Effect of surfactant concentration and interfacial slip on the flow past a viscous drop at low surface Péclet number

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Abstract

The motion of a viscous drop is investigated when the interface is fully covered with a stagnant layer of surfactant in an arbitrary unsteady Stokes flow for the low surface Péclet number limit. The effect of the interfacial slip coefficient on the behavior of the flow field is also considered. The hydrodynamic problem is solved by the solenoidal decomposition method and the drag force is computed in terms of Faxen's laws using a perturbation ansatz in powers of the surface Péclet number. The analytical expressions for the migration velocity of the drop are also obtained in powers of the surface Péclet number. Further instances corresponding to a given ambient flow as uniform flow, Couette flow, Poiseuille flow are analyzed. Moreover, it is observed that, a surfactant-induced cross-stream migration of the drop occur towards the centre-line in both Couette flow and Poiseuille flow cases. The variation of the drag force and migration velocity is computed for different parameters such as Péclet number, Marangoni number etc.

1 Introduction

The motion of drops and bubbles is a common phenomenon understanding which is important to realize many industrial and chemical applications. Some properties such as deformability, inertia, and external (thermal or chemical) gradients influence the migration of drops. The variation of temperature or the presence of surfactants causes variations in the interfacial gradient. Young, Goldstein and Block [1] were the first to study the flow past a drop by considering thermal effects. Subramanian and Balasubramaniam [2] have computed the drag force in terms of Faxen's laws by considering the thermal effects in an axisymmetric Stokes flow. Subramanian [3] calculated the settling velocity of a drop by considering thermal effects in a steady axisymmetric flow. The unsteady motion of a vertically falling liquid drop in an axisymmetric flow has been analyzed by Chisnell [4]. Dill and Balasubramaniam [5] have studied the thermocapillary migration of a drop in an axisymmetric unsteady Stokes flow. Choudhuri and Padmavathi [6] have calculated the drag and torque in terms of Faxen's laws for an oscillatory Stokes flow past a drop. Choudhuri and Raja Sekhar [7] have obtained the thermocapillary drift of a spherical drop in a steady arbitrary Stokes flow. Ramachandran et al. [8] discussed the impact of interfacial slip on the dynamics of a drop in a Stokes flow by using a numerical approach based on the boundary integral method. Ramachandran and Leal [9] studied the effect of interfacial slip on the drop deformation in a steady Stokes flow by using Navier slip boundary conditions. Mandal et al. [10] computed the shape of a drop by considering the interfacial slip effect in an arbitrary steady Stokes flow by using Lamb's solution.

While these works are mostly on the migration of viscous drops in pure ambient viscous flows, or in presence of thermocapillary effects, there are also studies concerned with the effect of surfactants on the motion of drops and bubbles in creeping flows. Surfactants are surface active agents that are adsorbed at a fluid-fluid interface or at a liquid-gas interface, where they typically lower the interfacial tension and cause a Marangoni effect. It is observed that even a small amount of surfactant can reduce the terminal velocity of a drop. For example, Levan

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and Newman [11] studied the effect of surfactants on the terminal velocity of a drop in an axi symmetric flow. Along the interface, the surfactant is governed by a convection-diffusion equation. Holbrook and LeVan [12] and Holbrook and LeVan [13] have used a collocation method to solve the convection-diffusion problem for high Péclet numbers and studied the retardation of drop motion when the surfactant is present. Sadhal and Johnson [14] studied the flow past a drop which is partially coated with a stagnant layer of surfactant for large surface Péclet number. Many authors have examined the effect of soluble and insoluble surfactants on the motion of drops using various numerical techniques (Ref. [15, 16]). Stone [17] derived a convection-diffusion equation for the surfactant transport along a deforming interface. Stone and Leal [18] used a numerical treatment to analyze the effect of surfactants on the deformation and breakup of a drop. Hanna and Vlahovska [19] discussed the surfactant-induced migration of a drop in an unbounded Poiseuille flow for large Péclet numbers. A simplified CFD simulation was performed to study the influence of surfactants on the rise of bubbles by Fleckenstein and Bothe [20]. Recently, Pak et al. [21] calculated the migration of a drop in a steady Poiseuille flow at low surface Péclet numbers.

The migration of a non-deforming clean spherical viscous drop at zero Reynolds number in a pressure driven flow moves only along the flow direction (Ref. [22]), i.e., there can be no cross migration in the absence of inertia and deformation on a clean spherical drop. It is experimentally observed that, for three dimensional Poiseuille flow and for Couette flow, the migration due to deformation occurs towards the center line (Ref. [23–25]). The cross migration due to inertial effects is also studied by many authors (Ref. [26, 27]). It is also found that the surfactant redistribution can also cause the cross stream migration of drops (Ref. [19, 21, 28]). Recently, Mandal et al. [10] have studied the effect of interfacial slip on the cross migration of a drop in an unbounded Poiseuille flow. However, these studies are restricted to steady case and ambient Poiseuille flow. We are generalizing the problem to an unsteady arbitrary ambient flow, by considering the effects of interfacial slip as well as surfactant concentration effects.

We are interested in the case of arbitrary Stokes flow past drops which is challenging due to its three dimensional nature. Note that the corresponding drag and torque can be obtained in a compact form similar to Faxen's laws. For example, the recent study by Choudhuri and Raja Sekhar [7] discussed thermocapillary migration of a viscous spherical drop and obtained the corresponding Faxen's laws. Consequently, Sharanya and Raja Sekhar [29] have addressed thermocapillary migration of a spherical drop in an arbitrary unsteady Stokes flow. We are motivated by these studies and consider the motion of a viscous spherical drop whose interface is covered with a stagnant layer of surfactant in an arbitrary unsteady Stokes flow. The arbitrary Stokes flow case is considered by Pak et al. [21], where they restrict the flow to be steady, and the surfactant coating the whole interface. The slip reduces the deformation of a drop in a shear-type flow (Ref. [8, 9]). Also, it is noted that due to this slip condition the disturbance flow produced by a drop is expected to be weakened in magnitude. In our present case, we attempt a more generalized problem of an arbitrary transient Stokes flow past a drop for low surface Péclet number. Also, we take into account the effect of interfacial slip on the flow. We solve the problem for any given ambient flow and consider some special cases to validate our results.

The objective of our present paper is to analyze the behavior of the flow when the interfacial slip effect and the surfactant concentration effect occurs for low surface Péclet numbers. We use the solenoidal decomposition method to solve the unsteady Stokes equations, which is motivated by the general solution proposed by Venkatalaxmi et al. [30]. We use slip boundary conditions to see the effect of interfacial slip on the flow behavior which has been previously used by Ramachandran et al. [8] and Ramachandran and Leal [9]. If we denote the surfactant concentration as Γ, we assume that Γ is governed by a convection-diffusion equation [14, 17, 31]. We find the surfactant concentration up to second order for an arbitrary Stokes flow, i.e., up to O(Pe_s^2) (Ref. [21]). We observe area-specific surfactant distribution on the interface of the drop. We also solve for the flow fields and obtain the settling velocity of the drop. We compute migration velocity corresponding to surfactant coated drop in Poiseuille flow and Couette flow and make some observations on the cross flow migration.

2 Problem Statement and Mathematical Formulation

We consider the motion of a liquid drop of radius a and viscosity μ^i in an unsteady Stokes flow, suspended in another unbounded Newtonian fluid of viscosity μ^e (see Fig. 1). Let the velocity of the fluid inside the drop

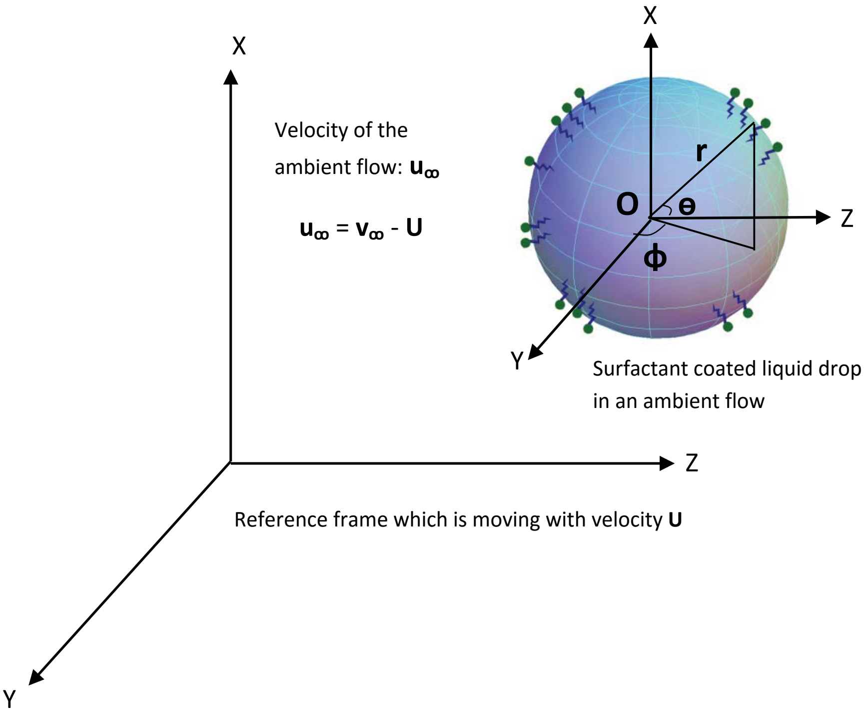

Figure 1: Geometry of the problem

be $\vec{v}^i$ and the velocity of the fluid outside the drop be $\vec{v}^e$. We assume that the settling velocity of the drop is **U**, which we determine later. The presence of a small amount of surface-active agents (surfactants) causes the variation in interfacial tension which influences the migration of the drop. We analyze the problem when the surfactant concentration effects and interfacial slip effects are considered. Surfactants are surface-active agents that lower the interfacial tension between two liquids. We neglect the inertial terms under negligible Reynolds number assumption. We assume a low surface Péclet number Pe_s. Further, we assume that the dimensional interfacial tension, σ^*, depends in an affine way on the dimensional surfactant concentration, Γ^*, i.e.,

$$\sigma^* = \sigma - RT\Gamma^*,$$

where σ is the interfacial tension when the interface is clean, R is the gas constant and T is the absolute temperature (Ref. [21]). We non-dimensionalize the lengths by the drop radius a, velocities by the characteristic velocity scale of the background flow, U_c, time by its characteristic time scale t_c and the surfactant concentration by its equilibrium value when the distribution is uniform, Γ_{eq}. The pressure is non dimensionalized by $\frac{\mu U_c}{a}$.

We assume that the flow inside and outside the drop is governed by the unsteady Stokes equations and the continuity equations which are given in the non dimensional form as follows:

for $r < 1$

$$\beta_i \frac{\partial \vec{v}^i}{\partial t} = -\vec{\nabla} p^i + \vec{\nabla}^2 \vec{v}^i; \qquad \vec{\nabla}.\vec{v}^i = 0, \tag{2.1}$$

and for $r > 1$

$$\beta_e \frac{\partial \vec{v}^e}{\partial t} = -\vec{\nabla} p^e + \vec{\nabla}^2 \vec{v}^e; \qquad \vec{\nabla}.\vec{v}^e = 0. \tag{2.2}$$

In the above equations, $\beta_e = \frac{a^2}{\nu_e t_c}$ and $\beta_i = \frac{a^2}{\nu_i t_c}$ represent the unsteadiness parameters corresponding to the flow inside and outside the drop respectively, which we assume to be unity, i.e., $t_c = \frac{a^2}{\nu_j}$.

We assume that the velocity field far from the drop approaches the undisturbed background flow, $\vec{v}^{\infty}$, i.e.,

$$\vec{v}^e \to \vec{v}_\infty \quad as \quad r \to \infty, \tag{2.3}$$

which together with some pressure field p_∞ satisfies the unsteady Stokes and continuity equations.

The surfactant transport is governed by an unsteady convection-diffusion equation, (Ref. Stone [17] and Sadhal and Johnson [14]), which is given in the non dimensional form as follows

$$Pr_s \frac{\partial \Gamma}{\partial t} + Pe_s \left[\vec{\nabla}_s.(\Gamma \vec{v}_s) + \Gamma(\vec{v}.\hat{n}) \vec{\nabla}_s.\hat{n} \right] = \vec{\nabla}_s^2 \Gamma, \tag{2.4}$$

where $\vec{v}_s = \vec{v}^e.\hat{t}$ is the velocity component tangential to the surface of the drop and $Pe_s = \frac{aU_c}{D_s}$ is the surface Péclet number which measures the importance of convection relative to diffusion. Here D_s is the dimensional surface-diffusion constant. $Pr_s = \frac{\nu_e}{D_s}$ is the Prandtl number which is dimensionless and is defined as the ratio of momentum diffusivity to surfactant diffusivity. Eq. (2.4) includes the convective and diffusive contribution to the surfactant transport and a source-like contribution accounting for the variation of surfactant concentration resulting from the local changes in the interfacial area. (Ref. [17]).

We solve the problem in a reference frame which is moving with the velocity of the drop, **U**, in which the drop appears to be stationary (see Fig. (1)). In this moving frame, the velocity fields inside and outside the drop are given respectively by

$$\vec{u}^i = \vec{v}^i - \mathbf{U},$$

$$\vec{u}^e = \vec{v}^e - \mathbf{U}.$$

One can observe that, these velocity fields also satisfy the unsteady Stokes and continuity equations given by

for $r < 1$

$$\frac{\partial \vec{u}^i}{\partial t} = -\vec{\nabla} p^i + \vec{\nabla}^2 \vec{u}^i; \qquad \vec{\nabla}.\vec{u}^i = 0, \tag{2.5}$$

and for $r > 1$

$$\frac{\partial \vec{u}^e}{\partial t} = -\vec{\nabla} p^e + \vec{\nabla}^2 \vec{u}^e; \qquad \vec{\nabla}.\vec{u}^e = 0. \tag{2.6}$$

The external velocity $\vec{u}^e$ is expected to meet the following far field condition in the reference frame

$$\vec{u}^e \to \vec{u}_\infty = \vec{v}_\infty - \mathbf{U} \quad as \quad r \to \infty. \tag{2.7}$$

We follow the physical interpretations discussed by various authors [2,32–34] and adopt the following kinematic boundary conditions on the surface of the drop in non-dimensional form:

Vanishing normal component of the velocities, i.e.,

$$\vec{u}^e.\hat{n} = 0; \qquad \vec{u}^i.\hat{n} = 0, \tag{2.8}$$

Slip in the tangential component of velocities, i.e.,

$$\vec{u}^e.\hat{t} - \vec{u}^i.\hat{t} = \alpha \tau^e_{\hat{n}\hat{t}}, \tag{2.9}$$

Tangential stress balance, i.e.,

$$\tau^e_{\hat{n}\hat{t}} - \mu \tau^i_{\hat{n}\hat{t}} = Ma \vec{\nabla}_s \Gamma.\hat{t}, \tag{2.10}$$

Since the stress fields and the surfactant concentration on the surface of the drop remain the same in both the laboratory frame and the moving frame, the tangential stress balance takes the same form as in both reference frames. We note that the surfactant transport equation given in Eq. (2.4) simplifies to

$$Pr_s \frac{\partial \Gamma}{\partial t} + Pe_s \left[\vec{\nabla}_s.(\Gamma \vec{u}_s) \right] = \vec{\nabla}_s^2 \Gamma, \tag{2.11}$$

in the moving reference frame. Here $\vec{u}_s$ is the velocity tangential to the surface of the drop in the moving frame.

3 Method of solution

We expand the velocity and pressure fields, surfactant concentration and migration velocity as a regular perturbation expansion for low surface Péclet number ($Pe_s \ll 1$), i.e.,

$$\begin{aligned}\left[\vec{u}^e, \vec{u}^i, p^e, p^i, \Gamma, \mathbf{U}\right] &= \left[\vec{u}_0^e, \vec{u}_0^i, p_0^e, p_0^i, \Gamma_0, \mathbf{U}_0\right] + Pe_s\left[\vec{u}_1^e, \vec{u}_1^i, p_1^e, p_1^i, \Gamma_1, \mathbf{U}_1\right] \\ &\quad + Pe_s^2\left[\vec{u}_2^e, \vec{u}_2^i, p_2^e, p_2^i, \Gamma_2, \mathbf{U}_2\right] + O(Pe_s^3). \end{aligned} \tag{3.1}$$

Since the boundary value problem defined in Eqs. (2.5) to (2.10) is independent of the perturbation parameter Pe_s, the velocity and pressure fields at all orders satisfy similar equations with the corresponding quantities as: leading order ($\vec{u}_0, p_0$), first order ($\vec{u}_1, p_1$), and second order ($\vec{u}_2, p_2$) etc. For brevity, we do not repeat these equations here.

