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BRIEF COMMUNICATION

DRAG AND MASS TRANSFER FOR FLOW OF A CARREAU FLUID PAST A SWARM OF NEWTONIAN DROPS

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INTRODUCTION

Knowledge of hydrodynamic aspects of relative motion between bubbles or drops and a continuous phase is a prerequisite to understanding and rationalizing many transport processes encountered in chemical and environmental operations. Examples of such operations include gas-liquid contacting, liquid-liquid extraction, emulsion polymerization and production of paints and liquid detergents. The drag experienced by the liquid drop and the information around the drop which could be used to obtain the mass transfer rate between the two phases are of central importance in all these applications. In the past, the variational principles were used to study the non-Newtonian flow over a swarm of drops and/or bubbles. Among others, Jarzebski & Malinowski (1986) used them to estimate the upper and lower bounds on the drag coefficient for a power law fluid and for a Carreau fluid (Jarzebski & Malinowski 1987a). Gummalam *et al.* have used them to predict the rising velocity of spherical bubbles in a power law fluid (Gummalam & Chhabra 1987) and in a Carreau fluid (Gummalam *et al.* 1988). In spite of their widespread utility, the variational results suffer from the drawback that the predictions are sensitive to the choice of trial functions used in the analysis. Experience also shows that the bounds diverge for higher degrees of non-Newtonian behavior and therefore, are not very useful for strongly non-Newtonian fluids. Jarzebski & Malinowski (1987b) obtained the solutions for the power law flow over a swarm of drops by linearizing the governing equations approximately. Their analysis, however, is limited to mildly shear-thinning fluids because of the approximations involved. All the aforementioned theoretical studies have employed the free surface cell model (Happel 1958) to simplify the interactions between drops. Zhu & Deng (1994) have recently used the free surface model to simulate numerically the flow behavior of a power law fluid over a swarm of droplets. Although the power law model provides the simplest representation of the shear thinning behavior, its inability to predict a constant viscosity in the limit of vanishingly small shear rates has raised some doubts as to its appropriateness for describing the creeping flow with stagnation points.

Papers published to date provide the following: (1) approximate, closed-form solutions to the problem, obtained using the linearization technique and hence applicable only for weakly pseudoplastic fluids; (2) variational bound-form solutions, obtained by choosing the trial function which may suffer from errors; and (3) numerical solutions for power law fluids which are unable to predict the zero shear viscosity and infinite shear viscosity most non-Newtonian fluids are known to exhibit. In this note, a numerical solution for the slow motion ofa Carreau fluid over Newtonian spherical drops or bubbles with a non-rigid interface is provided to assess the effects of a wide range of shear thinning behaviors, holdups and viscosity ratios on the drag and the mass transfer rate and to evaluate the effects of the Newtonian plateau seen in the viscosity functions of most non-Newtonian shear thinning fluids.

STATEMENT OF PROBLEM AND NUMERICAL IMPLEMENTATION

Consider the steady, creeping and axisymmetric flow of an incompressible Carreau fluid past a swarm of monosized spherical Newtonian drops of radius R with a superficial velocity V_0 . Happel's cell model assumes that each drop is surrounded by a hypothetical spherical envelope whose surface is frictionless and whose radius is given by $R_1 = R\Phi^{-1/3}$, where Φ is the overall average volumetric holdup (or content) of drops. In a spherical coordinate system, the governing equations are:

$$
E^{*2}\Psi_{\circ}^{*} = \omega_{\circ}^{*}\xi\sin\theta\tag{1}
$$

$$
[\beta + (1 - \beta)(1 + 2\lambda^2 \Pi_o^*)^{(n-1)/2}] E^{*2}(\omega_o^* \xi \sin \theta) + (n-1)\lambda^2 (1 - \beta)(1 + 2\lambda^2 \Pi_o^*)^{(n-3)/2}
$$

$$
\times \left[\frac{\partial \Pi_o^*}{\partial \xi} \frac{\partial}{\partial \xi} (\omega_o^* \xi \sin \theta) + \frac{\partial \Pi_o^*}{\partial \theta} \frac{1}{\xi^2} \frac{\partial}{\partial \theta} (\omega_o^* \xi \sin \theta) \right] = 2\lambda^2 (1 - \beta)(1 - n) F(\xi, \theta) \sin \theta \quad [2]
$$

