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Mass transfer around a slender drop in a nonlinear extensional flow

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Abstract: Mass transfer around a slender drop in a nonlinear extensional and creeping flow is theoretically studied. The fluid mechanics problem is governed by three dimensionless parameters: The capillary number ($Ca \gg 1$), the viscosity ratio ($\lambda \ll 1$), and the nonlinear intensity of the flow ($|E| \ll 1$). The transfer of mass around such a drop is studied for the two asymptotic cases of large and zero Peclet numbers (Pe). The results show that as the capillary number increases, the drop becomes longer, thinner, its surface area increases, leading to larger mass transfer rates, especially at large Peclet numbers, since then convection contributes to the overall mass transfer as well. Taking a slender inviscid drop ($\lambda = 0$) in a linear extensional flow ($E = 0$) as our reference case, we find that the addition of nonlinear effects to the flow sometimes increases ($E\lambda^{-1}Ca^{-2} < 64/9$) and sometimes decreases ($E\lambda^{-1}Ca^{-2} > 64/9$) the rate of mass transfer.

Keywords: Creeping Flow, Drop, Fluid Mechanics, Mass Transfer, Nonlinear Flows

1 Introduction

Many industrial products such as foods, paints, pharmaceuticals and cosmetics are made of emulsions in which a drop is embedded in a liquid. These emulsions are generally processed in mixing devices generating shear, extensional or even more complicated flows. The type of flow as well as the transfer of mass between such a drop and the continuous liquid, were the subjects of many scientific studies since they affect the properties of the emulsion.

Let an initially non-buoyant spherical drop of radius a and viscosity μ_{in} be placed in another liquid having viscosity μ . When the external liquid is subjected to shear

or extensional flow, the drop will deform. For Newtonian fluids under creeping flow conditions, the fluid mechanics problem is governed by two dimensionless parameters: the capillary number $Ca = \mu Aa/\sigma$ and the viscosity ratio $\lambda = \mu_{in}/\mu$, with A being the shear or extension rate and σ the surface tension. While the majority of the literature is dedicated to small deformations ($Ca \ll 1$), this report is dedicated to slender drops only; these are obtained at large capillary numbers ($Ca \gg 1$) and small viscosity ratios ($\lambda \ll 1$). More information on this fundamental research area and its industrial applications, can be found in reviews by Rallison [1], Stone [2], and Briscoe et al. [3]; and in books by Clift et. al [4], Levitskiy and Shulman [5], Sadhal et al. ([6], Zapryanov and Tabakova [7], and Chhabra [8].

Slender drops in shear and extensional flows were reported long time ago in the classical paper of Taylor [9]. Thirty years later, Taylor [10] was the first to suggest an approximated model for the deformation and breakup of a slender drop in an axisymmetric linear extensional creeping flow, a case where the cross section of the drop is circular. In the following years, the theory, that was refined by Buckmaster [11, 12], Acrivos and Lo [13] and others, predicts that the slender drop has a parabolic radius profile and that the ends of the drop are pointed. As the capillary number increases, the steady stable drop becomes thinner, longer, its surface area increases, until the breakup point is reached at a critical value of $Ca\lambda^{1/6} = 0.148$. It follows, that a slender inviscid drop or a bubble ($\lambda = 0$) cannot be broken.

The experiments conducted by Taylor [9] in his four-roller apparatus cannot produce a three-dimensional axisymmetric extensional flow, but rather a two-dimensional extensional flow. Therefore, Hinch and Acrivos [14] considered a slender drop in a two-dimensional linear extensional creeping flow, a case where the drop cross-section is not circular. They found that the cross-section of the drop is an approximate ellipse with an axis ratio of 1.5, and that the deformation and breakup criteria were almost identical to the axisymmetric findings. Antanovskii [15], in his study on the formation of a pointed drop, mentions that the flow, in Taylor's four-roller apparatus, is better described by a nonlinear two-dimensional extensional flow rather than by the linear flow.

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A slender drop in a nonlinear axisymmetric extensional creeping flow was treated by Sherwood [16]. The theory that was recently reviewed and expanded by Favelukis [17] suggest that apart from the capillary number ($Ca \gg 1$) and the viscosity ratio ($\lambda \ll 1$), the fluid mechanics problem is also governed by the nonlinear intensity of the flow ($|E| \ll 1$), where $E = Ba^2/A$ and B is coefficient connected to the nonlinear velocity term defined in Eq. (1). Contrary to linear extensional flow ($E = 0$), where an inviscid drop ($\lambda = 0$) or a bubble cannot be broken, the addition of nonlinear terms to the external flow ($E \neq 0$) can cause the bubble to break. Similar to the linear flow, in the nonlinear flow the end shape of the slender drop is also pointed. Finally, three types of breakup mechanisms were discovered: a center pinching mode, indefinitely elongation and mechanism that remind us of tip-streaming where a cusp is developed at the end of the drop.