3.1 Representation of velocity

By eliminating the pressure from the unsteady Stokes equations, one can verify that the velocity fields inside and outside the droplet satisfy

$$\vec{\nabla}^2\left(\vec{\nabla}^2 - \frac{\partial}{\partial t}\right)\vec{u}^j = 0 \quad \text{for } j = i, e. \tag{3.2}$$

By using the general solution for the unsteady Stokes equation together with the equation of continuity, we can have the following representation for the velocity and pressure fields (see [30])

$$\vec{u}^j = \vec{\nabla} \times \vec{\nabla} \times (\mathbf{r}\chi^j) + \vec{\nabla} \times (\mathbf{r}\eta^j), \tag{3.3}$$

$$p^j = p_\infty^j + \rho_j \frac{\partial}{\partial r}\left(\mathbf{r}\left(\vec{\nabla}^2\chi^j - \frac{\partial \chi^j}{\partial t}\right)\right), \tag{3.4}$$

where the scalars χ^j and η^j are solutions of

$$\vec{\nabla}^2\left(\vec{\nabla}^2 - \frac{\partial}{\partial t}\right)\chi^j = 0, \tag{3.5}$$

$$\left(\vec{\nabla}^2 - \frac{\partial}{\partial t}\right)\eta^j = 0. \tag{3.6}$$

Here $\mathbf{r}$ is the position vector and p_∞ is a constant. Hence, the problem can now be handled in terms of the scalars χ^j and η^j. Accordingly, the boundary conditions in terms of χ^j and η^j are given by

Vanishing normal component of the velocity

$$\chi^e = \chi^i = 0 \quad \text{on } r = 1\,. \tag{3.7}$$

Slip in the tangential component of velocity

$$\frac{\partial \chi^e}{\partial r} - \frac{\partial \chi^i}{\partial r} = \alpha \frac{\partial^2 \chi^e}{\partial r^2}, \quad \eta^e - \eta^i = \alpha \frac{\partial}{\partial r}\left(\frac{\eta^e}{r}\right) \quad \text{on } r = 1\,. \tag{3.8}$$

Tangential stress balance

$$\frac{\partial}{\partial \theta}\left(\frac{\partial^2 \chi^e}{\partial r^2} - \mu \frac{\partial^2 \chi^i}{\partial r^2}\right) = Ma \frac{\partial \Gamma}{\partial \theta} \quad \text{on } r = 1\,, \tag{3.9}$$

$$\frac{\partial}{\partial \phi}\left(\frac{\partial^2\chi^e}{\partial r^2}-\mu\frac{\partial^2\chi^i}{\partial r^2}\right)=Ma\frac{\partial \Gamma}{\partial \phi} \quad \text{on } r=1\,, \tag{3.10}$$

$$\frac{\partial}{\partial r}\left(\frac{\eta^e}{r}\right)=\mu\frac{\partial}{\partial r}\left(\frac{\eta^i}{r}\right) \quad \text{on } r=1. \tag{3.11}$$

Finite velocity and pressure fields inside the drop require that

$$\chi^i<\infty, \quad \eta^i<\infty. \tag{3.12}$$

3.2 Leading order problem

The zeroth order surfactant transport equation corresponding to the general case given in (2.11) is

$$Pr_s\frac{\partial \Gamma_0}{\partial t}=\vec{\nabla}_s^2\Gamma_0\,. \tag{3.13}$$

In order to obtain the leading order surfactant concentration Γ_0, we express Γ_0 in terms of spherical harmonics, i.e.,

$$\Gamma_0=\sum_{n=0}^{\infty}R_n^0(\theta,\phi)e^{-\lambda^2t/Pr_s}\,, \tag{3.14}$$

where

$$R_n(\theta,\phi)=\sum_{m=0}^{n}\left(E_{nm}^0\cos m\phi+F_{nm}^0\sin m\phi\right)P_n^m(\cos\theta), \tag{3.15}$$

are the spherical harmonics, $P_n^m(\eta)$ are associated Legendre polynomials and E_{nm}^0, F_{nm}^0 have to be determined such that Γ_0 satisfies (3.13). Substituting the above expression (3.14) in (3.13), we obtain $n(n+1)=-\lambda^2/Pr_s$. This is possible only when $n=0$ and $\lambda=0$ since we have $\lambda^2>0$. Therefore we have that Γ_0 is a constant, which we take as unity, i.e., $\Gamma_0=1$.

We represent the far-field ambient flow in terms of χ_0^∞ and η_0^∞, given by

$$\chi_0^\infty=\sum_{n=1}^{\infty}\left[\alpha_n^0r^n+\beta_n^0f_n(\lambda_e r)\right]S_n(\theta,\phi)e^{\lambda_e^2t}, \tag{3.16}$$

$$\eta_0^\infty=\sum_{n=1}^{\infty}\left[\gamma_n^0f_n(\lambda_e r)\right]T_n(\theta,\phi)e^{\lambda_e^2t}, \tag{3.17}$$

where

$$S_n^0(\theta,\phi)=\sum_{m=0}^{n}P_n^m(\eta)\left[A_{nm}^0\cos m\phi+B_{nm}^0\sin m\phi\right], \tag{3.18}$$

$$T_n^0(\theta,\phi)=\sum_{m=0}^{n}P_n^m(\eta)\left[C_{nm}^0\cos m\phi+D_{nm}^0\sin m\phi\right], \tag{3.19}$$

are spherical harmonics, and α_n^0, β_n^0, γ_n^0, A_{nm}^0, B_{nm}^0, C_{nm}^0 and D_{nm}^0 are the known coefficients. These coefficients are controlled by the choice of the ambient flow. For example, in case of uniform ambient flow, $\chi_0^\infty=\frac{1}{2}r\cos\theta e^{\lambda_e^2t}$, $\eta_0^\infty=0$; and hence $\alpha_1^0=\frac{1}{2}$, $\alpha_n^0=0$ for $n\neq 1$, $\beta_n^0=0$, $\gamma_n^0=0$, $A_{10}^0=1$, $A_{nm}^0=0$ for $n\neq 1$ or $m\neq 0$, $B_{nm}^0=0$, $C_{nm}^0=0$ and $D_{nm}^0=0$. Here, $f_n(\lambda_j r)$ and $g_n(\lambda_j r)$ $(j=i,e)$ are modified spherical Bessel function of first and second kind, respectively. Note that, for the bounded solution as $t \to \infty$, we require $\lambda_j^2 < 0$. In the presence of the spherical drop, the resultant flow due to the disturbance can be represented as general solution of Eqs. (3.5) and (3.6) as follows,

for $r < 1$

$$\chi_0^i = \sum_{n=1}^{\infty} \left[\bar{\alpha}_n^0 r^n + \bar{\beta}_n^0 f_n(\lambda_i r)\right] S_n^0(\theta,\phi) e^{\lambda_i^2 t}, \tag{3.20}$$

$$\eta_0^i = \sum_{n=1}^{\infty} \left[\bar{\gamma}_n^0 f_n(\lambda_i r)\right] T_n^0(\theta,\phi) e^{\lambda_i^2 t}, \tag{3.21}$$

and for $r > 1$

$$\chi_0^e = \sum_{n=1}^{\infty} \left[\alpha_n^0 r^n + \frac{\hat{\alpha}_n^0}{r^{n+1}} + \beta_n^0 f_n(\lambda_e r) + \hat{\beta}_n^0 g_n(\lambda_e r)\right] S_n^0(\theta,\phi) e^{\lambda_e^2 t}, \tag{3.22}$$

$$\eta_0^e = \sum_{n=1}^{\infty} \left[\gamma_n^0 f_n(\lambda_e r) + \hat{\gamma}_n^0 g_n(\lambda_e r)\right] T_n^0(\theta,\phi) e^{\lambda_e^2 t}, \tag{3.23}$$

where $\bar{\alpha}_n^0$, $\bar{\beta}_n^0$, $\bar{\gamma}_n^0$, $\hat{\alpha}_n^0$, $\hat{\beta}_n^0$, $\hat{\gamma}_n^0$ are the unknown coefficients which are to be determined subject to the boundary conditions (3.7) to (3.12), and λ_i, λ_e are the amplification factors corresponding to the flow inside and outside of the drop which can be found if the initial conditions are provided (Ref. [30]). Moreover, the far field condition turns out to be $\chi_0^e \to \chi_0^\infty$ and $\eta_0^e \to \eta_0^\infty$ as $r \to \infty$. The unknown coefficients can be expressed in terms of the known ambient flow variables using the boundary conditions. We present these details in Appendix A.

The zeroth order drag force experienced by a spherical drop can be computed using the formula

$$\vec{D} = \int_{\theta=0}^{\pi} \int_{\phi=0}^{2\pi} \bar{\bar{\tau}}.\hat{n}\, dS, \tag{3.24}$$

where dS represents the surface element, $\hat{n}$ is the unit normal to the boundary of the drop, **r** is the position vector and $\bar{\bar{\tau}}$ is the stress tensor. We have computed zeroth order thermocapillary drift in case of transient Stokes flow past a viscous drop, and expressed in terms of Faxen's laws, given by

$$\vec{D}_0 = 4\pi\lambda_e^2 \hat{\alpha}_1^0 \left(A_{11}^0 \hat{\imath} + B_{11}^0 \hat{\jmath} + A_{10}^0 \hat{k}\right) e^{\lambda_e^2 t}. \tag{3.25}$$

Note that the above structure in terms of the known vector (A_{11}^0, B_{11}^0, A_{10}^0) is due to the spherical harmonics $S_n^0(\theta,\phi)$ given in (3.18). Corresponding to a given ambient flow, one can determine the coefficient $\hat{\alpha}_1^0$. For example, in case of uniform ambient flow, we have $n = 1$ and the corresponding expression for $\hat{\alpha}_1^0$ can be obtained using $\hat{\alpha}_n^0$ given in Appendix A. Consequently from Eq. (3.25), we have the following expression for the drag force

$$\vec{D}_0 = 2\pi \left[\frac{Y + \mu X + \alpha P}{W + \mu Z + \alpha G}[\vec{u}_{0\infty}]_0 + \frac{V + \mu U + \alpha H}{W + \mu Z + \alpha G}[\vec{\nabla}^2 \vec{u}_{0\infty}]_0\right]. \tag{3.26}$$

The above quantity depends on $\mu = \frac{\mu^i}{\mu^e}$, the ratio of the viscosities, and α the dimensionless slip coefficient. Since $\Gamma_0 = 1$, $\vec{\nabla}_s \Gamma_0$ vanishes and the tangential stress becomes continuous. Hence at leading order, we do not observe any influence of the surfactant. The expanded form of the quantities $X, Y, P, G, Z, W, U, V, H$ etc., are given in Appendix B. It may be noted that the above compact form is due to the following relations

$$[\vec{u}_{0\infty}]_0 = (2\alpha_1^0 + \frac{2}{3}\lambda_e \beta_1^0)(A_{11}^0 \hat{\imath} + B_{11}^0 \hat{\jmath} + A_{10}^0 \hat{k}) e^{\lambda_e^2 t},$$

$$[\vec{\nabla}^2\vec{u}_{0\infty}]_0 = \frac{2}{3}\lambda_e^3\beta_1^0(A_{11}^0\hat{\imath} + B_{11}^0\hat{\jmath} + A_{10}^0\hat{k})e^{\lambda_e^2 t},$$

$$[\vec{\nabla}\times\vec{u}_{0\infty}]_0 = \frac{2\lambda_e}{3}\gamma_1^0(C_{11}^0\hat{\imath} + D_{11}^0\hat{\jmath} + C_{10}^0\hat{k})e^{\lambda_e^2 t}.$$

One may observe that, when the slip coefficient in the zeroth order drag force is equal to zero (i.e., $\alpha = 0$), then the drag force reduces to

$$\vec{D}_0 = 2\pi\left[\frac{Y+\mu X}{W+\mu Z}[\vec{u}_{0\infty}]_0 + \frac{V+\mu U}{W+\mu Z}[\vec{\nabla}^2\vec{u}_{0\infty}]_0\right]. \tag{3.27}$$

In the context of thermocapillary migration of a spherical drop, Sharanya and Raja Sekhar [29] obtained an expression for the drag force exerted on the spherical drop. The above expression (3.27) agrees with their results when the thermocapillary effects are neglected. Table (1) gives some additional understanding in this regard. Note that the zeroth order drag force given in (3.26) is with respect to a reference frame which is moving with a velocity $\boldsymbol{U}_0$. Therefore the drag force in the laboratory reference frame in terms of a given ambient hydrodynamic field is given by

$$\vec{D}_0 = 2\pi\left[\frac{Y+\mu X+\alpha P}{W+\mu Z+\alpha G}\left([\vec{v}_{0\infty}]_0 - \mathbf{U}_0\right) + \frac{V+\mu U+\alpha H}{W+\mu Z+\alpha G}[\vec{\nabla}^2\vec{v}_{0\infty}]_0\right], \tag{3.28}$$

where $\mathbf{U}_0$ is the zeroth order migration velocity which is yet to be determined.

The force balance in the absence of gravity when the flow is transient is given by (Refs. [2,4]),

$$M\frac{d\mathbf{U}}{dt} = \vec{D}, \tag{3.29}$$

where $M = \frac{4}{3}\pi\rho_i$ is the mass of the drop with unit radius. Here, ρ_i is the density of the drop. From the above equation (3.29), we have the leading order force balance as follows

$$M\frac{d\mathbf{U}_0}{dt} = \vec{D}_0\,. \tag{3.30}$$

On using the expression for the drag given in (3.28) (general case), this would enable us to obtain the following expression for the migration velocity of the drop

$$\begin{aligned}\mathbf{U}_0 \;=\; & \frac{3}{2\rho_i+\rho_e}\left[\frac{Y+\mu X+\alpha P}{W+\mu Z+\alpha G}[\vec{v}_{0\infty}]_0 + \frac{V+\mu U+\alpha H}{W+\mu Z+\alpha G}[\vec{\nabla}^2\vec{v}_{0\infty}]_0\right]\\ & \left(\frac{3}{2\rho_i+\rho_e}\frac{Y+\mu X+\alpha P}{W+\mu Z+\alpha G} + \lambda_e^2\right)^{-1}.\end{aligned} \tag{3.31}$$

We may observe that, when the slip coefficient is zero, the above zeroth order migration velocity reduces to the one that is obtained by Sharanya and Raja Sekhar [29] provided thermal effects are neglected. In this case, we have

$$\begin{aligned}\mathbf{U}_0 \;=\; & \frac{3}{2\rho_i+\rho_e}\left[\frac{Y+\mu X}{W+\mu Z}[\vec{v}_{0\infty}]_0 + \frac{V+\mu U}{W+\mu Z}[\vec{\nabla}^2\vec{v}_{0\infty}]_0\right]\\ & \left(\frac{3}{2\rho_i+\rho_e}\frac{Y+\mu X}{W+\mu Z} + \lambda_e^2\right)^{-1}.\end{aligned} \tag{3.32}$$

If we consider the limiting case of no oscillations in the hydrodynamic flow field, i.e., $\lambda_i = \lambda_e = 0$, and zero slip coefficient, i.e., $\alpha = 0$, then the zeroth order terminal velocity reduces to

$$\mathbf{U}_0 = [\vec{v}_\infty]_0 + \frac{\mu}{4+6\mu}[\vec{\nabla}^2\vec{v}_\infty]_0, \tag{3.33}$$

which is exactly matching with the one that is obtained by Pak, Feng and Stone [21].

3.2.1 Stationary drop

If we assume that the drop is stationary, then we have $\vec{v}_\infty = \vec{u}_\infty$. In this case, the zeroth order drag force is given by

$$\vec{D}_0 = 2\pi \left[\frac{Y + \mu X + \alpha P}{W + \mu Z + \alpha G} [\vec{v}_{0\infty}]_0 + \frac{V + \mu U + \alpha H}{W + \mu Z + \alpha G} [\vec{\nabla}^2 \vec{v}_{0\infty}]_0 \right], \tag{3.34}$$

which agrees with the corresponding result that is obtained by Choudhuri and Padmavati [6] when the slip coefficient is zero (Ref. Table (1)).