$$
E^{*2}\Psi_i^* = \omega_i^* \xi \sin \theta \tag{3}
$$

$$
E^{*2}(\omega_i^* \xi \sin \theta) = 0 \tag{4}
$$

where,

$$
E^{*2} = \frac{\partial^2}{\partial \xi^2} + \frac{\sin \theta}{\xi^2} \frac{\partial}{\partial \theta} \left(\frac{1}{\sin \theta} \frac{\partial}{\partial \theta} \right)
$$
 [5]

$$
F(\xi,\theta) = \frac{\partial}{\partial \xi} \left[(1+2\lambda^2 \Pi_o^*)^{(n-3)/2} \left(\xi D_{(\xi\theta)_0}^* \frac{\partial \Pi_o^*}{\partial \xi} + D_{(\theta\theta)_0}^* \frac{\partial \Pi_o^*}{\partial \theta} \right) \right] - \frac{\partial}{\partial \theta} \left[(1+2\lambda^2 \Pi_o^*)^{(n-3)/2} \left(D_{(\xi_0)_0}^* \frac{\partial \Pi_o^*}{\partial \xi} + \frac{D_{(\xi\theta)_0}^*}{\xi} \frac{\partial \Pi_o^*}{\partial \theta} \right) \right] \tag{6}
$$

The relevant boundary conditions are: on the drop surface,

$$
v_{(\xi)0}^*(1,\theta) = v_{(\xi)i}^*(1,\theta) = 0
$$
\n[7a]

$$
v_{(\theta)\circ}^*(1,\theta) = v_{(\theta)i}^*(1,\theta) \tag{7b}
$$

$$
[\beta + (1 - \beta)(1 + 2\lambda^2 \Pi_o^*)^{(n-1)/2}]D_{(\xi\theta)0}^*(1, \theta) = \alpha D_{(\xi\theta)i}^*(1, \theta)
$$
 [7c]

and on the surface of the outer sphere,

$$
v_{(\xi)\circ}^*(s,\theta) = \cos\theta \tag{8a}
$$

$$
D_{\langle \xi \theta \rangle o}^*(s, \theta) = 0 \tag{8b}
$$

and $v_{(\xi)i}^*$ $v_{(\theta)i}^*$ remain finite as ξ tends to zero.

Subscripts "i" and "o" indicate the quantities are calculated for inner dispersed Newtonian phase and outer continuous non-Newtonian phase, respectively. Superscript "*" denotes the quantities in dimensionless form. ζ and θ are dimensionless spherical co-ordinates; Ψ and ω are stream function and vorticity, respectively; $v_{(i)}$ is a physical component of velocity; λ and n are parameters in the Carreau fluid model; Π is the second invariant of rate of deformation tensor $D_{(ij)}$; η_0 and η_{α} are zero shear and infinite shear viscosities of fluids; $\alpha = \mu/\eta_0$, $\beta = \eta_{\alpha}/\eta_0$, $s = R_1/R = \Phi^{-1/3}$; and μ is the viscosity of dispersed Newtonian fluids. The above equations can be obtained in similar way with Zhu & Deng (1994) by using Carreau fluid model in lieu of power law fluid model.

The flow drag on the drop is given as:

$$
F_{\rm D} = 2\pi R^2 \left[\int_0^{\pi} (-p + \tau_{(rr)})_{r=R} \cos \theta \sin \theta \, \mathrm{d}\theta - \int_0^{\pi} (\tau_{(r\theta)})_{r=R} \sin^2 \theta \, \mathrm{d}\theta \right]
$$
 [9]

where $\tau_{(rr)}$ and $\tau_{(r\theta)}$ are the physical components of the deviatoric stress tensor.

The correction factor for the drag coefficient for non-Newtonian behavior is given by:

$$
Y_{\rm D} = \frac{C_{\rm D}}{24/\text{Re}}\tag{10}
$$

where C_D is the drag coefficient and Reynolds number, Re, is defined as $Re = \rho V_0 2R/\eta_0$.