So far, our discussion has been limited to the fluid mechanics problem which can provide us, for example, with the disturbed velocity profile, the shape and the surface area of the slender drop. Mass transfer between such a drop and the external liquid is proportional to the surface area of the drop. It follows that slender drops, with their extremely large surface area, when compared to that of a spherical drop having the same volume, are excellent candidates for mass transfer operations. For example, in the polymer processing industry (Tadmor and Gogos [18]), we find it in polymer melt devolatilization (Albalak [19]) and in the production of polymeric foam materials (Lee et al. [20]; Lee and Park [21]).

The continuity equation for the solute in a binary mixture is governed by the Peclet number, the ratio of convection to diffusion, defined in our problem as: $Pe = Aa^2/D$, with D being the diffusion coefficient. When $Pe = \infty$, convection is large when compared to diffusion, and the well known thin concentration boundary layer approximation can be applied. On the other hand, if $Pe = 0$ convection is absent, and mass transfer is by diffusion only with a thick concentration boundary layer.

Mass transfer around slender drops in an axisymmetric linear extensional flow, for the two limits of the Peclet number, are summarized in Favelukis [22, 23]. For both asymptotic limits of the Peclet number, we found that as the elongation rate or the capillary number increases, the drop becomes thinner, longer and its surface area increases, leading to larger mass transfer rates. Clearly that higher mass transfer rates are obtained at large Peclet numbers since then convection is also present.

Mixing devices used to process foams or emulsions cannot generally produce linear flows but rather more complicated flows which may include nonlinear terms to

the velocity field. In Sherwood [16] and Favelukis [17], the effects of the nonlinear terms on the fluid mechanics problem were investigated revealing new phenomena and it is anticipated that new and interesting results will be discovered in the mass transfer problem as well. Thus, it is the purpose of this theoretical study to explore the mass transfer around a slender drop in a nonlinear extensional creeping flow.

2 Fluid mechanics

In Fig. 1 we consider a slender drop positioned at the origin of a cylindrical coordinate system and subjected to a nonlinear axisymmetric extensional flow. The drop has a local radius $R(z)$ and a half-length L such that $R/L \ll 1$. Far away from the drop, we assume the nonlinear undisturbed motion suggested by Sherwood [16]:

$$v_r = -\frac{1}{2}Ar - \frac{3}{2}Brz^2, \quad v_z = Az + Bz^3 \quad (1)$$

We define this nonlinear axisymmetric extensional flow with $A > 0$, however B can obtain both positive or negative values. If $B = 0$, the familiar linear extensional flow is recovered and A becomes the extension rate.

Outside and near the drop as well as on the surface of the drop, and following Acrivos and Lo [13], Favelukis [17] developed the velocity components of the steady disturbed motion:

$$v_r = A \left\{ - \left(1 + 3 \frac{B}{A} z^2 \right) \frac{r}{2} + \frac{R}{r} \left[\left(1 + 3 \frac{B}{A} z^2 \right) \frac{R}{2} + \left(1 + \frac{B}{A} z^2 \right) z \frac{dR}{dz} \right] \right\}, \quad v_z = Az \left(1 + \frac{B}{A} z^2 \right) \quad (2)$$

Note that the slender body approximation suggests that the axial component of the velocity is the same for both the disturbed and the undisturbed motion. The radial disturbed velocity can be found from the continuity equation and the kinematic condition stating that for a stationary drop the normal surface velocity equals zero.