3.3 First-order correction

The first order surfactant transport equation due to the expansion (3.1) and Eq. (2.11) is given by

$$Pr_s \frac{\partial \Gamma_1}{\partial t} + \vec{\nabla}_s . \vec{u}_{0s} = \vec{\nabla}_s^2 \Gamma_1, \tag{3.35}$$

where $\vec{u}_{0s}$ is the zeroth order tangential velocity vector on the drop surface. Assuming that the surfactant concentration is oscillatory, i.e., $\Gamma_1(\theta, \phi, t) = \Gamma_1(\theta, \phi) e^{-i\omega t} = \Gamma_1(\theta, \phi) e^{-l^2 t / Pr_s}$, Eq. (3.35) reduces to

$$(\vec{\nabla}_s^2 + l^2)\Gamma_1 = \vec{\nabla}_s . \vec{u}_{0s}. \tag{3.36}$$

In order to obtain the first order surfactant concentration Γ_1, we express Γ_1 in terms of spherical harmonics, i.e.,

$$\Gamma_1 = \sum_{n=1}^{\infty} R_n^1(\theta, \phi) e^{-l^2 t / Pr_s}, \tag{3.37}$$

where

$$R_n^1(\theta, \phi) = \sum_{m=0}^{n} \left(E_{nm}^1 \cos m\phi + F_{nm}^1 \sin m\phi \right) P_n^m(\cos\theta), \tag{3.38}$$

are the spherical harmonics, and E_{nm}^1, F_{nm}^1 have to be determined such that Γ_1 satisfies the Eq.(3.37). Since $\vec{\nabla}_s^2 R_n^1(\theta, \phi) = -n(n+1) R_n^1(\theta, \phi)$, we observe that $\vec{\nabla}_s^2 \Gamma_1 = -\sum\limits_{n=1}^{\infty} n(n+1) R_n^1(\theta, \phi) e^{l^2 t / Pr_s}$. The coefficients in $R_n^1(\theta, \phi)$ can be determined as follows:

$$\sum_{n=0}^{\infty} \sum_{m=0}^{n} (-n(n+1) + l^2) \left[E_{nm}^1 \cos m\phi + F_{nm}^1 \sin m\phi \right] P_n^m(\cos\theta) e^{-l^2 t / Pr_s} = \vec{\nabla}_s . \vec{u}_{0s}. \tag{3.39}$$

This enables us to write the following relations

$$\begin{aligned} E_{kj}^1 \pi \frac{2(k+j)!}{(2k+1)(k-j)!} e^{-l^2 t / Pr_s} &= \frac{-1}{k(k+1) - l^2} \int_{\phi=0}^{2\pi} \int_{\theta=0}^{\pi} (\vec{\nabla}_s . \vec{u}_{0s}) P_k^j(\cos\theta) \\ &\quad \cos j\phi \sin\theta \, d\theta \, d\phi, \end{aligned} \tag{3.40}$$

$$\begin{aligned} F_{kj}^1 \pi \frac{2(k+j)!}{(2k+1)(k-j)!} e^{-l^2 t / Pr_s} &= \frac{-1}{k(k+1) - l^2} \int_{\phi=0}^{2\pi} \int_{\theta=0}^{\pi} (\vec{\nabla}_s . \vec{u}_{0s}) P_k^j(\cos\theta) \\ &\quad \sin j\phi \sin\theta \, d\theta \, d\phi, \end{aligned} \tag{3.41}$$

which implies $-l^2 / Pr_s = \lambda_e^2 (< 0)$ and

$$\begin{aligned} E_{nm}^1 &= \Big[(n+1)\alpha_n^0 + \beta_n^0 \left(\lambda_e f_{n+1}(\lambda_e) + (n+1) f_n(\lambda_e) \right) \\ &\quad - n\hat{\alpha}_n^0 + \hat{\beta}_n^0 \left((n+1) g_n(\lambda_e) - \lambda_e g_{n+1}(\lambda_e) \right) \Big] \times \left[A_{nm}^0 \frac{n(n+1)}{n(n+1) + \lambda_e^2 Pr_s} \right], \end{aligned} \tag{3.42}$$

$$F^1_{nm} = \Big[(n+1)\alpha^0_n + \beta^0_n\left(\lambda_e f_{n+1}(\lambda_e) + (n+1) f_n(\lambda_e)\right) \\ - n\hat{\alpha}^0_n + \hat{\beta}^0_n\left((n+1) g_n(\lambda_e) - \lambda_e g_{n+1}(\lambda_e)\right)\Big] \times \left[B^0_{nm}\frac{n(n+1)}{n(n+1)+\lambda_e^2 Pr_s}\right]. \tag{3.43}$$

The first-order pressure and velocity fields satisfy the unsteady Stokes and continuity equations. Correspondingly, we express χ^i_1, η^i_1, χ^e_1 and η^e_1 as follows

$$\chi^i_1 = \sum_{n=1}^{\infty}\left[\bar{\alpha}^1_n r^n + \bar{\beta}^1_n f_n(\lambda_i r)\right] S^1_n(\theta,\phi) e^{\lambda_i^2 t}, \tag{3.44}$$

$$\eta^i_1 = \sum_{n=1}^{\infty}\left[\bar{\gamma}^1_n f_n(\lambda_i r)\right] T^1_n(\theta,\phi) e^{\lambda_i^2 t}, \tag{3.45}$$

$$\chi^e_1 = \sum_{n=1}^{\infty}\left[\alpha^1_n r^n + \frac{\hat{\alpha}^1_n}{r^{n+1}} + \beta^1_n f_n(\lambda_e r) + \hat{\beta}^1_n g_n(\lambda_e r)\right] S^1_n(\theta,\phi) e^{\lambda_e^2 t}, \tag{3.46}$$

$$\eta^e_1 = \sum_{n=1}^{\infty}\left[\gamma^1_n f_n(\lambda_e r) + \hat{\gamma}^1_n g_n(\lambda_e r)\right] T^1_n(\theta,\phi) e^{\lambda_e^2 t}, \tag{3.47}$$

where $S^1_n(\theta,\phi)$ and $T^1_n(\theta,\phi)$ are spherical harmonics of order n. The interfacial surfactant that is coupled via the boundary conditions (3.9) and (3.10) together with the form of Γ_1 given in (3.37) enforces $S^1_n(\theta,\phi) = R^1_n(\theta,\phi)$. However, we have

$$T^1_n(\theta,\phi) = \sum_{m=0}^{n}\left(E'_{nm}\cos m\phi + F'_{nm}\sin m\phi\right) P^m_n(\cos\theta). \tag{3.48}$$

We have given the expressions for the unknown coefficients, α^1_n, $\hat{\alpha}^1_n$, β^1_n, $\hat{\beta}^1_n$, γ^1_n, $\hat{\gamma}^1_n$, $\bar{\alpha}^1_n$, $\bar{\beta}^1_n$ and $\bar{\gamma}^1_n$, in Appendix C. Following a similar approach that is used to solve the leading order problem, we compute the first order drag given by

$$\vec{D}_1 = 2\pi\Big[-\frac{Y+\mu X+\alpha P}{W+\mu Z+\alpha G}\mathbf{U}_1 + \frac{2Ma\lambda_e^2 f_2(\lambda_i) g_1(\lambda_e)}{(W+\mu Z+\alpha G)} \\ \times (E^1_{11}\hat{i} + F^1_{11}\hat{j} + E^1_{10}\hat{k}) e^{\lambda_e^2 t}\Big]. \tag{3.49}$$

The force balance $M\frac{d\mathbf{U}_1}{dt} = \vec{D}_1$ together with the expression for $\vec{D}_1$ given in Eq. (3.49) leads to the first order migration velocity of the drop

$$\mathbf{U}_1 = \frac{3}{2\rho_i+\rho_e}\left[\frac{2Ma\lambda_e^2 f_2(\lambda_i) g_1(\lambda_e)}{(W+\mu Z+\alpha G)}(E^1_{11}\hat{i} + F^1_{11}\hat{j} + E^1_{10}\hat{k}) e^{\lambda_e^2 t}\right] \\ \times \left(\frac{3}{2\rho_i+\rho_e}\frac{Y+\mu X+\alpha P}{W+\mu Z+\alpha G} + \lambda_e^2\right)^{-1}, \tag{3.50}$$

where

$$E^1_{11} = \frac{2A^0_{11}}{(2+\lambda_e^2 Pr_s)}\left[2\alpha^0_n + \beta^0_n\left(\lambda_e f_2(\lambda_e) + 2 f_1(\lambda_e)\right) - \hat{\alpha}^0_n + \hat{\beta}^0_n\left(2 g_1(\lambda_e) - \lambda_e g_2(\lambda_e)\right)\right], \tag{3.51}$$

$$F_{11}^1 = \frac{2B_{10}^0}{(2+\lambda_e^2 Pr_s)} \left[2\alpha_n^0 + \beta_n^0 \left(\lambda_e f_2(\lambda_e) + 2f_1(\lambda_e)\right) - \hat{\alpha}_n^0 + \hat{\beta}_n^0 \left(2g_1(\lambda_e) - \lambda_e g_2(\lambda_e)\right) \right], \tag{3.52}$$

and

$$E_{10}^1 = \frac{2A_{10}^0}{(2+\lambda_e^2 Pr_s)} \left[2\alpha_n^0 + \beta_n^0 \left(\lambda_e f_2(\lambda_e) + 2f_1(\lambda_e)\right) - \hat{\alpha}_n^0 + \hat{\beta}_n^0 \left(2g_1(\lambda_e) - \lambda_e g_2(\lambda_e)\right) \right], \tag{3.53}$$

Here we observe that, only three modes of concentration E_{11}^1, F_{11}^1 and E_{10}^1 are contributing to the drag and migration velocity. If we consider the special case of steady flow past a droplet, i.e., $\lambda_e = \lambda_i = 0$, the first order migration velocity reduces to

$$\mathbf{U}_1 = \frac{2Ma}{6+9\mu+18\alpha\mu}(e_{11}^1\hat{i} + f_{11}^1\hat{j} + e_{10}^1\hat{k}), \tag{3.54}$$

where

$$e_{kj}^1 \pi \frac{2(k+j)!}{(2k+1)(k-j)!} = \frac{-1}{k(k+1)} \int_{\phi=0}^{2\pi} \int_{\theta=0}^{\pi} (\vec{\nabla}_s.\vec{u}_{0s}) P_k^j(\cos\theta) \cos j\phi \sin\theta \, d\theta \, d\phi, \tag{3.55}$$

$$f_{kj}^1 \pi \frac{2(k+j)!}{(2k+1)(k-j)!} = \frac{-1}{k(k+1)} \int_{\phi=0}^{2\pi} \int_{\theta=0}^{\pi} (\vec{\nabla}_s.\vec{u}_{0s}) P_k^j(\cos\theta) \sin j\phi \sin\theta \, d\theta \, d\phi. \tag{3.56}$$

In particular,

$$e_{11}^1 = A_{11}^0 \left[\frac{\alpha_1^0 (1+3\alpha\mu)}{1+\mu+3\alpha\mu} \right], \tag{3.57}$$

$$f_{11}^1 = B_{10}^0 \left[\frac{\alpha_1^0 (1+3\alpha\mu)}{1+\mu+3\alpha\mu} \right], \tag{3.58}$$

and

$$e_{10}^1 = A_{10}^0 \left[\frac{\alpha_1^0 (1+3\alpha\mu)}{1+\mu+3\alpha\mu} \right]. \tag{3.59}$$

If the slip coefficient $\alpha = 0$, this result is matching with the one obtained by Pak, Feng, Stone [21].

3.3.1 Stationary drop

If we assume that the drop is stationary, the first order drag force is given by

$$\vec{D}_1 = 2\pi \left[\frac{2Ma\lambda_e^2 f_2(\lambda_i) g_1(\lambda_e)}{(W+\mu Z+\alpha G)} (E_{11}^1\hat{i} + F_{11}^1\hat{j} + E_{10}^1\hat{k}) e^{\lambda_e^2 t} \right]. \tag{3.60}$$

3.4 Second-order correction

The second order surfactant transport equation is given by

$$Pr_s \frac{\partial \Gamma_2}{\partial t} + \vec{\nabla}_s.(\Gamma_0 \vec{u}_{1s} + \Gamma_1 \vec{u}_{0s}) = \vec{\nabla}_s^2 \Gamma_1, \tag{3.61}$$

where $\vec{u}_{0s}$, $\vec{u}_{1s}$ are the zeroth order and first order tangential velocity components on the drop surface respectively. Assuming that the surfactant concentration is oscillatory, i.e., $\Gamma_2(\theta, \phi, t) = \Gamma_2(\theta, \phi) e^{-i\omega_2 t} = \Gamma_2(\theta, \phi) e^{-l_2^2 t/Pr_s}$, Eq. (3.61) reduces to

$$(\vec{\nabla}_s^2 + l_2^2)\Gamma_2 = \vec{\nabla}_s.(\Gamma_0 \vec{u}_{1s} + \Gamma_1 \vec{u}_{0s}). \tag{3.62}$$

In order to obtain the second order surfactant concentration, Γ_2, we adopt a similar procedure that is used in Section. (3.3). We express Γ_2 in terms of spherical harmonics, i.e.,

$$\Gamma_2 = \sum_{n=1}^{\infty} R_n^2(\theta, \phi) e^{-l_2^2 t/Pr_s}, \tag{3.63}$$

where

$$R_n^2(\theta, \phi) = \sum_{m=0}^{n} \left(E_{nm}^2 \cos\, m\phi + F_{nm}^2 \sin\, m\phi \right) P_n^m(\cos\, \theta), \tag{3.64}$$

and E_{nm}^2, F_{nm}^2 have to be determined such that Γ_2 satisfies the Eq.(3.62). Correspondingly, the coefficients $R_n^2(\theta, \phi)$ can be determined as follows:

$$E_{kj}^2 \pi \frac{2(k+j)!}{(2k+1)(k-j)!} e^{-l_2^2 t/Pr_s} = \frac{-1}{k(k+1) - l_2^2} \int_{\phi=0}^{2\pi} \int_{\theta=0}^{\pi} (\vec{\nabla}_s.(\Gamma_0 \vec{u}_{1s} + \Gamma_1 \vec{u}_{0s})) P_k^j(\cos\, \theta) \cos\, j\phi \sin\, \theta \, d\theta \, d\phi, \tag{3.65}$$

$$F_{kj}^2 \pi \frac{2(k+j)!}{(2k+1)(k-j)!} e^{-l_2^2 t/Pr_s} = \frac{-1}{k(k+1) - l_2^2} \int_{\phi=0}^{2\pi} \int_{\theta=0}^{\pi} (\vec{\nabla}_s.(\Gamma_0 \vec{u}_{1s} + \Gamma_1 \vec{u}_{0s})) P_k^j(\cos\, \theta) \sin\, j\phi \sin\, \theta \, d\theta \, d\phi, \tag{3.66}$$

which implies $-l_2^2/Pr_s = \lambda_e^2$, and

$$\begin{aligned} E_{kj}^2 &= \left[-k\hat{\alpha}_k^2 + \hat{\beta}_k^2 \left((k+1) g_k(\lambda_e) - \lambda_e g_{k+1}(\lambda_e) \right) \right] \left[E_{kj}^1 \frac{k(k+1)}{k(k+1) + \lambda_e^2 Pr_s} \right] - \frac{(2k+1)(k-j)!}{2\pi(k+j)!} \\ &\times \frac{e^{-\lambda_e^2 t}}{k(k+1) + \lambda_e^2 Pr_s} \int_{\phi=0}^{2\pi} \int_{\theta=0}^{\pi} (\vec{\nabla}_s.(\Gamma_1 \vec{u}_{0s})) P_k^j(\cos\, \theta) \cos\, j\phi \sin\, \theta \, d\theta \, d\phi, \end{aligned} \tag{3.67}$$

$$\begin{aligned} F_{kj}^2 &= \left[-k\hat{\alpha}_k^2 + \hat{\beta}_k^2 \left((k+1) g_k(\lambda_e) - \lambda_e g_{k+1}(\lambda_e) \right) \right] \left[F_{kj}^1 \frac{k(k+1)}{k(k+1) + \lambda_e^2 Pr_s} \right] - \frac{(2k+1)(k-j)!}{2\pi(k+j)!} \\ &\times \frac{e^{-\lambda_e^2 t}}{k(k+1) + \lambda_e^2 Pr_s} \int_{\phi=0}^{2\pi} \int_{\theta=0}^{\pi} (\vec{\nabla}_s.(\Gamma_1 \vec{u}_{0s})) P_k^j(\cos\, \theta) \sin\, j\phi \sin\, \theta \, d\theta \, d\phi. \end{aligned} \tag{3.68}$$

Evaluating the double integral on the right hand side is difficult for any given arbitrary flow. However, these can be evaluated for a given ambient flow so that we have the second order concentration. Accordingly, we compute these double integrals for specific cases like uniform flow, Couette flow etc.