A theoretical prediction of the mass transfer coefficient around a drop can be obtained by using the thin concentration boundary layer approximation of Lochiel & Calderbank (1964) as follows:

Sh/Pe^{1/2} =
$$
\left[\frac{2}{\pi}\right]^{1/2} \left\{\int_0^{\pi} (-v_{(\theta)0}^*)_{\xi=1} \sin^2 \theta \ d\theta \right\}^{1/2}
$$
 [11]

The use of this equation is restricted to high Peclet number and low Reynolds number regime.

The finite difference technique is used to obtain numerical solutions of the governing equations [1]-[4]. An iteration procedure is used to treat the difficulties arising from the boundary conditions as follows (Zhu & Deng 1994):

- (1) Let $m = 0$, estimate an initial value of the azimuthal velocity at the interface, v_{θ}^{*} (1, θ)⁽⁰⁾
- (2) Let

$$
v_{(\theta)\sigma}^*(1,\,\theta)^{(m)} = v_{(\theta)i}^*(1,\,\theta)^{(m)}.\tag{12}
$$

- (3) Set up the discretized system for [1] and [2] and solve this system under the boundary conditions [7a], [12] and [8] until it has converged.
- (4) If

$$
\|[\beta + (1-\beta)(1+2\lambda^2\Pi_o^{*(m)})^{(n-1)/2}]D_{(\xi\theta)\circ}^*(1,\theta)^{(m)} - \alpha D_{(\xi\theta)i}^*(1,\theta)^{(m)}\| \leq \epsilon_1 \|D_{(\xi\theta)i}^*(1,\theta)^{(m)}\|
$$

for some prescribed tolerance ϵ_1 , then terminate the iteration, otherwise.

(5) Let

$$
D_{(\xi\theta)i}^{*}(1,\theta)^{(m)} = \frac{1}{\alpha} \left[\beta + (1-\beta)(1+2\lambda^{2}\Pi_{o}^{* (m)})^{(n-1)/2} \right] D_{(\xi\theta)o}^{*}(1,\theta)^{(m)}.
$$
 [13]

- (6) Set up the discretized system for [3] and [4] and solve this system under the boundary conditions [7a] and [13] until it has converged.
- (7) Let $m \rightarrow m + 1$.
- (8) Repeat steps (2) – (6) .

In the numerical calculations, the value of tolerance, ϵ_1 , has been reduced gradually until the further reduction of ϵ_1 changed the final results of drag coefficient by no more than 0.01%. The final value of ϵ_1 used in the calculations is 5×10^{-5} . The grid size used in the calculations has been determined by a trial-and-error procedure in a similar way and it is found to be mainly dependent on α as well as fluid parameters λ and n. Finer mesh has to be used for more shear thinning fluid and consequently the computational time increases significantly. The grid has been found to be only weakly dependent on the value of Φ . The increase of Φ has two implications. Firstly, it causes fluids to undergo a higher level of shearing and consequently a finer grid is required to simulate the greater gradient of flow variables over the domain caused by a stronger shearing action. On the other hand, increase of drop content decreases the size of the flow region and therefore the number of grids required decreases accordingly. Consequently, the value of Φ has less significant influence as a result of two nullifying mechanisms. The number of grids has to be increased with the increasing value of α . The tolerance for the convergence used in solving the governing equations [1] and [2] in step 3 and [3] and [4] in step 6 is 10^{-5} .

RESULTS AND DISCUSSION

The effect of the dimensionless characteristic time λ on the drag is shown in figure 1(a) for a single drop and in figure 1(b) for a swarm of drops (holdup $\Phi = 0.4$), in terms of the correction factor Y_{D} , for different values of the flow behavior index n when $\alpha = 1.0$ and $\beta = 0.0$. Included in the same figure are the upper bounds of Jarzebski & Malinowski's solutions (1987a) based on the

variational principles. There is an appreciable discrepancy between the results predicted by the numerical technique and the upper bounds of the variational solutions. Apparently, the upper bounds of the variational solutions overestimate the drag experienced by the fluid drops, especially in the cases of very strong non-Newtonian flow behavior. The results of the drag coefficient $\Phi = 0.4$ obtained from the present computation are given in figure 2 for the viscosity ratio parameter α equal to 0.1 and 10, and β taking the values of 0 and 0.1, respectively.