Fig. 1: A slender drop in a nonlinear extensional flow: $R(z)$ is the local radius and L is the half-length of the drop

Assuming incompressible Newtonian fluids under creeping flow conditions (both inside and outside the drop), the fluid mechanics problem is governed by three dimensionless parameters: the capillary number, the viscosity ratio, and the nonlinear intensity of the flow, which contrary to the other two parameters, it can be positive or negative:

$$Ca = \frac{\mu A a}{\sigma}, \lambda = \frac{\mu_{in}}{\mu}, E = \frac{B a^2}{A} \quad (3)$$

In the above definitions, μ and μ_{in} are the viscosities of the external and internal fluid respectively, a is the equivalent radius (the radius of an equal volume spherical drop) and σ is the surface tension. An order of magnitude analysis suggests that slender drops ($R/L \ll 1$) must obey the following conditions: $Ca^3 \gg 1$, $\lambda^{1/2} \ll 1$ and $|E|^{3/4} \ll 1$, suggesting large capillary numbers, small viscosity ratios and a small intensity of the nonlinear extensional flow. A further exploration of governing equations reveals that the number of dimensionless governing parameters can be reduced from three to two having the order of magnitude of one:

$$F = Ca \lambda^{1/6}, G = Ca^4 E \quad (4)$$

And we shall name F as the positive viscous strength of the flow and G as the nonlinear strength of the flow which can be positive or negative.

We now introduce a few dimensionless drop shape parameters, these are the radius at the center and at the end of the drop, the volume of the drop which is conserved before and after deformation, and the surface area of the drop (S):

$$R^*(0) = \frac{1}{2\nu Ca}, R^*(L^*) = 0 \quad (5)$$

$$\int_0^{L^*} R^{*2} dz^* = \frac{2}{3} \quad (6)$$

$$S^* = \frac{S}{4\pi a^2} = \int_0^{L^*} R^* dz^* \quad (7)$$

In the above equations, all the lengths were made dimensionless with respect to the equivalent radius (a). In the first Eq. (5) the dimensionless parameter ν is related to the internal steady pressure at the center of the drop: $\nu = -1 + P^*(0)/2$, with $P^*(0) = P(0)/(\mu A)$.

Favelukis [17] explored the governing equations near the center and close to the end of the drop, performed a stability analysis and draw the following steady-state conclusions. Contrary to the linear extensional flow case (G

= 0), in which the local radius decreases monotonically (for $z \geq 0$), in the nonlinear case ($G \neq 0$), two possible shapes exist. Steady shapes (stable or unstable) with a monotonically decreasing local radius, and steady shapes (unstable) where the local radius of the drop obtains a maximum (apart from the one at the center). Furthermore, steady slender drops with or without nonlinear effects, have pointed ends.

2.1 An inviscid drop ($\lambda = 0$)

For the case of an inviscid drop or a bubble, $F = 0$, the shape of the drop can be constructed as a power series (Favelukis [17]):

$$R^* = \frac{1}{Ca} \sum_{k=0}^{\infty} a_k \eta^k; \eta = Ez^{*2} \quad (8)$$

Where the first coefficients are given by:

$$a_0 = \frac{1}{2\nu}, a_1 = \frac{9}{4\nu(\nu-2)}, a_2 = \frac{117}{8\nu(\nu-2)(\nu-4)} \quad (9)$$

which can be expressed in a general form as:

$$a_k = \frac{\prod_{j=0}^{k-1} (2j+9/2)}{2 \prod_{j=0}^k (\nu-2j)} \quad (10)$$

Note that the specific case of $j = 0$, the numerator in the last equation is defined as 1. Eq.(10) has singular points at $\nu = 0, 2, 4, 6, \dots$ suggesting a multiple branch solution depending on ν .

The solid line in Fig. 2(a) shows the deformation curve ($0 < \nu < 4$) for a drop with a monotonically decreasing local radius ($z \geq 0$). It starts on the right hand side where ν is small, G is large and L^*/Ca^2 is small. As ν increases, G decreases and L^*/Ca^2 increases. When $G = 0$, the inviscid drop in linear extensional creeping flow is obtained ($\nu = 2$), which according to Acrivos and Lo [13] it is stable and cannot be broken. With a further increase of ν , the deformation curve enters a region where $G < 0$, which is enlarged in Fig. 2(b). The curve turns back at the bifurcation turning point ($\nu = 2.51$) and terminates at $\nu = 4$, $G = -0$ and $L^*/Ca^2 = 60.0$.

The stability analysis performed by Favelukis [17] shows that steady stable shapes are located at the lower branch up to the bifurcation turning point at $\nu = 2.51$, while steady unstable drops are located at the upper branch and in other branches having higher values of ν which are not shown in Fig. 2. The bifurcation turning point is the breakup point, due to fracture as opposed to tip-streaming, located at: $\nu = 2.51$, $G = -9.62 \cdot 10^{-5}$ and L^*/Ca^2

Fig. 2: The deformation curve for a bubble or an inviscid drop ($\lambda = 0$): (a) general view; (b) turning point zone. Solid line: stable drops at lower branch, unstable drops at upper branch, breakup point at turning point ($v = 2.51$); dotted line: approximated solution. Filled circles represent different values of v .