Once we obtain the second order concentration for a given flow, one can solve the above equations by following similar procedure that is used to solve the zeroth and first order equations. The second order drag is given by

$$\vec{D}_2 = 2\pi\left[\frac{Y+\mu X+\alpha P}{W+\mu Z+\alpha G}(-\mathbf{U}_2)+\frac{2Ma\lambda_e^2 f_2(\lambda_i)g_1(\lambda_e)}{(W+\mu Z+\alpha G)}\right.$$
$$\left.\times\,(E_{11}^2\hat{i}+F_{11}^2\hat{j}+E_{10}^2\hat{k})e^{\lambda_e^2 t}\right]. \tag{3.69}$$

The force balance $M\frac{d\mathbf{U}_2}{dt}=\vec{D}_2$ leads to

$$\mathbf{U}_2 = \frac{3}{2\rho_i+\rho_e}\left[\frac{2Ma\lambda_e^2 f_2(\lambda_i)g_1(\lambda_e)}{(W+\mu Z+\alpha G)}(E_{11}^2\hat{i}+F_{11}^2\hat{j}+E_{10}^2\hat{k})e^{-\lambda_e^2 t}\right]$$
$$\left(\frac{3}{2\rho_i+\rho_e}\frac{Y+\mu X+\alpha P}{W+\mu Z+\alpha G}+\lambda_e^2\right)^{-1}, \tag{3.70}$$

where X,Y,P,G,Z,W,U,V,H etc., are given in the Appendix B. We therefore, conclude that the second order migration velocity and drag depend only on three modes of the concentration namely, E_{11}^2,F_{11}^2 and E_{10}^2. If we consider the special case of steady flow past a drop, i.e., $\lambda_e=\lambda_i=0$, the second order migration velocity reduces to

$$\mathbf{U}_2=\frac{2Ma}{6+9\mu+18\alpha\mu_i}(e_{11}^2\hat{i}+f_{11}^2\hat{j}+e_{10}^2\hat{k}), \tag{3.71}$$

where

$$e_{nm}^2=\frac{-1}{n(n+1)}\int_{\phi=0}^{2\pi}\int_{\theta=0}^{\pi}(\vec{\nabla}_s.(\Gamma_0\vec{u}_{1s}+\Gamma_1\vec{u}_{0s}))P_n^m(\cos\,\theta)\cos\,m\phi\sin\,\theta\,d\theta\,d\phi, \tag{3.72}$$

$$f_{nm}^2=\frac{-1}{n(n+1)}\int_{\phi=0}^{2\pi}\int_{\theta=0}^{\pi}(\vec{\nabla}_s.(\Gamma_0\vec{u}_{1s}+\Gamma_1\vec{u}_{0s}))P_n^m(\cos\,\theta)\sin\,m\phi\sin\,\theta\,d\theta\,d\phi. \tag{3.73}$$

If the slip coefficient $\alpha=0$, this result also agrees with the one that is obtained by Pak et al. [21].

3.4.1 Stationary drop

If we assume that the drop is stationary, the second order drag force is given by

$$\vec{D}_2=2\pi\left[\frac{2Ma\lambda_e^2 f_2(\lambda_i)g_1(\lambda_e)}{(W+\mu Z+\alpha G)}(E_{11}^2\hat{i}+F_{11}^2\hat{j}+E_{10}^2\hat{k})e^{\lambda_e^2 t}\right]. \tag{3.74}$$

4 Results and discussion

Now, we present important observations with reference to some special cases such as uniform ambient flow, Couette flow, etc.

4.1 Uniform ambient flow

Consider a uniform flow along the $x-$axis, past a liquid drop of unit radius whose center is at its origin. In this case, $\vec{u}_\infty=\vec{u}_{0\infty}=\hat{i}e^{\lambda_e^2 t}$.

Therefore, the corresponding scalar functions χ_0^∞ and η_0^∞ are given by

$$\chi_0^\infty=\frac{1}{2}r\sin\,\theta\cos\,\phi e^{\lambda_e^2 t},\quad \eta_0=0.$$

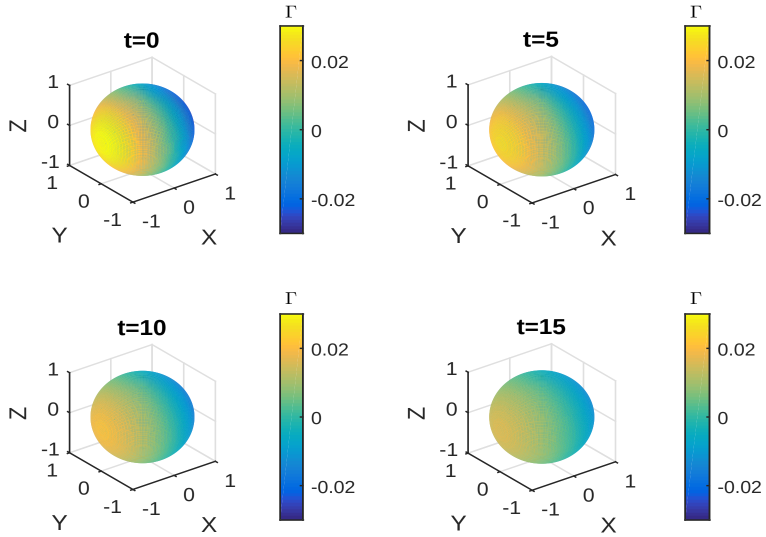

Figure 2: Variation of first order surfactant distribution with the time t corresponding to uniform flow, with $\lambda_e^2 = -0.04$, $\lambda_i^2 = -0.04$, $\mu = 5$ $Ma = 400$ and $\alpha = 0.1$.

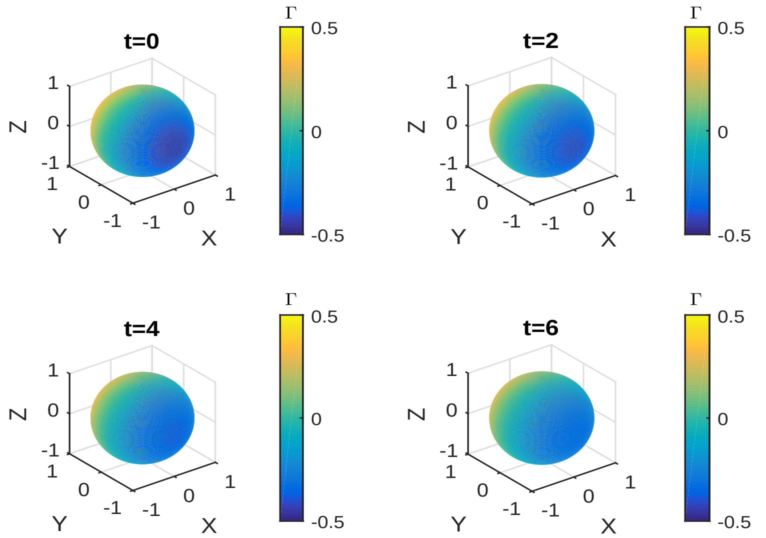

Figure 3: Variation of second order surfactant distribution with the time t corresponding to uniform flow, with $\lambda_e^2 = -0.04$, $\lambda_i^2 = -0.04$, $\mu = 5$ $Ma = 400$ and $\alpha = 0.1$.

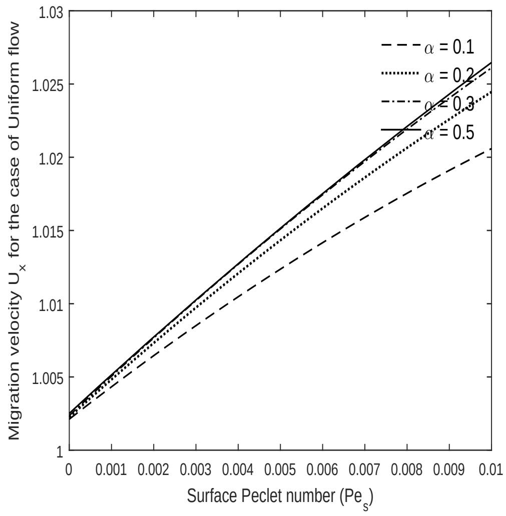

Figure 4: Variation of migration velocity with Pe_s for different slip parameters corresponding to uniform flow, α, $\lambda_e^2 = -0.01$, $\lambda_i^2 = -0.01$, $Ma = 400$ and $\mu = 5$.

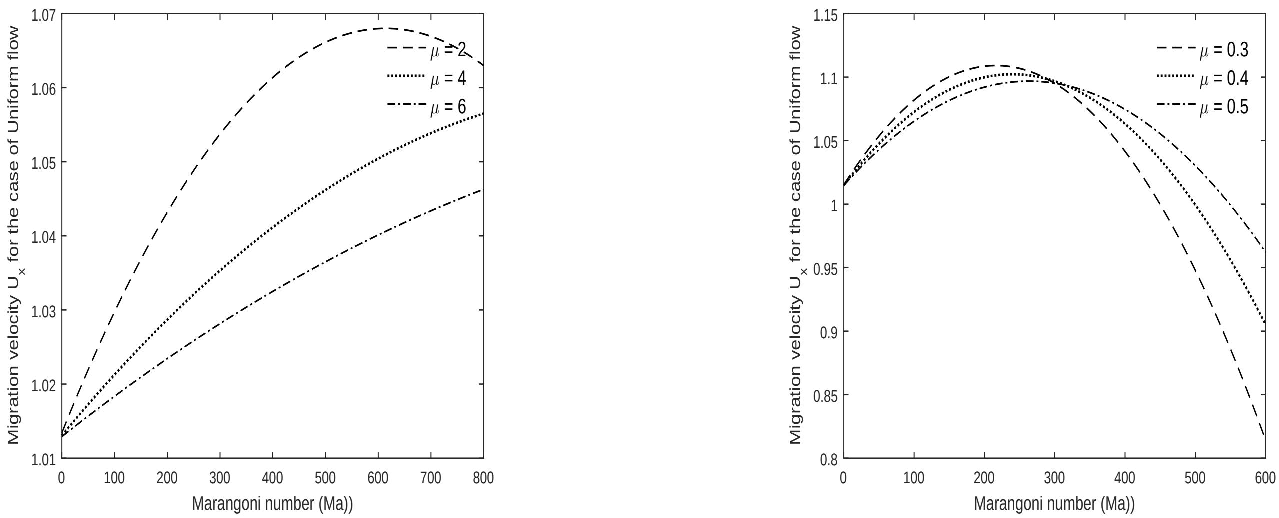

Figure 5: Variation of migration velocity with Marangoni number (Ma) for different viscosity ratios corresponding to uniform flow, μ, $\lambda_e^2 = -0.04$, $\lambda_i^2 = -0.04$, $\alpha = 0.2$ and $Pe_s = 0.01$.

The above choice indicates that $\alpha_1^0 = \frac{1}{2}$, $\beta_1^0 = 0$, $\gamma_1^0 = 0$ in Eqs. (3.16) and (3.17). Therefore the corresponding drag on the spherical drop is given by

$$\vec{D} = \vec{D_0} + Pe_s \vec{D_1} + Pe_s^2 \vec{D_2} + O(Pe_s^3), \tag{4.1}$$

where

$$\vec{D}_0 = 2\pi \left[\frac{Y + \mu X + \alpha P}{W + \mu Z + \alpha G} \right] e^{\lambda_e^2 t} \hat{i}, \tag{4.2}$$

$$\vec{D}_1 = 2\pi \left[\frac{Y + \mu X + \alpha P}{W + \mu Z + \alpha G} (-\mathbf{U}_1) + \frac{2Ma\lambda_e^2 f_2(\lambda_i) g_1(\lambda_e)}{(W + \mu Z + \alpha G)} E_{11}^1 \hat{i} e^{\lambda_e^2 t} \right], \tag{4.3}$$

$$\vec{D}_2 = 2\pi \left[\frac{Y + \mu X + \alpha P}{W + \mu Z + \alpha G} (-\mathbf{U}_2) + \frac{2Ma\lambda_e^2 f_2(\lambda_i) g_1(\lambda_e)}{(W + \mu Z + \alpha G)} E_{11}^2 \hat{i} e^{\lambda_e^2 t} \right]. \tag{4.4}$$

Here

$$E_{11}^1 = \frac{2}{(2 + \lambda_e^2 Pr_s)} \frac{\left(3g_1(\lambda_e)\lambda_e^2 \left(f_2(\lambda_i) + \alpha\mu \left(-2f_2(\lambda_i) + f_1(\lambda_i)\lambda_i\right)\right)\right)}{\delta_1}, \tag{4.5}$$

where

$$\begin{aligned} \delta_1 \quad = \quad & \left(2 \left(g_1(\lambda_e)\lambda_e^2 \left(f_2(\lambda_i) + \alpha\mu \left(-2f_2(\lambda_i) + f_1(\lambda_i)\lambda_i\right)\right) + g_2(\lambda_e)\lambda_e \left(f_1(\lambda_i)\mu\lambda_i \left(1 + 2\alpha\right)\right.\right.\right. \\ & \left. - 2f_2(\lambda_i) \left(-1 + \mu + 2\alpha\mu\right)\right) + 3g_1(\lambda_e) \left(-f_1(\lambda_i)\mu\lambda_i \left(1 + 2\alpha\right)\right. \\ & \left.\left.\left. + 2f_2(\lambda_i) \left(-1 + \mu + 2\alpha\mu\right)\right)\right)\right), \end{aligned} \tag{4.6}$$

and

$$\begin{aligned} E_{11}^2 \quad = \quad & \frac{2E_{11}Ma}{(2 + \lambda_e^2 Pr_s)} \left(f_2(\lambda_i) \left(-3g_1(\lambda_e) + g_2(\lambda_e)\lambda_e\right)\right) / \left(g_1(\lambda_e)\lambda_e^2 \left(-f_2(\lambda_i) + \alpha\mu \left(2f_2(\lambda_i) - f_1(\lambda_i)\lambda_i\right)\right)\right. \\ & + 3g_1(\lambda_e) \left(f_1(\lambda_i)\mu\lambda_i \left(1 + 2\alpha\right) - 2f_2(\lambda_i) \left(-1 + \mu + 2\alpha\mu\right)\right) \\ & \left. + g_2(\lambda_e)\lambda_e \left(-f_1(\lambda_i)\mu\lambda_i \left(1 + 2\alpha\right) + 2f_2(\lambda_i) \left(-1 + \mu + 2\alpha\mu\right)\right)\right). \end{aligned} \tag{4.7}$$

The migration velocity is given by

$$\mathbf{U} = \mathbf{U}_0 + Pe_s \mathbf{U}_1 + Pe_s^2 \mathbf{U}_2 + O(Pe_s^3). \tag{4.8}$$

In this case the zeroth order migration velocity $\boldsymbol{U}_0$, given in Eq. (3.31) reduces to

$$\mathbf{U}_0 = \frac{3}{2\rho_i + \rho_e} \left[\frac{Y + \mu X + \alpha P}{W + \mu Z + \alpha G} \right] \left(\frac{3}{2\rho_i + \rho_e} \frac{Y + \mu X + \alpha P}{W + \mu Z + \alpha G} + \lambda_e^2 \right)^{-1} [\vec{v}_{0\infty}]_0, \tag{4.9}$$

where $\vec{v}_{0\infty}$ can be obtained from the relation

$$\begin{aligned} [\vec{u}_{0\infty}]_0 \quad & = \quad [\vec{v}_{0\infty}]_0 - \mathbf{U}_0 \\ & = \quad \left(1 - \frac{3}{2\rho_i + \rho_e} \left[\frac{Y + \mu X + \alpha P}{W + \mu Z + \alpha G} \right] \left(\frac{3}{2\rho_i + \rho_e} \frac{Y + \mu X + \alpha P}{W + \mu Z + \alpha G} + \lambda_e^2 \right)^{-1} \right) \\ & \quad\quad \times [\vec{v}_{0\infty}]_0, \end{aligned} \tag{4.10}$$

which implies,

$$\begin{aligned} [\vec{v}_{0\infty}]_0 \quad = \quad & \left[1 - \frac{3}{2\rho_i + \rho_e} \left[\frac{Y + \mu X + \alpha P}{W + \mu Z + \alpha G} \right] \left(\frac{3}{2\rho_i + \rho_e} \frac{Y + \mu X + \alpha P}{W + \mu Z + \alpha G} + \lambda_e^2 \right)^{-1} \right]^{-1} \\ & e^{\lambda_e^2 t} \hat{i}. \end{aligned} \tag{4.11}$$

