M., "Fluid Dynamics of a Double Emulsion Droplet in an Electric Field," *Phys. Fluids*, **11**, 1029 (1999a).

Ha, J.-W. and Yang, S.-M., "Breakup of a Multiple Emulsion Drop in a Uniform Electric Field," *Colloid Interface Sci.*, **213**, 92 (1999b).

Kang, I. S. and Leal, L. G., "Small-amplitude Perturbations of Shape for a Nearly Spherical Bubble in an Inviscid Straining Flow (Steady Shape and Oscillatory Motion)," *J. Fluid Mech.*, **187**, 231 (1988).

Leal, L. G., "The Slow Motion of Slender Rod-like Particles in a Second-order Fluid," *J. Fluid Mech.*, **69**, 305 (1975).

Leal, L. G., "Laminar Flow and Convective Transport Processes-scaling Principles and Asymptotic Analysis," Butterworth-Heinemann, Boston (1992).

Mackay, M. E. and Boger, D. V., "An Explanation of the Rheological Properties of Boger Fluids," *J. Non-Newtonian Fluid Mech.*, **22**, 235 (1987).

Melcher, J. R. and Taylor, G. I., "Electrohydrodynamics: a Review of the Role of Interfacial Shear Stresses," *Ann. Rev. Fluid Mech.*, **1**, 111 (1969).

Miksis, M. J., "Shape of a Drop in an Electric Field," *Phys. Fluids*, **24**, 1967 (1981).

Moriya, S., Adachi, K. and Kotaka, T., "Deformation of Droplets Suspended in Viscous Media in an Electric Field: I. Rate of Deformation, II. Burst Behavior," *Langmuir*, **2**, 155 (1986).

Saville, D. A., "Electrohydrodynamic Stability : Fluid Cylinder in Longitudinal Electric Fields," *Phys. Fluids*, **13**, 2987 (1970).

Saville, D. A., "Electrohydrodynamics : the Taylor-Melcher Leaky Dielectric Model," *Annu. Rev. Fluid Mech.*, **29**, 27 (1997).

Taylor, G. I., "Disintegration of Water Drops in an Electric Field," *Proc. R. Soc. Lond.*, **A291**, 159 (1964).

Taylor, G. I., "Electrically Driven Jets," *Proc. R. Soc. Lond.*, **A313**, 453 (1969)

Taylor, G. I., "Studies in Electrohydrodynamics. I. The Circulation

Produced in a Drop by an Electric Field," *Proc. R. Soc. Lond.*, **A291**, 159 (1966).

Torza, S., Cox, R. G. and Mason, S. G., "Electrohydrodynamic Deformation and Burst of Liquid Drops," *Phil. Trans. R. Soc. Lond.*, **269**, 259 (1971).

Tam, K. C. and Tiu, C., "A Low Viscosity, Highly Elastic Ideal Fluid," *J. Non-Newtonian Fluid Mech.*, **31**, 163 (1989).

Venugopal, G. and Krause, S., "Development of Phase Morphologies of Poly(methyl methacrylate)-polystyrene-toluene Mixtures in Electric Fields," *Macromolecules*, **25**, 4626 (1992).

Vizika, O. and Saville, D. A. 1992, "The Electrohydrodynamic Deformation of Drops Suspended in Liquids in Steady and Oscillatory Electric Fields," *J. Fluid Mech.*, **239**, 1 (1992).

Yang, S. M., Feng, Z. C. and Leal, L. G., "Nonlinear Effects in the Dynamics of Shape and Volume Oscillations for a Gas Bubble in an External Flow," *J. Fluid Mech.*, **247**, 417 (1993).

Xi, K. and Krause, S., "Droplet Deformation and Structure Formation in Two-phase Polymer/polymer/toluene Mixtures in an Electric Field," *Macromolecules*, **31**, 3974 (1998).