$= 28.7$. Contrary to the linear case ($G = 0$), where the inviscid drop is always stable and breakup is not possible, the addition of nonlinear terms ($G \neq 0$), can cause the bubble to break. There is another family of solutions, not shown in Fig. 2, belonging to steady unstable drops covering the range $2 < v < 4$ with positive values of G . This family suggests strange and unstable drops where the local radius has a maximum (apart from the one at the center) and therefore will not be treated in this report.

A much simpler and elegant approximated solution, described in Favelukis [17], can be obtained by considering the first two terms of Eq. (8):

$$R^* = \frac{1}{2vCa} \left[1 - \left(\frac{z^*}{L^*} \right)^2 \right], L^* = 5v^2 Ca^2, S^* = \frac{5}{3} vCa \quad (11)$$

$$G = \frac{2(2-v)}{225v^4} \quad (12)$$

Equation (11) represents a parabolic radius profile and can only describe the shape corresponding to a monotonically decreasing local radius ($z \geq 0$). The two-term approximation is described by the dotted line in Fig. 2 and its bifurcation turning point is positioned at: $v = 8/3 = 2.67$, $G = -1.17 \cdot 10^{-4}$ and $L^*/Ca^2 = 35.6$. When the extensional flow is linear ($G = 0$), $v = 2$, $L^*/Ca^2 = 20$, the approximate Eq. (11) is also the exact solution.

2.2 A viscous drop ($\lambda \neq 0$)

We continue with a viscous drop, $F \neq 0$, and we introduce a dimensionless parameter representing the ratio of the nonlinear strength of the flow to the viscous strength of the flow:

$$H = \frac{G}{F^6} = \frac{E}{\lambda Ca^2} \quad (13)$$

The parameter H covers the complete numerical range: $-\infty < H < +\infty$. When $H = 0$ the linear extensional flow ($G = 0$) is recovered, and $H \rightarrow \pm \infty$ suggests an inviscid drop ($\lambda = 0$) in a nonlinear ($G \neq 0$) extensional flow.

Similar to the inviscid drop case, Favelukis [17] presented a solution to the steady shape of the drop in the form of a power series around the center of the drop:

$$R^* = \frac{1}{Ca} \sum_{k=0}^{\infty} b_k \phi^k, \phi = \lambda Ca^2 z^{*2} \quad (14)$$

where the first coefficient are:

$$b_0 = \frac{1}{2v}, b_1 = \frac{9H - 16v^2}{4v(v-2)}, b_2 = \frac{H[117H - 16v^2(11+v)]}{8v(v-2)(v-4)} \quad (15)$$

suggesting once again, a multiple branch solution with singular points at $v = 0, 2, 4, 6, \dots$ and so on. Again, we shall focus on drops with a monotonically decreasing local radius ($z \geq 0$) having low values of v , since there it is anticipated to locate steady stable shapes.

The deformation curve for viscous drops is described by the solid lines in Fig. 3. The first family of solutions corresponds to $H < 64/9 = 7.11$ (positive or negative) with $L^*/Ca^2 > 20$ and $v > 2$. On the left of the linear extensional case ($H = 0$), we find curves having $H < 0$. These solutions are stable at the lower branch and unstable at the upper branch, with the turning point (filled circle) being the breakup point. To the right of the $H = 0$ case we observe curves with $H > 0$, still under the $H < 64/9$ criterion. Some curves stop after the turning point ($H = 1$) and some stop before the turning point ($H = 5$). Curves with bifurcation turning points are stable at the lower branch and unstable at the upper branch, while lines without a bifurcation turning point are stable everywhere. Table 1 lists some

parameters at breakup point, for this family of solutions having: $H < 64/9$ (positive or negative) having bifurcation turning points.

Next is the family of solutions corresponding to $H > 64/9$, $L^*/Ca^2 < 20$ and $\nu < 2$. The stability analysis shows that these lines are stable everywhere. The last family of solutions corresponding to $H > 64/9$ and $\nu > 2$ is not shown in the figure and will not be treated in our mass transfer studies. As previously mentioned, this family suggests steady unstable strange drops shapes having a maximum, besides the one at the center of the drop.