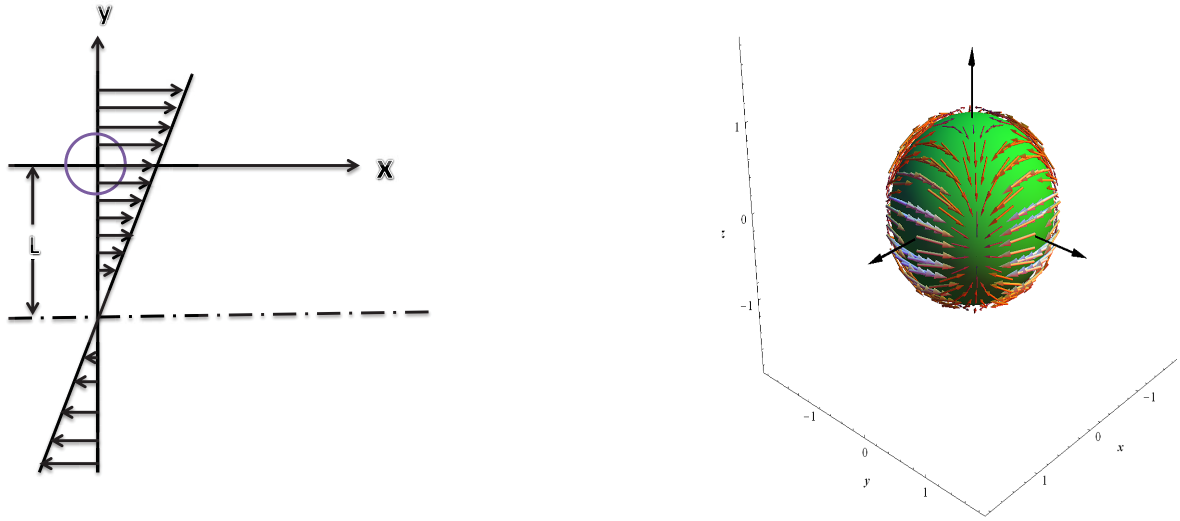

Figure 6: a) Geometry of the problem and velocity vector corresponding to Coutte ambient flow, b) surface velocity vector field corresponding to Coutte flow

The first order migration velocity $\mathbf{U}_1$, given in (3.50) reduces to

$$\mathbf{U}_1 = \frac{3}{2\rho_i + \rho_e}\left[\frac{2Ma\lambda_e^2 f_2(\lambda_i) g_1(\lambda_e)}{(W + \mu Z + \alpha G)}\right]\left(\frac{3}{2\rho_i + \rho_e}\frac{Y + \mu X + \alpha P}{W + \mu Z + \alpha G} + \lambda_e^2\right)^{-1} E_{11}^1 e^{\lambda_e^2 t}\hat{i},$$

and the second order migration velocity $\boldsymbol{U}_2$, given in (3.70) reduces to

$$\mathbf{U}_2 = \frac{3}{2\rho_i + \rho_e}\left[\frac{2Ma\lambda_e^2 f_2(\lambda_i) g_1(\lambda_e)}{(W + \mu Z + \alpha G)}\right]\left(\frac{3}{2\rho_i + \rho_e}\frac{Y + \mu X + \alpha P}{W + \mu Z + \alpha G} + \lambda_e^2\right)^{-1} E_{11}^2 e^{\lambda_e^2 t}\hat{i}.$$

It may be noted that the corresponding migration velocity is only along the flow direction and avoids cross migration.

We show the variation of first and second order surfactant distributions with time (figures 2 and 3). Here, we have noticed that, the surfactant concentration decreases with time.

For a fixed viscosity ratio, the slip parameter reduces the resistance offered by the drop. Accordingly, the migration velocity increases same is observed in figure (4). It may be noted that surface Péclet number measures the importance of convection relative to diffusion. Therefore, as Pe_s increases, migration velocity increases. The same is observed in figure (4). As the viscosity ratio is increasing, the drop behaves like a solid and hence, the migration velocity decreases.

It may be noted that Marangoni number is the ratio of surface tension forces to viscous forces. Therefore, for small viscosity ratios, with increasing Ma, the surface forces dominate and hence, the drag force increases with the increase of Marangoni number. Accordingly, the migration velocity decreases with Marangoni number. But, for large viscosity ratios, the viscous forces dominates the surface forces. And hence, migration velocity increases with increasing Marangoni number. The same is observed in figure (5).

We have observed in figures (2) and (3) as time increases, the both first and second order surfactant concentration decreases as expected.

4.2 Couette flow

Consider a Couette flow past a liquid drop of unit radius whose center is at the origin (see Fig. 6). In this case, $\vec{v}_\infty = (F(y+L)\hat{i})e^{\lambda_e^2 t}$, where L is the distance of the center of the droplet from the point of zero velocity and F is the shear (Ref. [22]).

Therefore the corresponding scalar functions χ_0^∞ and η_0^∞ are given by

$$\chi_0^\infty = \left(\frac{FL}{2} r P_1^1(\cos\theta)\cos\phi + \frac{F}{36} r^2 \sin 2\phi P_2^2(\cos\theta)\right) e^{\lambda_e^2 t}, \quad \eta_0 = 0.$$

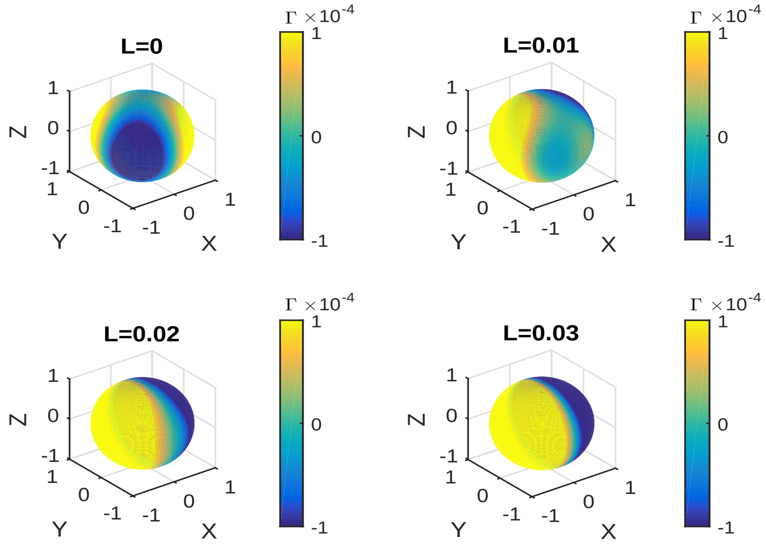

Figure 7: Variation of first order surfactant distribution for different time values corresponding to Coutte flow, with $\lambda_e^2 = -0.04$, $\lambda_i^2 = -0.04$, $\mu = 5$,$Ma = 400$, $F = 1$, $t = 1$, $Pe = 0.01$ and $\alpha = 0.1$.

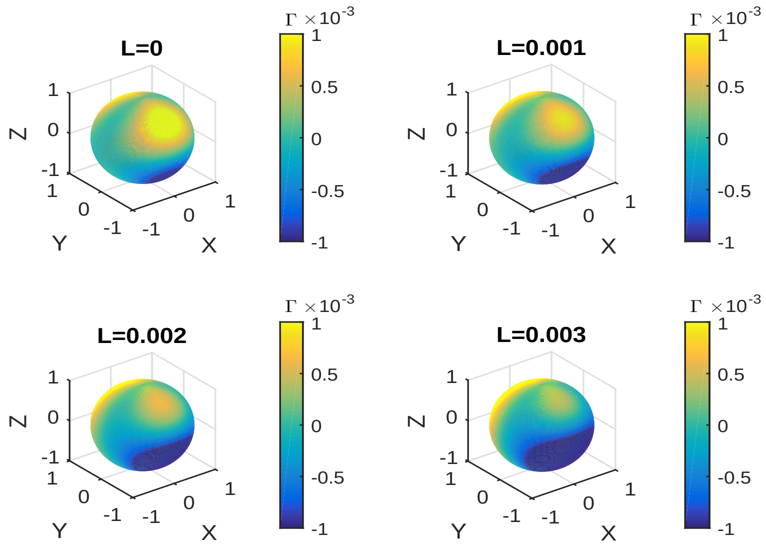

Figure 8: Variation of second order surfactant distribution for different time values corresponding to Coutte flow, with $\lambda_e^2 = -0.04$, $\lambda_i^2 = -0.04$, $\mu = 5$,$Ma = 400$, $F = 1$, $t = 1$, $Pe = 0.01$ and $\alpha = 0.1$.

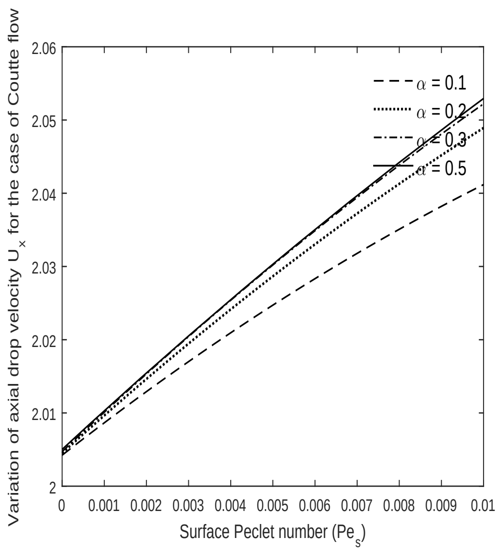

Figure 9: Variation of migration velocity with Pe_s for different slip parameters corresponding to Coutte flow, α, $\lambda_e^2 = -0.01$, $\lambda_i^2 = -0.01$, $Ma = 400$, $F = 1$, $L = 2$ and $\mu = 5$.

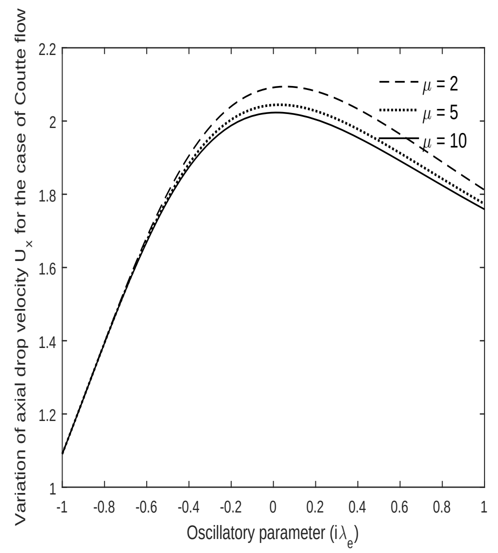

Figure 10: Variation of migration velocity with $i\lambda_e$ for different viscosity ratios corresponding to Coutte flow, α, $F = 1$, $L = 2$, $\lambda_i^2 = -0.04$, $Ma = 400$ and $\mu = 5$.

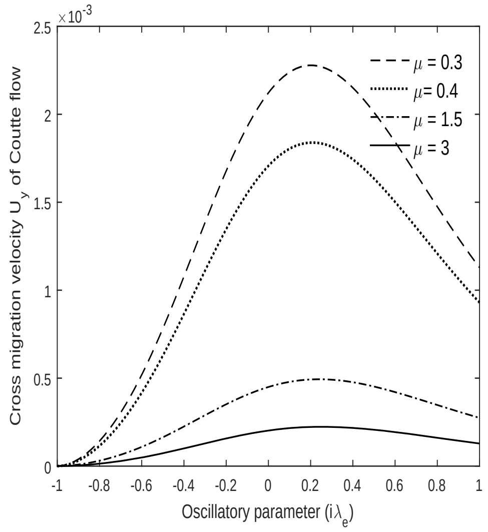

Figure 11: Variation of cross migration velocity with $i\lambda_e$ for different viscosity ratios corresponding to Coutte flow, μ, with $\lambda_i^2 = -0.04$, $\alpha = 0.2$, $Ma = 400$, $F = 1$, $L = 2$ and $Pe = 0.01$.

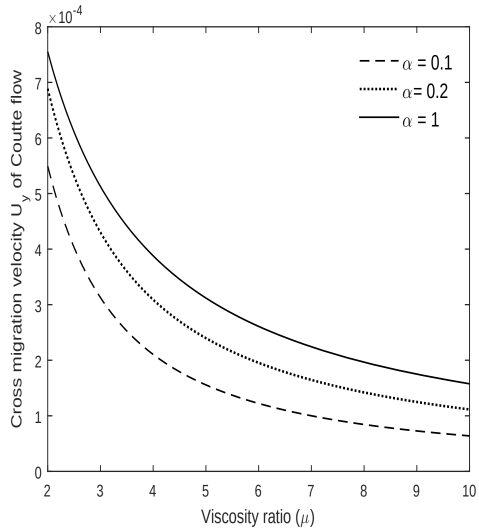

Figure 12: Variation of cross migration velocity with viscosity ratio, μ, for different α corresponding to Coutte flow with $\lambda_e^2 = -0.04$, $\lambda_i^2 = -0.04$, $\alpha = 0.1$, $Ma = 400$, $F = 1$, $L = 4$ and $Pe = 0.01$.

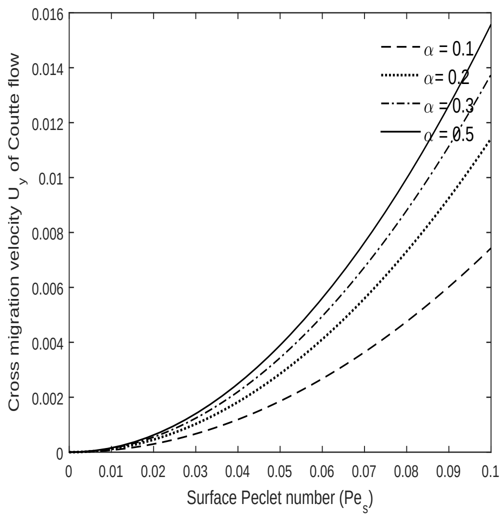

Figure 13: Variation of cross migration velocity with Pe_s for different slip parameters corresponding to Coutte flow, α, with $\lambda_e^2 = -0.04$, $\lambda_i^2 = -0.04$, $Ma = 400$, $F = 1$, $L = 2$ and $\mu = 5$.

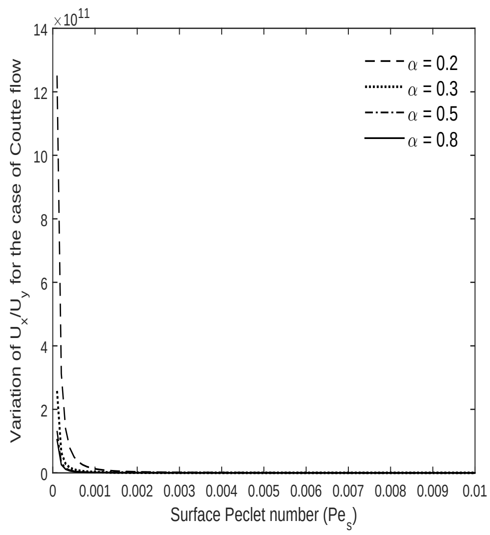

Figure 14: Variation of U_x/U_y with Pe_s for different slip parameters corresponding to Coutte flow, α, with $\lambda_e^2 = -0.04$, $\lambda_i^2 = -0.04$, $Ma = 400$, $F = 1$, $L = 2$ and $\mu = 5$.

The above choice indicates that $\alpha_1^0 = \frac{FL}{2}$, $\alpha_2^0 = \frac{F}{36}$, $\beta_1^0 = 0$, $\gamma_1^0 = 0$ in Eqs. (3.16) and (3.17). Therefore the corresponding surfactant concentration distribution on the spherical drop is given by

$$\Gamma = \Gamma_0 + Pe_s\Gamma_1 + Pe_s^2\Gamma_2 + O(Pe_s^3), \tag{4.12}$$

where

$$\Gamma_0 = 1, \tag{4.13}$$

$$\Gamma_1 = \left(-E_{11}^1 \sin\theta\cos\phi + F_{22}^1 \sin 2\phi P_2^2(\cos\theta)\right) e^{\lambda_e^2 t}, \tag{4.14}$$

$$\begin{aligned}\Gamma_2 \;=\;& \big(-E_{11}^2 \sin\theta\cos\phi - F_{11}^2\sin\theta\sin\phi + F_{22}^2 \sin 2\phi P_2^2(\cos\theta) \\ &+ F_{31}^2\sin\phi P_3^1(\cos\theta) + F_{33}^2 \sin 3\phi P_3^3(\cos\theta) + E_{20}^2 P_2^0(\cos\theta) \\ &+ E_{22}^2\cos 2\phi P_2^2(\cos\theta) + E_{40}^2 P_4^0(\cos\theta) + E_{44}^2\cos 4\phi P_4^4(\cos\theta)\big) \\ & e^{\lambda_e^2 t},\end{aligned} \tag{4.15}$$

where few quantities E_{11}^1, F_{22}^1 etc. are listed in the Appendix (D).