Numerical predictions of the Sherwood number normalized by Newtonian results are plotted in figure 3 as functions of the flow behavior index n at different values of the viscosity ratio parameters for $\Phi = 0.4$. Included in the same figure are the predictions of Jarzebski & Malinowski (1987a) based on the variational principles. The appreciable discrepancy between the present numerical solutions and the solutions of the variational principles has been observed for high values of the viscosity ratio parameters. The flow behavior index has very little influence on the mass transfer rate for intermediate values of the holdup when the viscosity ratio parameter is small (swarm of bubbles in Carreau fluids as its extreme case) and the mass transfer rate decreases with the decrease in the flow behavior index when the viscosity ratio parameter is large (assemblage of solid spheres as its extreme case).

The results obtained using power law fluids of previous study were also included in figure 2 in comparison with those obtained using Carreau fluids in the present study. All dimensionless quantities for power law fluids were defined based on the value of viscosity at some characteristic second invariant of the rate of deformation tensor (Zhu & Deng 1994). In order to facilitate the comparison between the present results and the previous results of power law fluids, the same characteristic second invariant of the rate of deformation tensor has been used to make the two results consistent and comparable. An inspection of these comparisons reveals that a remarkable discrepancy exists between these two predictions. The smaller the value of λ , the bigger the discrepancy. This discrepancy is presumably due to the inability of power fluid to predict zero shear viscosity. It is interesting to notice that the power law fluid underestimated the flow drag compared

with Carreau fluids. Also, when the ratio between viscosities of Newtonian dispersed drops and non-Newtonian continuous phase is small ($\alpha = 0$ for the limiting case of a swarm of bubbles in non-Newtonian fluids), the large discrepancy can be observed because the lower shear stress level can be expected for small value of α (the shear stress vanishes at the interface when $\alpha = 0$). This is again due to the inability of the power law fluid to predict viscosity at a low shearing level.

Although both the power law fluid and Carreau fluid predict the drag decrease of most non-Newtonian fluid flows in multiple drop systems, the degree of drag reduction differs quantitatively. The predictions of drag coefficient from these two models can differ by more than 30% for $\Phi = 0.4$, $\alpha = 0.1$, $\lambda = 10$ and $n = 0.2$. The Carreau model possesses all the features which time independent shear thinning fluids are known to exhibit. It describes the shear rate dependent viscosity for a variety of materials over several orders of magnitude of shear rates. So it appears to be most suitable among the generalized Newtonian fluid models and therefore provides more accurate predictions of a real system.

For most shear thinning fluids, β is usually small and was neglected in previous studies of the problems (Gummalam *et al.,* 1988; Jarzebski & Malinowski 1987a). Experimental results of Park *et al.* (1975) for polyvinylpyrrolidone solutions have indicated a range of 0.046-0.146 while Xu's (1988) experiments for polyacrylamide solutions have revealed a range of 0.005-0.05 for the values of β . The present study has shown that even though the value of β is small for most shear thinning fluids, β has a significant influence on the drag coefficient (see figure 2). Quantitatively, the values of drag coefficient for $\beta = 0.0$ and $\beta = 0.1$ differ from each other by an order of magnitude for $\Phi = 0.4$, and $\lambda = 10$. The larger the value of α , the larger the differences.

CONCLUDING REMARKS

The upper bounds of the solutions based on the variational principles, as suggested by previous investigators, predict the drag coefficient and the mass transfer rate with a remarkable error. The reduction in the drag coefficient due to the pseudoplasticity of the fluids is more significant for a larger value of the holdup and that the shear thinning behavior of the fluids results in a reduction in the holdup effect on the drag coefficient. There is very little influence of the pseudoplastic behavior of the fluids on the mass transfer rate for the swarm of drops when the viscosity ratio parameter is small and the mass transfer rate is reduced by the pseudoplasticity of the fluids when the viscosity ratio parameter is large. The degree of the reduction in mass transfer rate due to the pseudoplastic behavior is more significant at high values of holdup. The effects of zero shear viscosity and infinite shear viscosity on the drag coefficient could have been underestimated in previous studies of the problems by either choosing a simplified constitutive equation of power law fluid which is unable to predict both the zero shear viscosity and the infinite shear viscosity or neglecting the infinite shear viscosity from a more realistic fluid model for the mathematical simplicity.

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