Fig. 3: The deformation curve of viscous drops ($\lambda \neq 0$), for different values of H . Solid lines: exact solution with filled circles placed at breakup points; dashed lines: approximate solution

Once again, if we take the first two terms of the series expansion in Eq. (14), we obtain an approximate solution representing, once again, a parabolic radius profile:

$$R^* = \frac{1}{2\nu Ca} \left[1 - \left(\frac{z^*}{L^*} \right)^2 \right], L^* = 5\nu^2 Ca^2, S^* = \frac{5}{3} \nu Ca \quad (16)$$

$$F = \frac{2^{1/6}}{5^{1/3} \nu^{2/3}} \left(\frac{\nu - 2}{16\nu^2 - 9H} \right)^{1/6} \quad (17)$$

The two-term approximation, given by Eqs (16)-(17) is described in Fig. 3 by the dashed lines. When the extensional flow is linear ($H = 0$), the approximate solution equals the exact solution. We conclude that the approximate solution, which is represented by a very simple mathematical formula, can be used to replace the exact solution (up to the breakup point) when fast and practical estimations are required.

Figure 4 represents the dimensionless parameter ν as a function of the viscous strength of the flow F , for different values of H , according to the approximate solution given by Eq. (17). Some care needs to be exercised for the case

$H = 5$ which according to the exact shape is always stable but from the two-term approximated shape has a bifurcation turning (breakup) point, see Fig. 3. Table 2 describes some parameters at breakup point, according to the approximate solution, for the family of solutions having: $H < 64/9$ (positive or negative) and bifurcation turning points.

Fig. 4: The parameter ν as a function of the viscous strength of the flow, for different values of H , according to the approximate solution. Filled circles placed at breakup points

3 Mass Transfer

Let the liquid outside the drop be a binary mixture containing a small concentration of a solute. In this section we shall investigate the mass transfer of the solute to or from the slender drop and especially the influence of the nonlinearity of the flow on this process. The continuity equation for a binary mixture in the continuous phase with the assumptions of steady-state, Fickian diffusion, laminar flow, constant total density (dilute solutions) and diffusion coefficient (D), is given by:

$$\nu \cdot \nabla c = D \nabla^2 c \quad (18)$$

Here ν is the disturbed external velocity and c is the solute concentration in the liquid outside the drop. We assume the usual boundary conditions presented in the literature, suggesting a surface concentration c_s , usually dictated by a thermodynamic equilibrium such as Henry's Law, a bulk concentration c_∞ and a fresh liquid at the bulk concentration c_∞ entering the concentration boundary layer. Eq. (18) is governed by the Peclet number, the ratio of convection

Table 1: Parameters at breakup point, according to the exact solution, for the family of solutions with $H < 64/9$ (positive or negative) and bifurcation turning points

H	v	G	F	L^*/Ca^2	S^*/Ca
$-\infty$	2.51	$-9.62 \cdot 10^{-5}$	0	28.7	4.06
-1000	2.51	$-9.53 \cdot 10^{-5}$	0.0676	28.7	4.06
-100	2.50	$-8.78 \cdot 10^{-5}$	0.0978	28.6	4.05
-10	2.44	$-4.95 \cdot 10^{-5}$	0.131	28.4	4.01
0	2.4	0	0.148	28.8	4
+1	2.39	$+1.18 \cdot 10^{-5}$	0.151	29.0	4.00

Table 2: Parameters at breakup point, according to the approximate solution, for the family of solutions with $H < 64/9$ (positive or negative) and bifurcation turning points

H	v	G	F	L^*/Ca^2	S^*/Ca
$-\infty$	2.67	$-1.17 \cdot 10^{-4}$	0	35.6	4.44
-1000	2.66	$-1.16 \cdot 10^{-4}$	0.0698	35.4	4.44
-100	2.62	$-1.04 \cdot 10^{-4}$	0.101	34.4	4.37
-10	2.49	$-5.39 \cdot 10^{-5}$	0.132	31.1	4.16
0	2.4	0	0.148	28.8	4
+1	2.38	$+1.16 \cdot 10^{-5}$	0.150	28.4	3.97

to diffusion:

$$Pe = \frac{Ua}{D} = \frac{Aa^2}{D} \quad (19)$$

Here $U = Aa$ is a characteristic velocity. When $Pe \rightarrow \infty$, convection is large (when compared to diffusion), the thin concentration boundary layer approximation can be applied suggesting that the diffusion normal to the surface is much larger than diffusion parallel to the surface. In the other asymptotic limit, corresponding to $Pe \rightarrow 0$, the governing Eq. (18) reduces to the Laplace equation suggesting mass transfer by diffusion only without convection and with a thick concentration boundary layer.