For an unbounded Coutte flow, the migration velocity of a force free drop is calculated as

$$\mathbf{U} = \mathbf{U}_0 + Pe_s\mathbf{U}_1 + Pe_s^2\mathbf{U}_2 + O(Pe_s^3). \tag{4.16}$$

In this case the zeroth order migration velocity $\mathbf{U}_0$, given in Eq. (3.31) reduces to

$$\mathbf{U}_0 = \frac{3}{2\rho_i+\rho_e}\left[\frac{Y+\mu X+\alpha P}{W+\mu Z+\alpha G}\right]\left(\frac{3}{2\rho_i+\rho_e}\frac{Y+\mu X+\alpha P}{W+\mu Z+\alpha G}+\lambda_e^2\right)^{-1} FLe^{\lambda_e^2 t}\hat{\imath}, \tag{4.17}$$

The first order migration velocity $\mathbf{U}_1$, given in (3.50) reduces to

$$\mathbf{U}_1 = \frac{3E_{11}^1\hat{\imath}}{2\rho_i+\rho_e}\left[\frac{2Ma\lambda_e^2 f_2(\lambda_i)g_1(\lambda_e)}{(W+\mu Z+\alpha G)}\right]\left(\frac{3}{2\rho_i+\rho_e}\frac{Y+\mu X+\alpha P}{W+\mu Z+\alpha G}+\lambda_e^2\right)^{-1} e^{\lambda_e^2 t}, \tag{4.18}$$

and the second order migration velocity $\mathbf{U}_2$, given in (3.70) reduces to

$$\mathbf{U}_2 = \frac{3(E_{11}^2\hat{\imath}+F_{11}^2\hat{\jmath})}{2\rho_i+\rho_e}\left[\frac{2Ma\lambda_e^2 f_2(\lambda_i)g_1(\lambda_e)}{(W+\mu Z+\alpha G)}\right]\left(\frac{3}{2\rho_i+\rho_e}\frac{Y+\mu X+\alpha P}{W+\mu Z+\alpha G}+\lambda_e^2\right)^{-1} e^{\lambda_e^2 t}. \tag{4.19}$$

By symmetry, it is expected that there will be no velocity component in z direction. In case if there is a cross migration (i.e., motion transverse to the flow direction), the same occurs towards the center line and should be in y direction (see Fig. (6)). In [21], a detailed explanation on cross migration of a surfactant coated viscous drop in Poiseuille flow is presented. Similar arguments followed in [10] while discussing migration of deformed drop with interfacial slip in in an unbounded Poiseuille flow. We also follow similar arguments to show that cross migration occurs only at second order with respect to the expansion of migration velocity in terms of surface Péclet number. Here, we have observed that, at leading order, we recover the case of clean spherical drop in an unbounded Couette flow (characterized by the velocity scale U_c). It is well known that, there can be no cross stream migration in the absence of inertia, deformation, and surfactant concentration. Accordingly, we observe that at leading order, there is no cross stream migration. This phenomena can also be supported mathematically as follows: The dimensional zeroth order migration velocity $\mathbf{U}_0^* = \mathbf{U}_0 U_c \propto U_c$. If there is a cross stream migration, for a reversal of the background flow direction ($U_c \to -U_c$), $\mathbf{U}_0^*$ also should change the direction. But, the cross stream migration should occur towards the center line. Therefore, there is no cross stream migration in leading order, which implies the symmetry condition at leading order is satisfied (Ref. [21]).

With the similar argument which is given for leading order migration velocity, we can say at first order also there can not be cross stream migration. We observe the dimensional first order migration velocity $\mathbf{U}_1^* = Pe_s \mathbf{U}_1 U_c \propto Pe_s Ma E_{11}^1 U_c \propto Pe_s Ma U_c$, and the product $Pe_s Ma$ is independent of U_c. Therefore, $\mathbf{U}_1^* \propto U_c$. If there is a cross migration velocity, $\mathbf{U}_1^*.\hat{j} \propto U_c$, which violates the symmetry requirement that the cross stream migration direction remains same upon reversal of the background flow direction. Lateral migration is therefore expected not to occur at leading and first order.

Observing second order migration velocity, we see $\mathbf{U}_2^*.\hat{i} = Pe_s^2 \mathbf{U}_2 U_c.\hat{i} \propto Pe_s^2 Ma E_{11}^2 U_c \propto Pe_s^2 Ma^2 U_c \propto U_c$, and $\mathbf{U}_2^*.\hat{j} = Pe_s^2 \mathbf{U}_2 U_c.\hat{j} \propto Pe_s^2 Ma F_{11}^2 U_c \propto Pe_s^2 Ma U_c \propto U_c^2$. Therefore, the transverse migration is invariant upon reversal of the direction of the ambient Coutte flow.

We also noted that, the transverse migration is linearly dependent on L (as $E_{11}^1 \propto L$) which respects the symmetry requirement that the transverse migration direction should reverse its sign when the drop is placed at the same distance but on the opposite side with respect to the center of the Coutte flow (see Fig. 6). The same is noted by Pak et al. [21] for the case of Poiseuille flow.

The first order and second order surfactant distributions are plotted with specific values of parameters for visualization in figures (7) and (8). Here, we have seen as L increases, the concentration increases (since $E_{11}^1 \propto L$).

We have observed the variation of axial migration velocity and cross-stream migration velocity for different parameters. The variation of axial migration is observed in figures (9) and (10). It can be seen that, we have the cross migration due to the linear term present in the ambient velocity of Coutte flow. And the migration in the axial direction is due to the constant term present in the Coutte flow. As a consequence, axial migration velocity in the case of Coutte flow behaves in the same manner as the migration velocity in the case uniform ambient flow.

We have observed the variation of cross migration velocity with the amplification factor λ_e in figure (11). Drop cross migration is oscillating with the amplification factor. From Fig. (12), we have seen that, with the increasing viscosity ratio, the magnitude of cross migration decreases as expected (as the viscosity ratio increases, the drop behaves like a solid).

It may be noted that surface Péclet number measures the importance of convection relative to diffusion. Therefore, as Pe_s increases, magnitude of migration velocity increases. Also, for a fixed viscosity ratio, the slip parameter reduces the resistance offered by the drop. Accordingly, the migration velocity increases same is observed in figure (13). From figure (14), we have observed the ratio of U_x and U_y decreases. From this, we can say, U_y increases faster than U_x with Péclet number.

4.3 Poiseuille flow

Consider a Poiseuille flow past a liquid drop of unit radius whose center is at origin (Ref. [21, 22] to see the geometrical setup of the problem). In this case, we calculated the ambient velocity as $\vec{v}_\infty = \vec{v}_{0\infty} = -\hat{k} e^{\lambda_e^2 t}\left(1 - \frac{J_0(i\lambda_e R)}{J_0(i\lambda_e R_0)}\right)\left(1 - \frac{1}{J_0(i\lambda_e R_0)}\right)^{-1}$. Here $R^2 = r^2 \sin^2\theta + b^2 + 2br\sin\theta\cos\phi$, velocity is non-dimensionalized with the characteristic velocity U_b, which is at a dimensionless distance b from the drop, and R_0 is the dimensionless distance to the point of zero velocity of the flow, λ_e is the amplification factor. We expanded $\vec{v}_\infty$ as series form for small λ_e to get χ_0^∞ and η_0^∞, which are given by

$$\chi_0^\infty = \left[\beta_{12} f_1(\lambda_e r) S_1(\theta,\phi) + \sum_{n=1}^{\infty} \alpha_{n2} r^n S_n(\theta,\phi)\right] e^{\lambda_e^2 t}, \quad \eta_0^\infty = 0,$$

where,

$$\alpha_1^0 = -\frac{1}{2}\left(1 - \frac{1}{J_0(i\lambda_e R_0)}\right)^{-1}\left(1 - \frac{b^2\lambda_e^2}{4J_0(i\lambda_e R_0)}\right), \tag{4.20}$$

$$\alpha_2^0 = -\left(1 - \frac{1}{J_0(i\lambda_e R_0)}\right)^{-1}\left(\frac{b\lambda_e^2}{36J_0(i\lambda_e R_0)}\right) \tag{4.21}$$

$$\alpha_3^0 = -\left(\frac{\lambda_e^2}{120J_0(i\lambda_e R_0)}\right)\left(1-\frac{1}{J_0(i\lambda_e R_0)}\right)^{-1} \tag{4.22}$$

$$\beta_1^0 = \left(\frac{3}{2\lambda_e J_0(i\lambda_e R_0)}\right)\left(1-\frac{1}{J_0(i\lambda_e R_0)}\right)^{-1} \tag{4.23}$$

Therefore the corresponding surfactant concentration distribution on the spherical drop is given by

$$\Gamma = \Gamma_0 + Pe_s\Gamma_1 + Pe_s^2\Gamma_2 + O(Pe_s^3), \tag{4.24}$$

where

$$\Gamma_0 = 1, \tag{4.25}$$

$$\Gamma_1 = \left(E_{10}^1 \cos\theta + E_{21}^1 \cos\phi P_2^1(\cos\theta) + E_{30}^1 P_3^0(\cos\theta)\right) e^{\lambda_e^2 t}, \tag{4.26}$$

$$\begin{aligned}\Gamma_2 \;=\; & \big(E_{10}^2 \cos\theta + E_{21}^2 \cos\phi P_2^1(\cos\theta) + E_{30}^2 P_3^0(\cos\theta) \\ & + E_{11}^2 P_1^1(\cos\theta)\cos\phi + E_{20}^2 P_2^0(\cos\theta) + E_{22}^2 \cos 2\phi P_2^2(\cos\theta) \\ & + E_{31}^2 \cos\phi P_3^1(\cos\theta) + E_{42}^2 \cos 2\phi P_4^2(\cos\theta) + E_{51}^2 \cos\phi P_5^1(\cos\theta) \\ & + E_{62}^2 \cos 2\phi P_6^2(\cos\theta) + E_{71}^2 \cos\phi P_7^1(\cos\theta) + E_{82}^2 \cos 2\phi P_8^2(\cos\theta) \\ & + E_{52}^2 \cos 2\phi P_5^2(\cos\theta) + E_{40}^2 P_4^0(\cos\theta) + E_{60}^2 P_6^0(\cos\theta)\big) \\ & e^{\lambda_e^2 t},\end{aligned} \tag{4.27}$$

where the constants E_{10}^1, E_{21}^1, etc can be computed Eqs. (3.42), (3.43), (3.67) and (3.68).

For an unbounded Poiseuille flow, the migration velocity of a force free drop is calculated as

$$\mathbf{U} = \mathbf{U}_0 + Pe_s\mathbf{U}_1 + Pe_s^2\mathbf{U}_2 + O(Pe_s^3). \tag{4.28}$$

In this case the zeroth order migration velocity $\mathbf{U}_0$, given in Eq. (3.31) reduces to

$$\begin{aligned}\mathbf{U}_0 \;=\; & \frac{3}{2\rho_i+\rho_e}\left[\frac{Y+\mu X+\alpha P}{W+\mu Z+\alpha G}[\vec{v}_{0\infty}]_0 + \frac{V+\mu U+\alpha H}{W+\mu Z+\alpha G}[\nabla^2\vec{v}_{0\infty}]_0\right] \\ & \left(\frac{3}{2\rho_i+\rho_e}\frac{Y+\mu X+\alpha P}{W+\mu Z+\alpha G}+\lambda_e^2\right)^{-1},\end{aligned} \tag{4.29}$$

where

$$[\vec{v}_{0\infty}]_0 \;=\; \left(\frac{I_0(b\lambda_e) - J_0(i\lambda_e R_0)}{(1-J_0(\lambda_e R_0))J_0(i\lambda_e R_0)}\right) e^{\lambda_e^2 t}\hat{k}. \tag{4.30}$$

and

$$[\nabla^2\vec{v}_{0\infty}]_0 \;=\; \frac{\lambda_e^2 I_0(b\lambda_e)}{I_0(\lambda_e R_0) - I_0(\lambda_e R_0)^2} e^{\lambda_e^2 t}\hat{k}. \tag{4.31}$$

If we consider the limiting case of no oscillations in the hydrodynamic flow field, i.e., $\lambda_i = \lambda_e = 0$, and zero slip coefficient, i.e., $\alpha = 0$, then the zeroth order terminal velocity reduces to

$$\mathbf{U}_0 = \left(1 - \frac{b^2}{R_0^2} - \frac{\mu}{4+6\mu}\frac{4}{R_0^2}\right)\hat{k}, \tag{4.32}$$

which is exactly matching with the one that is obtained by Pak, Feng and Stone [21].

The first order migration velocity $\mathbf{U}_1$, given in (3.50) reduces to

$$\mathbf{U}_1 = \frac{3E_{10}^1\hat{k}}{2\rho_i+\rho_e}\left[\frac{2Ma\lambda_e^2 j_2(\lambda_i)h_1(\lambda_e)}{(W+\mu Z+\alpha G)}\right]\left(\frac{3}{2\rho_i+\rho_e}\frac{Y+\mu X+\alpha P}{W+\mu Z+\alpha G}+\lambda_e^2\right)^{-1} e^{\lambda_e^2 t}, \tag{4.33}$$

and the second order migration velocity $\mathbf{U}_2$, given in (3.70) reduces to

$$\mathbf{U}_2 = \frac{3(E_{11}^2\hat{i}+E_{10}^2\hat{k})}{2\rho_i+\rho_e}\left[\frac{2Ma\lambda_e^2 j_2(\lambda_i)h_1(\lambda_e)}{(W+\mu Z+\alpha G)}\right]\left(\frac{3}{2\rho_i+\rho_e}\frac{Y+\mu X+\alpha P}{W+\mu Z+\alpha G}+\lambda_e^2\right)^{-1} e^{\lambda_e^2 t}. \tag{4.34}$$

Similar to the arguments made in Section (4.2), by symmetry, it is expected that there will be no velocity component in y direction. In case if there is a cross migration, the same occurs towards the center line and should be in x direction (Ref. [21]). We also follow similar arguments to show that, there is no cross stream migration in leading order and first order, which implies the symmetry condition at leading order is satisfied (Ref. [21]).

Similarly, observing second order migration velocity, we see $\mathbf{U}_2^*.\hat{k} = Pe_s^2\mathbf{U}_2U_c.\hat{k} \propto Pe_s^2MaE_{10}^2U_c \propto Pe_s^2Ma^2U_c \propto U_c$, and $\mathbf{U}_2^*.\hat{i} = Pe_s^2\mathbf{U}_2U_c.\hat{i} \propto Pe_s^2MaE_{11}^2U_c \propto Pe_s^2MaU_c \propto U_c^2$. Therefore, the transverse migration is invariant upon reversal of the direction of the ambient Poiseuille flow.

4.4 Validation

We have compared our results with some existing literature to validate our results. These are shown in the Table (1).

5 Conclusions

In this paper, we have considered an arbitrary transient Stokes flow with a given ambient flow past a spherical drop. We analyzed the effects of surface-active agents on the motion of the drop. We have solved the unsteady convection-diffusion equation to find the surfactant transport on the surface of the drop for low surface Péclet number. We have also considered the effects of interfacial slip. We found a closed form expression for drag and migration velocity in terms of Marangoni number, slip parameter and viscosity ratios up to second order in the surface Péclet number, i.e., up to $O(Pe_s^2)$. We have analyzed the variation of surfactants for different viscosity ratios and Marangoni number. We have observed that the impurities residing on the surface do not show much effect on the behavior of the drop for increasing viscosity ratios. We considered various special cases and computed drag and migration velocity up to second order in the surface Péclet number in each case. We have also compared the results with the existing literature for some limiting cases.

6 acknowledgements

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Table 1: Limiting cases of the magnitude of drag force of the present study to get that of different existing literature.