A summary on mass transfer around slender drops in a linear extensional flow ($G = 0$), for the two limits of the Peclet number, can be found in two recent publications (Favelukis [22, 23]). We now extend these studies to the case where the extensional flow is not linear ($G \neq 0$).

3.1 Large Peclet numbers

A general theory for the steady mass transfer around axisymmetric drops of revolution, at large Peclet numbers, was proposed by Lochiel and Calderbank [24], with the only requirements being the shape of the drop (R) and the tangential surface velocity (v_{zs}). The main result is of course the dimensionless mass transfer rate, the average flux times the surface area of the drop, which after being adapted to the present case reads (Favelukis and

Semiat [25], Favelukis [22, 26]):

$$\bar{S}h \cdot S^* = \frac{\bar{k}a}{D} S^* = \frac{2}{\sqrt{\pi}} \left(\int_0^{L^*} v_{zs}^* R^{*2} dz^* \right)^{1/2} Pe^{1/2} \quad (20)$$

Here $\bar{S}h$ is the average Sherwood number (dimensionless flux), S^* is the dimensionless surface area defined in Eq. (7) and $\bar{k}$ is the average mass transfer coefficient (the ratio of the average surface flux to solute the concentration difference).

The dimensionless tangential surface velocity can be obtained from Eq. (2) and the characteristic velocity (defined in the Peclet number):

$$v_{zs}^* = \frac{v_{zs}}{Aa} = z^* (1 + Ez^{*2}) \quad (21)$$

Substituting the above tangential surface velocity, together with the excellent approximation for the shape of the drop given by Eqs (11) or (16), we find for both inviscid or viscous drops:

$$\bar{S}h \cdot S^* = 5 \sqrt{\frac{2}{3\pi}} \left(\frac{v}{2} \right) \left(1 + \frac{25}{4} G v^4 \right)^{1/2} CaPe^{1/2} \quad (22)$$

For the case of an inviscid drop ($\lambda = 0$), the relation between G and v is obtained by Eq. (12) and for viscous drops ($\lambda \neq 0$) we can relate G , F and H with v via Eqs (13) and (17). The above discussion leads to the following equation, valid for inviscid ($F = 0$) as well as viscous ($F \neq 0$) drops:

$$\bar{S}h \cdot S^* = 5 \sqrt{\frac{2}{3\pi}} \left(\frac{v}{2} \right) \left[1 + \frac{1}{18} (2 - v + 200F^6 v^6) \right]^{1/2} CaPe^{1/2} \quad (23)$$

Figures 5 and 6 describe the dimensionless mass transfer rate as a function of the strength of the flow at large Peclet numbers. Figure 5 represents an inviscid drop where the abscissa is the nonlinear strength of the flow (G), while Fig. 6 describes viscous drops, with this time the viscous strength of the flow (F) being the abscissa. Both figures represent stable steady shapes as we stop the curves at the breakup point. As previously mentioned, the two term approximated shape can be used as an excellent replacement of the exact shape, but up to the breakup point.

For an inviscid drop ($\lambda = 0$), depicted in Fig. 5, the range of the plot corresponds to values of ν below the critical point ($\nu < \nu_{cr}$), see Fig. 2. As ν increases, G decreases and the mass transfer rate increases. Notice to the sharp increase in the rate of mass transfer near the breakup point and its similar behavior like the half-length of the drop presented in Fig. 2. For positive values of G ($\nu < 2$), the mass transfer rate is lower than the linear case ($G = 0$, $\nu = 2$), while negative values of G ($\nu > 2$), represent rates of mass transfer larger than the linear case.

Fig. 5: The mass transfer rate as a function of the nonlinear strength of the flow for a bubble or an inviscid drop ($\lambda = 0$) at $Pe \rightarrow \infty$. Filled circle is the breakup point; empty circle is the linear flow

Next is Fig. 6 corresponding to a viscous drop ($\lambda \neq 0$). First, let us observe the family of solutions corresponding to $H < 64/9 = 7.11$ (positive or negative) which also includes the linear extensional case ($H = 0$). Here all plots represent the stable branch located at $2 < \nu < \nu_{cr}$. We find that as F increases (at constant H), ν increases (see Fig. 4) and the mass transfer rate increases especially near the breakup point. The next family satisfying $\nu < 2$ and $H > 64/9$ predicts lower mass transfer rates than the first family. Here as F increases (at constant H), ν decreases and the rate of mass transfer decreases.

Two general conclusions can be obtained for both inviscid and viscous drops. First, as ν increases, the radius at the center of the drop decreases (see Eq. 5), the length of the drop increases (in order to conserve the volume), the surface area and the tangential surface velocity increase leading to larger mass transfer rates. Second, and taking an inviscid drop in linear extensional flow: $\nu = 2$ or $\overline{Sh} \cdot S^* = 2.30CaPe^{1/2}$ as our reference case ($G = 0$ in Fig. 5 or $F = 0$ in Fig. 6), we find that nonlinear contributions to the flow sometimes increase ($H < 64/9$) and sometimes decrease ($H > 64/9$) the rate of mass transfer.

Fig. 6: The mass transfer rate as a function of the viscous strength of the flow, for a viscous drop ($\lambda \neq 0$), at different values of H and at $Pe \rightarrow \infty$. The filled circles are located at breakup points

3.2 Small Peclet numbers

We now proceed to the other asymptotic limit of $Pe = 0$, suggesting mass transfer by diffusion only, with convection being neglected. For the case of a linear extensional flow ($G = 0$) analytical solutions are available (Favelukis [23]; Favelukis and Chiam [27]) and we shall extend the theory to the present case of a nonlinear ($G \neq 0$) extensional flow.

Making use of the bispherical coordinate system, our previous reports showed that the shape of a spindle has, at first approximation, a parabolic local radius with pointed ends which can be represented by Eqs (11) or (16). Fortunately, the electrostatic capacity of a spindle was solved many years ago by Szegő [28] and Payne [29]. Thus, by applying the analogy between electrostatics and diffusion, both governed by the Laplace equation, we can adapt the results presented in Favelukis [23] to our present physical

situation:

$$\bar{Sh} \cdot S^* = 40 \left(\frac{\nu}{2}\right)^2 Ca^2 \int_0^\infty \frac{K_\alpha(-t_s)}{K_\alpha(t_s) \cosh(\pi\alpha)} d\alpha \quad (24)$$

$$K_\alpha(t) = \frac{2}{\pi} \cosh(\pi\alpha) \int_0^\infty (2 \cosh u + 2t)^{1/2} \cos(\alpha u) du \quad (25)$$

$$\theta_s = \pi - \frac{1}{40(\nu/2)^3 Ca^3} \quad (26)$$

Where $K_\alpha(t)$ is the conal function, $t = \cos \theta$, $t_s = \cos \theta_s$, θ is a bispherical coordinate and θ_s is its value at the surface of the slender spindle ($\theta_s \rightarrow \pi$). The conal function is equivalent to the Legendre function $P_n(t)$ with $n = -1/2 + i\alpha$.

In order to avoid the usage of the cumbersome conal functions, Favelukis [23] showed that diffusion around a slender prolate spheroid (rounded ends) is practically identical to diffusion around a slender drop (pointed ends), given by Eqs (11) or (16), which are the exact solution for the linear flow and represent an excellent approximation for the nonlinear case:

$$\bar{Sh} \cdot S^* = \frac{L^*}{\ln[2L^*/R^*(0)]} = \frac{20(\nu/2)^2 \cdot Ca^2}{\ln[160(\nu/2)^3 \cdot Ca^3]} \quad (27)$$

In Fig. 7 we describe the diffusion around stable slender drops as a function of the capillary number for different values of the parameter ν . First, let us observe the case $\nu = 2$ corresponding to an inviscid drop ($\lambda = 0$) in a linear extensional flow ($G = 0$) and, as before, we shall refer this case as our reference. The first family of solutions, for which $H < 64/9 = 7.11$ (positive or negative) and located at $2 < \nu < \nu_{cr}$ (see Fig. 4), represent higher mass transfer than the reference, with the highest rate obtained for an inviscid drop ($\lambda = 0$) in a nonlinear flow ($G \neq 0$) located according to Eqs (11) – (12) at $\nu_{cr} = 2.67$. The next family at $H > 64/9$ and $\nu < 2$ suggests lower mass transfer rates than the reference case. We conclude that as the capillary number increases or as the parameter ν increases, the drop becomes thinner and longer, its surface area increases resulting in larger mass transfer rates.