	Main contribution	Limiting cases of current study to get others as listed
Present study	Effect of surfactant concentration and interfacial slip α on the unsteady Stokes flow past a viscous drop for low Pe_s (Surface Péclet numbar).	
Pak et al. [21]	Effect of surfactant concentration on the steady Stokes flow past a viscous drop for low Pe_s (Surface Péclet numbar).	$D(Pe_s, Ma, \mu, \alpha \to 0, Pr_s \to 0, \lambda_e \to 0, \lambda_i \to 0, t) = D_1(Pe_s, Ma, \mu)$
Sharanya and Raja Sekhar [29]	Thermocapillary migration of a spherical drop in an arbitrary transient Stokes flow	$D(Pe_s \to 0, Ma \to 0, \mu, \alpha \to 0, Pr_s, \lambda_e, \lambda_i, t) = D_2(Ma \to 0, \mu, Pr, \lambda_e, \lambda_i, t)$
Choudhuri and Raja Sekhar [7]	Thermocapillary migration of a spherical drop in an arbitrary transient Stokes flow	$D(Pe_s \to 0, Ma \to 0, \mu, \alpha \to 0, Pr_s \to 0, \lambda_e \to 0, \lambda_i \to 0, t) = D_3(Ma \to 0, \mu)$
Choudhuri and Padmavati [6]	Oscillatory Stokes flow past a viscous drop	$D(Pe_s \to 0, Ma \to 0, \mu, \alpha \to 0, Pr_s \to 0, \lambda_e, \lambda_i, t) = D_4(\lambda_e, \lambda_i, \mu)$

A The unknown coefficients (for leading order problem)

The unknown coefficients in (3.20) and (3.23) can be found using the boundary conditions given in (3.7) to (3.12) which are given as follows:

$$
\begin{aligned}
\hat{\alpha}_n^0 \;=\;& \big(-2f_{n+1}(\lambda_i)g_{n+1}(\lambda_e)\alpha_n^0\lambda_e + 2f_{n+1}(\lambda_i)g_2(\lambda_e)\mu\alpha_n^0\lambda_e - 2f_{n+1}(\lambda_e)f_{n+1}(\lambda_i)g_n(\lambda_e)\beta_n^0\lambda_e \\
& - 2f_n(\lambda_e)f_{n+1}(\lambda_i)g_{n+1}(\lambda_e)\beta_n^0\lambda_e + 2f_{n+1}(\lambda_e)f_{n+1}(\lambda_i)g_n(\lambda_e)\mu\beta_n^0\lambda_e + 2f_n(\lambda_e)f_{n+1}(\lambda_i)g_{n+1}(\lambda_e)\mu\beta_n^0\lambda_e \\
& - f_{n+1}(\lambda_i)g_n(\lambda_e)\alpha_n^0\lambda_e^2 - f_n(\lambda_i)g_{n+1}(\lambda_e)\mu\alpha_n^0\lambda_e\lambda_i - f_n(\lambda_i)f_{n+1}(\lambda_e)g_n(\lambda_e)\mu\beta_n^0\lambda_e\lambda_i \\
& - f_n(\lambda_e)f_n(\lambda_i)g_{n+1}(\lambda_e)\mu\beta_n^0\lambda_e\lambda_i + 4f_{n+1}(\lambda_i)g_{n+1}(\lambda_e)\alpha\mu\alpha_n^0\lambda_e + 4f_{n+1}(\lambda_e)f_{n+1}(\lambda_i)g_n(\lambda_e)\alpha\mu\beta_n^0\lambda_e \\
& + 4f_n(\lambda_e)f_{n+1}(\lambda_i)g_{n+1}(\lambda_e)\alpha\mu\beta_n^0\lambda_e + 2f_{n+1}(\lambda_i)g_n(\lambda_e)\alpha\mu\alpha_n^0\lambda_e^2 - 2f_n(\lambda_i)g_{n+1}(\lambda_e)\alpha\mu\alpha_n^0\lambda_e\lambda_i \\
& - 2f_n(\lambda_i)f_{n+1}(\lambda_e)g_n(\lambda_e)\alpha\mu\beta_n^0\lambda_e\lambda_i \\
& - 2f_n(\lambda_e)f_n(\lambda_i)g_{n+1}(\lambda_e)\alpha\mu\beta_n^0\lambda_e\lambda_i - f_n(\lambda_i)g_n(\lambda_e)\alpha\mu\alpha_n^0\lambda_e^2\lambda_i\big) \\
& /\,(-2f_{n+1}(\lambda_i)g_n(\lambda_e) - 4f_{n+1}(\lambda_i)g_n(\lambda_e)n + 2f_{n+1}(\lambda_i)g_n(\lambda_e)\mu + 4f_{n+1}(\lambda_i)g_n(\lambda_e)n\mu \\
& + 2f_{n+1}(\lambda_i)g_{n+1}(\lambda_e)\lambda_e - 2f_{n+1}(\lambda_i)g_{n+1}(\lambda_e)\mu\lambda_e + f_{n+1}(\lambda_i)g_n(\lambda_e)\lambda_e^2 - f_n(\lambda_i)g_n(\lambda_e)\mu\lambda_i \\
& - 2f_n(\lambda_i)g_n(\lambda_e)n\mu\lambda_i + f_n(\lambda_i)g_{n+1}(\lambda_e)\mu\lambda_e\lambda_i + 4f_{n+1}(\lambda_i)g_n(\lambda_e)\alpha\mu + 8f_{n+1}(\lambda_i)g_n(\lambda_e)n\alpha\mu \\
& - 4f_{n+1}(\lambda_i)g_{n+1}(\lambda_e)\alpha\mu\lambda_e - 2f_{n+1}(\lambda_i)g_n(\lambda_e)\alpha\mu\lambda_e^2 - 2f_n(\lambda_i)g_n(\lambda_e)\alpha\mu\lambda_i \\
& - 4f_n(\lambda_i)g_n(\lambda_e)n\alpha\mu\lambda_i + 2f_n(\lambda_i)g_{n+1}(\lambda_e)\alpha\mu\lambda_e\lambda_i + f_n(\lambda_i)g_n(\lambda_e)\alpha\mu\lambda_e^2\lambda_i\big)\,, \qquad \text{(A.1)}
\end{aligned}
$$

$$
\begin{aligned}
\hat{\beta}_n^0 \;=\;& \big(2f_{n+1}(\lambda_i)\alpha_n^0 + 4f_{n+1}(\lambda_i)n\alpha_n^0 - 2f_{n+1}(\lambda_i)\mu\alpha_n^0 - 4f_{n+1}(\lambda_i)n\mu\alpha_n^0 + 2f_n(\lambda_e)f_{n+1}(\lambda_i)\beta_n^0 \\
& + 4f_n(\lambda_e)f_{n+1}(\lambda_i)n\beta_n^0 - 2f_n(\lambda_e)f_{n+1}(\lambda_i)\mu\beta_n^0 - 4f_n(\lambda_e)f_{n+1}(\lambda_i)n\mu\beta_n^0 \\
& + 2f_{n+1}(\lambda_e)f_{n+1}(\lambda_i)\beta_n^0\lambda_e - 2f_{n+1}(\lambda_e)f_{n+1}(\lambda_i)\mu\beta_n^0\lambda_e - f_n(\lambda_e)f_{n+1}(\lambda_i)\beta_n^0\lambda_e^2 \\
& + f_n(\lambda_i)\mu\alpha_n^0\lambda_i + 2f_n(\lambda_i)n\mu\alpha_n^0\lambda_i + f_n(\lambda_e)f_n(\lambda_i)\mu\beta_n^0\lambda_i \\
& + 2f_n(\lambda_e)f_n(\lambda_i)n\mu\beta_n^0\lambda_i + f_n(\lambda_i)f_{n+1}(\lambda_e)\mu\beta_n^0\lambda_e\lambda_i - 4f_{n+1}(\lambda_i)\alpha\mu\alpha_n^0 \\
& - 8f_{n+1}(\lambda_i)n\alpha\mu\alpha_n^0 - 4f_n(\lambda_e)f_{n+1}(\lambda_i)\alpha\mu\beta_n^0 - 8f_n(\lambda_e)f_{n+1}(\lambda_i)n\alpha\mu\beta_n^0 \\
& - 4f_{n+1}(\lambda_e)f_{n+1}(\lambda_i)\alpha\mu\beta_n^0\lambda_e + 2f_n(\lambda_e)f_{n+1}(\lambda_i)\alpha\mu\beta_n^0\lambda_e^2 + 2f_n(\lambda_i)\alpha\mu\alpha_n^0\lambda_i \\
& + 4f_n(\lambda_i)n\alpha\mu\alpha_n^0\lambda_i + 2f_n(\lambda_e)f_n(\lambda_i)\alpha\mu\beta_n^0\lambda_i + 4f_n(\lambda_e)f_n(\lambda_i)n\alpha\mu\beta_n^0\lambda_i \\
& + 2f_n(\lambda_i)f_{n+1}(\lambda_e)\alpha\mu\beta_n^0\lambda_e\lambda_i - f_n(\lambda_e)f_n(\lambda_i)\alpha\mu\beta_n^0\lambda_e^2\lambda_i\big)\,/ \\
& (-2f_{n+1}(\lambda_i)g_n(\lambda_e) - 4f_{n+1}(\lambda_i)g_n(\lambda_e)n + 2f_{n+1}(\lambda_i)g_n(\lambda_e)\mu + 4f_{n+1}(\lambda_i)g_n(\lambda_e)n\mu \\
& + 2f_{n+1}(\lambda_i)g_{n+1}(\lambda_e)\lambda_e - 2f_{n+1}(\lambda_i)g_{n+1}(\lambda_e)\mu\lambda_e + f_{n+1}(\lambda_i)g_n(\lambda_e)\lambda_e^2 \\
& - f_n(\lambda_i)g_n(\lambda_e)\mu\lambda_i - 2f_n(\lambda_i)g_n(\lambda_e)n\mu\lambda_i + f_n(\lambda_i)g_{n+1}(\lambda_e)\mu\lambda_e\lambda_i \\
& + 4f_{n+1}(\lambda_i)g_n(\lambda_e)\alpha\mu + 8f_{n+1}(\lambda_i)g_n(\lambda_e)n\alpha\mu - 4f_2(\lambda_i)g_2(\lambda_e)\alpha\mu\lambda_e \\
& - 2f_{n+1}(\lambda_i)g_n(\lambda_e)\alpha\mu\lambda_e^2 - 2f_n(\lambda_i)g_n(\lambda_e)\alpha\mu\lambda_i - 4f_n(\lambda_i)g_n(\lambda_e)n\alpha\mu\lambda_i \\
& + 2f_n(\lambda_i)g_{n+1}(\lambda_e)\alpha\mu\lambda_e\lambda_i + f_n(\lambda_i)g_n(\lambda_e)\alpha\mu\lambda_e^2\lambda_i\big)\,, \qquad \text{(A.2)}
\end{aligned}
$$

$$
\begin{aligned}
\bar{\alpha}_n^0 \;=\;& -e^{t\left(\lambda_e^2-\lambda_i^2\right)}f_n(\lambda_i)\lambda_e^2\left((g_n(\lambda_e)+2g_n(\lambda_e)n)\alpha_n^0 + (f_{n+1}(\lambda_e)g_n(\lambda_e)+f_n(\lambda_e)g_{n+1}(\lambda_e))\beta_n^0\lambda_e\right) \\
& /\left(\lambda_i\left(g_n(\lambda_e)\lambda_e^2\left(f_{n+1}(\lambda_i)+\alpha\mu\left(-2f_{n+1}(\lambda_i)+f_n(\lambda_i)\lambda_i\right)\right)\right.\right. \\
& + g_{n+1}(\lambda_e)\lambda_e\left(f_n(\lambda_i)\mu\lambda_i\left(1+2\alpha\right)-2f_{n+1}(\lambda_i)\left(-1+\mu+2\alpha\mu\right)\right) \\
& \left.\left. + g_n(\lambda_e)(1+2n)\left(-f_n(\lambda_i)\mu\lambda_i\left(1+2\alpha\right)+2f_{n+1}(\lambda_i)\left(-1+\mu+2\alpha\mu\right)\right)\right)\right)\,, \qquad \text{(A.3)}
\end{aligned}
$$

$$
\begin{aligned}
\hat{\beta}_n^0 \;=\;& e^{t\left(\lambda_e^2-\lambda_i^2\right)}\lambda_e^2\left((g_n(\lambda_e)+2g_n(\lambda_e)n)\alpha_n^0 + (f_{n+1}(\lambda_e)g_n(\lambda_e)+f_n(\lambda_e)g_{n+1}(\lambda_e))\beta_n^0\lambda_e\right) \\
& /\left(\lambda_i\left(g_n(\lambda_e)\lambda_e^2\left(f_{n+1}(\lambda_i)+\alpha\mu\left(-2f_{n+1}(\lambda_i)+f_n(\lambda_i)\lambda_i\right)\right)\right.\right. \\
& + g_{n+1}(\lambda_e)\lambda_e\left(f_n(\lambda_i)\mu\lambda_i\left(1+2\alpha\right)-2f_{n+1}(\lambda_i)\left(-1+\mu+2\alpha\mu\right)\right) \\
& \left.\left. + g_n(\lambda_e)(1+2n)\left(-f_n(\lambda_i)\mu\lambda_i\left(1+2\alpha\right)+2f_{n+1}(\lambda_i)\left(-1+\mu+2\alpha\mu\right)\right)\right)\right)\,, \qquad \text{(A.4)}
\end{aligned}
$$

$$
\begin{aligned}
\hat{\gamma}_n^0 \;=\;& \gamma_n^0 \left(f_{n+1}(\lambda_e)\lambda_e \left(f_n(\lambda_i) + e^{t\lambda_e^2}\alpha\mu \left(f_n(\lambda_i)(-1+n) + f_{n+1}(\lambda_i)\lambda_i\right)\mu_e\right)\right. \\
& + f_n(\lambda_e)\left(f_{n+1}(\lambda_i)\mu\lambda_i \left(-1 + \left(-1 + e^{t\lambda_e^2} n\right)\alpha\right)\right. \\
& \left.\left. + f_n(\lambda_i)(-1+n)\left(1 - \mu + \left(-1 + e^{t\lambda_e^2} n\right)\alpha\mu\right)\right)\right) / \\
& \left(g_{n+1}(\lambda_e)\lambda_e \left(f_n(\lambda_i) + e^{t\lambda_e^2}\alpha\mu \left(f_n(\lambda_i)(-1+n) + f_{n+1}(\lambda_i)\lambda_i\right)\mu_e\right)\right. \\
& - g_n(\lambda_e)\left(f_{n+1}(\lambda_i)\mu\lambda_i \left(-1 + \left(-1 + e^{t\lambda_e^2} n\right)\alpha\right)\right. \\
& \left.\left. + f_n(\lambda_i)(-1+n)\left(1 - \mu + \left(-1 + e^{t\lambda_e^2} n\right)\alpha\mu\right)\right)\right),
\end{aligned} \tag{A.5}
$$

$$
\begin{aligned}
\hat{\gamma}_n^0 \;=\;& \left(e^{t(\lambda_e^2-\lambda_i^2)}\gamma_n^0 (f_{n+1}(\lambda_e)g_n(\lambda_e) + f_n(\lambda_e)g_{n+1}(\lambda_e))\lambda_e \left(-1 + \left(-1 + e^{t\lambda_e^2}\right)\alpha\right)\right) \\
& / \left(-g_{n+1}(\lambda_e)\lambda_e \left(f_n(\lambda_i) + e^{t\lambda_e^2}\alpha\mu \left(f_n(\lambda_i)(-1+n) + f_{n+1}(\lambda_i)\lambda_i\right)\mu_e\right)\right. \\
& + g_n(\lambda_e)\left(f_{n+1}(\lambda_i)\mu\lambda_i \left(-1 + \left(-1 + e^{t\lambda_e^2} n\right)\alpha\right)\right. \\
& \left.\left. + f_n(\lambda_i)(-1+n)\left(1 - \mu + \left(-1 + e^{t\lambda_e^2} n\right)\alpha\mu\right)\right)\right).
\end{aligned} \tag{A.6}
$$

B Symbols

The constants given in (3.26) are given as follows:

$$
\begin{gathered}
X = \lambda_e^3\{\lambda_i f_1(\lambda_i) - 2f_2(\lambda_i)\}g_2(\lambda_e), \\
Y = \lambda_e^3\{\lambda_i g_1(\lambda_e) + 2g_2(\lambda_e)\}f_2(\lambda_i), \\
P = -\mu\lambda_e^3\{-\lambda_i f_1(\lambda_i) + 2f_2(\lambda_i)\}\{2g_2(\lambda_e) + g_1(\lambda_e)\}, \\
G = \mu\{2g_2(\lambda_e)\lambda_e - 6g_1(\lambda_e) + \lambda_e^2 g_1(\lambda_e)\}\{\lambda_i f_1(\lambda_i) - 2f_2(\lambda_i)\}, \\
Z = \{\lambda_i f_1(\lambda_i) - 2f_2(\lambda_i)\}\{\lambda_e g_2(\lambda_e) - 3g_1(\lambda_e)\}, \\
W = \{2\lambda_e g_2(\lambda_e) - (6 - \lambda_e^2)g_1(\lambda_e)\}f_2(\lambda_i), \\
S = 3\{f_2(\lambda_e)g_1(\lambda_e)\}\{\lambda_i f_1(\lambda_i) - 2f_2(\lambda_i)\}, \\
T = 6f_2(\lambda_i)\{f_2(\lambda_e)g_1(\lambda_e) + f_1(\lambda_e)g_2(\lambda_e)\}, \\
Q = -3\mu\{f_2(\lambda_e)g_1(\lambda_e) + f_1(\lambda_e)g_2(\lambda_e)\}\{4f_2(\lambda_i) - 2\lambda_i f_2(\lambda_i)\}, \\
U = S - \frac{X}{\lambda_e^2}; V = T - \frac{Y}{\lambda_e^2}; H = Q - \frac{P}{\lambda_e^2}.
\end{gathered}
$$