4 Conclusions

Mass transfer around a slender drop in an axisymmetric nonlinear extensional and creeping flow is theoretically studied. The fluid mechanics problem, that was first suggested by Sherwood [16] and recently reviewed and expanded by Favelukis [17], is governed by three dimension-

Fig. 7: The mass transfer rate as a function of the capillary number, for different values of ν at $Pe \rightarrow 0$. Solid lines: a slender drop with pointed ends; dashed lines: a slender prolate spheroid with rounded ends

less parameters: The capillary number ($Ca \gg 1$), the viscosity ratio ($\lambda \ll 1$), and the nonlinear intensity of the flow ($|E| \ll 1$) which contrary to the other two parameters, it can be positive or negative. The number of dimensionless parameters can be reduced from three to two: the viscous strength of the flow $F = Ca\lambda^{1/6}$ and the nonlinear strength of the flow $G = Ca^4 E$, both having the order of magnitude of 1.

In a linear extensional flow ($G = 0$), the steady slender drop has a parabolic radius profile with pointed ends. As the capillary number increases the drop becomes thinner, longer and its surface area increases. We follow Acrivos and Lo [13] suggestion by defining the radius at the center of the drop times Ca as $1/(2\nu)$, where ν is a dimensionless parameter related to the steady pressure at the center of the drop. The deformation curve is composed from a lower steady stable branch at $2 \leq \nu < 2.4$ and an upper unstable steady branch at $\nu > 2.4$. These two branches are separated by a bifurcation turning (breakup) point at $\nu = 2.4$ and $F = Ca\lambda^{1/6} = 0.148$. It follows that an inviscid drop ($\lambda = 0$, $\nu = 2$) in linear extensional creeping flow cannot be broken.

When nonlinear effects are present ($G \neq 0$), one can represent the shape of the drop as an infinite power series around the center of the drop. By taking the first two terms, an approximate solution can be constructed suggesting, once again, a parabolic radius profile with pointed ends. This approximate solution, which is represented by a very simple mathematical formula, can be used as an excellent replacement to the exact solution but, up to the breakup point only. Let $H = G/F^6$ be a dimensionless parameter covering the entire numerical range: $-\infty < H < +\infty$. When

$H = 0$ the linear extensional flow ($G = 0$) is recovered, and $H \rightarrow \pm \infty$ corresponds to a bubble or an inviscid drop ($\lambda = 0$) in a nonlinear ($G \neq 0$) extensional flow.

According to the two-term approximate solution, the deformation curve contains two families of solutions. In the first family at $H < 64/9$ (positive or negative), the deformation curve behaves in a similar form like the linear case. That is, a lower steady stable branch at $2 < \nu < \nu_{cr}$ and an upper steady unstable branch at $\nu > \nu_{cr}$ with ν_{cr} being the breakup point located at the bifurcation turning point. Here, contrary to linear case, an inviscid drop ($\lambda = 0$) under negative nonlinear effects ($H = -\infty$), can be broken. The next family of solutions, located at $H > 64/9$ and $\nu < 2$, contains a single branch which is stable everywhere.

The transfer of mass around such a drop is studied for the two asymptotic limits of the Peclet number (Pe). When $Pe \rightarrow \infty$, convection is large (compared to diffusion) and the thin concentration boundary layer approximation can be applied. On the other hand, at $Pe \rightarrow 0$ the problem is satisfied by the Laplace equation suggesting mass transfer by diffusion only (without convection) and with a thick concentration boundary layer.

In order to clarify the results we shall take the case of an inviscid drop in a linear extensional flow ($\nu = 2$) as our reference state. For both regimes of the Peclet number we find that as ν increases, the radius at the center of the drop decreases, the length of the drop increases (to conserve the volume), the surface area increases, leading to larger mass transfer rates. Clearly, much higher mass transfer rates are obtained at the large Peclet numbers regime, since then, convection contributes to the overall mass transfer as well. Thus, compared to our reference case ($\nu = 2$), we conclude that the addition of nonlinear effects to the flow sometimes increase ($H < 64/9$ and $\nu > 2$) and sometimes decrease ($H > 64/9$ and $\nu < 2$) the rate of mass transfer around slender drops in a nonlinear extensional creeping flow.

The exchange of mass between a slender drop and a viscous liquid is a topic of basic scientific interest, with many industrial applications. This process is sometimes performed in rotating devices generating shear, extensional but generally much more complicated flows which include nonlinear terms to the velocity field. This report, which addresses the importance of the nonlinear intensity of the flow on the mass transfer around such drops, provides a better understanding for the design of such equipment.

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