C The unknown coefficients (for first order problem)

The unknown coefficients given in (3.44) to (3.47) are given by

$$
\begin{aligned}
\bar{\alpha}_n^1 \;=\;& \left(e^{t\lambda_e^2 - t\lambda_i^2} f_n(\lambda_i) Ma \left(g_n(\lambda_e) + 2g_n(\lambda_e)n - g_{n+1}(\lambda_e)\lambda_e + 2g_n(\lambda_e)\alpha\right.\right. \\
& \left.\left. + 4g_n(\lambda_e)n\alpha - 2g_{n+1}(\lambda_e)\alpha\lambda_e - g_n(\lambda_e)\alpha\lambda_e^2\right)\right) \\
& / \left(\lambda_i \left(2f_{n+1}(\lambda_i)g_n(\lambda_e) + 4f_{n+1}(\lambda_i)g_n(\lambda_e)n - 2f_{n+1}(\lambda_i)g_n(\lambda_e)\mu - 4f_{n+1}(\lambda_i)g_n(\lambda_e)n\mu\right.\right. \\
& - 2f_{n+1}(\lambda_i)g_{n+1}(\lambda_e)\lambda_e + 2f_{n+1}(\lambda_i)g_{n+1}(\lambda_e)\mu\lambda_e - f_{n+1}(\lambda_i)g_n(\lambda_e)\lambda_e^2 + f_n(\lambda_i)g_n(\lambda_e)\mu\lambda_i \\
& + 2f_n(\lambda_i)g_n(\lambda_e)n\mu\lambda_i - f_n(\lambda_i)g_{n+1}(\lambda_e)\mu\lambda_e\lambda_i - 4f_{n+1}(\lambda_i)g_n(\lambda_e)\alpha\mu \\
& - 8f_{n+1}(\lambda_i)g_n(\lambda_e)n\alpha\mu + 4f_{n+1}(\lambda_i)g_{n+1}(\lambda_e)\alpha\mu\lambda_e + 2f_{n+1}(\lambda_i)g_n(\lambda_e)\alpha\mu\lambda_e^2 \\
& + 2f_n(\lambda_i)g_n(\lambda_e)\alpha\mu\lambda_i + 4f_n(\lambda_i)g_n(\lambda_e)n\alpha\mu\lambda_i \\
& \left.\left. - 2f_n(\lambda_i)g_{n+1}(\lambda_e)\alpha\mu\lambda_e\lambda_i - f_n(\lambda_i)g_n(\lambda_e)\alpha\mu\lambda_e^2\lambda_i\right)\right),
\end{aligned} \tag{C.1}
$$

$$
\begin{aligned}
\bar{\beta}_n^1 \;=\; & -\Big(e^{t\lambda_e^2 - t\lambda_i^2} Ma \left(g_n(\lambda_e) + 2g_n(\lambda_e)n - g_{n+1}(\lambda_e)\lambda_e + 2g_n(\lambda_e)\alpha\right. \\
& \left. + 4g_n(\lambda_e)n\alpha - 2g_{n+1}(\lambda_e)\alpha\lambda_e - g_n(\lambda_e)\alpha\lambda_e^2\right)\Big) \\
& / \left(\lambda_i \left(2f_{n+1}(\lambda_i)g_n(\lambda_e) + 4f_{n+1}(\lambda_i)g_n(\lambda_e)n - 2f_{n+1}(\lambda_i)g_n(\lambda_e)\mu - 4f_{n+1}(\lambda_i)g_n(\lambda_e)n\mu\right.\right. \\
& - 2f_{n+1}(\lambda_i)g_{n+1}(\lambda_e)\lambda_e + 2f_{n+1}(\lambda_i)g_{n+1}(\lambda_e)\mu\lambda_e - f_{n+1}(\lambda_i)g_n(\lambda_e)\lambda_e^2 + f_n(\lambda_i)g_n(\lambda_e)\mu\lambda_i \\
& + 2f_n(\lambda_i)g_n(\lambda_e)n\mu\lambda_i - f_n(\lambda_i)g_{n+1}(\lambda_e)\mu\lambda_e\lambda_i - 4f_{n+1}(\lambda_i)g_n(\lambda_e)\alpha\mu \\
& - 8f_{n+1}(\lambda_i)g_n(\lambda_e)n\alpha\mu + 4f_{n+1}(\lambda_i)g_{n+1}(\lambda_e)\alpha\mu\lambda_e + 2f_{n+1}(\lambda_i)g_n(\lambda_e)\alpha\mu\lambda_e^2 \\
& + 2f_n(\lambda_i)g_n(\lambda_e)\alpha\mu\lambda_i + 4f_n(\lambda_i)g_n(\lambda_e)n\alpha\mu\lambda_i \\
& \left.\left. - 2f_n(\lambda_i)g_{n+1}(\lambda_e)\alpha\mu\lambda_e\lambda_i - f_n(\lambda_i)g_n(\lambda_e)\alpha\mu\lambda_e^2\lambda_i\right)\right), \qquad \text{(C.2)}
\end{aligned}
$$

$$
\bar{\gamma}_n^1 = 0, \qquad \text{(C.3)}
$$

$$
\alpha_n^1 = 0, \qquad \text{(C.4)}
$$

$$
\begin{aligned}
\hat{\alpha}_n^1 \;=\; & \left(f_{n+1}(\lambda_i)g_n(\lambda_e)Ma\right) \\
& / \left(2f_{n+1}(\lambda_i)g_n(\lambda_e) + 4f_{n+1}(\lambda_i)g_n(\lambda_e)n - 2f_{n+1}(\lambda_i)g_n(\lambda_e)\mu - 4f_{n+1}(\lambda_i)g_n(\lambda_e)n\mu\right. \\
& - 2f_{n+1}(\lambda_i)g_{n+1}(\lambda_e)\lambda_e + 2f_{n+1}(\lambda_i)g_{n+1}(\lambda_e)\mu\lambda_e - f_{n+1}(\lambda_i)g_n(\lambda_e)\lambda_e^2 + f_n(\lambda_i)g_n(\lambda_e)\mu\lambda_i \\
& + 2f_n(\lambda_i)g_n(\lambda_e)n\mu\lambda_i - f_n(\lambda_i)g_{n+1}(\lambda_e)\mu\lambda_e\lambda_i - 4f_{n+1}(\lambda_i)g_n(\lambda_e)\alpha\mu \\
& - 8f_{n+1}(\lambda_i)g_n(\lambda_e)n\alpha\mu + 4f_{n+1}(\lambda_i)g_{n+1}(\lambda_e)\alpha\mu\lambda_e + 2f_{n+1}(\lambda_i)g_n(\lambda_e)\alpha\mu\lambda_e^2 \\
& + 2f_n(\lambda_i)g_n(\lambda_e)\alpha\mu\lambda_i + 4f_n(\lambda_i)g_n(\lambda_e)n\alpha\mu\lambda_i \\
& \left. - 2f_n(\lambda_i)g_{n+1}(\lambda_e)\alpha\mu\lambda_e\lambda_i - f_n(\lambda_i)g_n(\lambda_e)\alpha\mu\lambda_e^2\lambda_i\right), \qquad \text{(C.5)}
\end{aligned}
$$

$$
\beta_n^1 = 0, \qquad \text{(C.6)}
$$

$$
\begin{aligned}
\hat{\beta}_n^1 \;=\; & -\left(f_{n+1}(\lambda_i)Ma\right) \\
& / \left(2f_{n+1}(\lambda_i)g_n(\lambda_e) + 4f_{n+1}(\lambda_i)g_n(\lambda_e)n - 2f_{n+1}(\lambda_i)g_n(\lambda_e)\mu - 4f_{n+1}(\lambda_i)g_n(\lambda_e)n\mu\right. \\
& - 2f_{n+1}(\lambda_i)g_{n+1}(\lambda_e)\lambda_e + 2f_{n+1}(\lambda_i)g_{n+1}(\lambda_e)\mu\lambda_e - f_{n+1}(\lambda_i)g_n(\lambda_e)\lambda_e^2 + f_n(\lambda_i)g_n(\lambda_e)\mu\lambda_i \\
& + 2f_n(\lambda_i)g_n(\lambda_e)n\mu\lambda_i - f_n(\lambda_i)g_{n+1}(\lambda_e)\mu\lambda_e\lambda_i - 4f_{n+1}(\lambda_i)g_n(\lambda_e)\alpha\mu \\
& - 8f_{n+1}(\lambda_i)g_n(\lambda_e)n\alpha\mu + 4f_{n+1}(\lambda_i)g_{n+1}(\lambda_e)\alpha\mu\lambda_e + 2f_{n+1}(\lambda_i)g_n(\lambda_e)\alpha\mu\lambda_e^2 \\
& + 2f_n(\lambda_i)g_n(\lambda_e)\alpha\mu\lambda_i + 4f_n(\lambda_i)g_n(\lambda_e)n\alpha\mu\lambda_i \\
& \left. - 2f_n(\lambda_i)g_{n+1}(\lambda_e)\alpha\mu\lambda_e\lambda_i - f_n(\lambda_i)g_n(\lambda_e)\alpha\mu\lambda_e^2\lambda_i\right), \qquad \text{(C.7)}
\end{aligned}
$$

$$
\gamma_n^1 = 0, \qquad \text{(C.8)}
$$

$$
\hat{\gamma}_n^1 = 0. \qquad \text{(C.9)}
$$

D Coefficients Couette flow

$$
E_{11}^1 = \frac{2FL}{(2 + \lambda_e^2 Pr_s)} \frac{\left(3g_1(\lambda_e)\lambda_e^2 \left(f_2(\lambda_i) + \alpha\mu\left(-2f_2(\lambda_i) + f_1(\lambda_i)\lambda_i\right)\right)\right)}{\delta_1}, \qquad \text{(D.1)}
$$

$$
F_{22}^1 = \frac{5Fg_2(\lambda_e)\lambda_e^2 \left(f_3(\lambda_i)(2\alpha\mu - 1) - \alpha f_2(\lambda_i)\mu\lambda_i\right)}{6\left(Pr_s\lambda_e^2 + 6\right)\delta_2} \qquad \text{(D.2)}
$$

where

$$\delta_2 = g_2(\lambda_e)\lambda_e^2\left(f_3(\lambda_i)(2\alpha\mu-1)-\alpha f_2(\lambda_i)\mu\lambda_i\right) + g_3(\lambda_e)\lambda_e\left(2f_3(\lambda_i)(2\alpha\mu+\mu-1)-(2\alpha+1)f_2(\lambda_i)\mu\lambda_i\right) + 5g_2(\lambda_e)\left((2\alpha+1)f_2(\lambda_i)\mu\lambda_i-2f_3(\lambda_i)(2\alpha\mu+\mu-1)\right), \quad \text{(D.3)}$$

and

$$E_{11}^2 = \frac{4MaE_{11}^1}{(2+\lambda_e^2 Pr_s)}\frac{f_2(\lambda_i)\left(-3g_1(\lambda_e)+g_2(\lambda_e)\lambda_e\right)}{\delta_1}, \quad \text{(D.4)}$$

$$F_{11}^2 = \frac{\lambda_e^2 e^{\lambda_e^2 t}}{5\left(2+Pr_s\lambda_e^2\right)}\left(\frac{18F_{22}^1 g_1(\lambda_e)(FL)\left(-\alpha f_1(\lambda_i)\mu\lambda_i+f_2(\lambda_i)(2\alpha\mu-1)\right)}{\delta_3} + \frac{5E_{11}^1 F g_2(\lambda_e)\left(-\alpha f_2(\lambda_i)\mu\lambda_i+f_3(\lambda_i)(2\alpha\mu-1)\right)}{\delta_2}\right), \quad \text{(D.5)}$$

where

$$\delta_3 = g_1(\lambda_e)\lambda_e^2\left(\alpha f_1(\lambda_i)\mu\lambda_i-2\alpha f_2(\lambda_i)\mu+f_2(\lambda_i)\right) + g_2(\lambda_e)\lambda_e\left((2\alpha+1)f_1(\lambda_i)\mu\lambda_i-2f_2(\lambda_i)(2\alpha\mu+\mu-1)\right) + 3g_1(\lambda_e)\left(2f_2(\lambda_i)(2\alpha\mu+\mu-1)-(2\alpha+1)f_1(\lambda_i)\mu\lambda_i\right) \quad \text{(D.6)}$$

$$F_{22}^2 = \frac{6F_{22}^1 f_3(\lambda_i)Ma\left(-5g_2(\lambda_e)+g_3(\lambda_e)\lambda_e\right)}{(\mathrm{Pr}\,\lambda_e^2+6)\,\delta_2}, \quad \text{(D.7)}$$

$$F_{31}^2 = \frac{2\lambda_e^2 e^{\lambda_e^2 t}}{45\left(12+Pr_s\lambda_e^2\right)}\left(-\frac{27F_{22}^1 g_1(\lambda_e)(FL)\left(-\alpha f_1(\lambda_i)\mu\lambda_i+f_2(\lambda_i)(2\alpha\mu-1)\right)}{\delta_3} - \frac{5E_{11}^1 F g_2(\lambda_e)\left(-\alpha f_2(\lambda_i)\mu\lambda_i+f_3(\lambda_i)(2\alpha\mu-1)\right)}{\delta_2}\right), \quad \text{(D.8)}$$

$$F_{33}^2 = \frac{-\lambda_e^2 e^{\lambda_e^2 t}}{45\left(12+Pr_s\lambda_e^2\right)}\left(-\frac{27F_{22}^1 g_1(\lambda_e)(FL)\left(-\alpha f_1(\lambda_i)\mu\lambda_i+f_2(\lambda_i)(2\alpha\mu-1)\right)}{\delta_3} - \frac{5E_{11}^1 F g_2(\lambda_e)\left(-\alpha f_2(\lambda_i)\mu\lambda_i+f_3(\lambda_i)(2\alpha\mu-1)\right)}{\delta_2}\right), \quad \text{(D.9)}$$

$$E_{20}^2 = \frac{\lambda_e^2 e^{\lambda_e^2 t}}{7\left(6+Pr_s\lambda_e^2\right)}\left(\frac{-20F_{22}^1 F g_2(\lambda_e)\left(-\alpha f_2(\lambda_i)\mu\lambda_i+f_3(\lambda_i)(2\alpha\mu-1)\right)}{\delta_2} + \frac{21E_{11}^1(FL)g_1(\lambda_e)\left(-\alpha f_1(\lambda_i)\mu\lambda_i+f_2(\lambda_i)(2\alpha\mu-1)\right)}{\delta_3}\right), \quad \text{(D.10)}$$

$$E_{22}^2 = \frac{-9\lambda_e^2 e^{\lambda_e^2 t}}{(6 + Pr_s\lambda_e^2)} \left(\frac{E_{11}^1(FL)g_1(\lambda_e)\left(-\alpha f_1(\lambda_i)\mu\lambda_i + f_2(\lambda_i)(2\alpha\mu - 1)\right)}{\delta_3}\right), \tag{D.11}$$

$$E_{40}^2 = \frac{\lambda_e^2 e^{\lambda_e^2 t}}{7\,(20 + Pr_s\lambda_e^2)} \left(\frac{20F_{22}^1 F g_2(\lambda_e)\left(-\alpha f_2(\lambda_i)\mu\lambda_i + f_3(\lambda_i)(2\alpha\mu - 1)\right)}{\delta_2}\right), \tag{D.12}$$

$$E_{44}^2 = \frac{-\lambda_e^2 e^{\lambda_e^2 t}}{84\,(20 + Pr_s\lambda_e^2)} \left(\frac{5F_{22}^1 F g_2(\lambda_e)\left(-\alpha f_2(\lambda_i)\mu\lambda_i + f_3(\lambda_i)(2\alpha\mu - 1)\right)}{\delta_2}\right), \tag{D.13}$$

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