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PARTICLE-LIQUID MASS TRANSFER IN VISCOELASTIC MEDIA

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Abstract—The problem of mass transfer from a solid sphere to a viscoelastic fluid has been examined theoretically. It is shown that fluid elasticity increases marginally the mass transfer rate in the creeping flow regime. This will have serious implications on the mass transfer from bubbles if impurities are present. Some conclusions on mass transfer at high Reynolds numbers are also offered.

INTRODUCTION

Mass transport phenomena in non-Newtonian fluids are of great importance. There are many instances, where mass transport occurs in a non-Newtonian fluid in a two phase dispersion (Astarita & Mashelkar, 1977). The common examples are sewage treatment processes, fermentation processes (e.g. antibiotic production), polymer processing (e.g. vacuum devolatilization of molten polymers) and polymer production (e.g. finishing stages of some polyesterification reactions). In many instances, bubbles and drops acquire surface rigidity due to the accumulation of surface active agents, which impede the interfacial motion. It thus becomes important to examine the mass transport rate in particle systems with rigid interfaces.

Expressions for the mass transfer rate from a bubble moving in a viscoelastic liquid were obtained by Moo-Young & Hirose [1972] and Tiefenbruck & Leal [1980] and the effect of fluid elasticity on the mass transfer rate from a drop was discussed by Shirotsuka & Kawase [1974]. However, mass transfer in a viscoelastic liquid from a fluid sphere, the surface of which has been immobilized by surface active agents, has not been analyzed so far.

In this communication, we shall consider the influence of the elasticity of the fluid on mass transfer characteristics from a solid sphere.

CREEPING FLOW REGION

Since the mass transport in non-Newtonian fluids occurs in the region of high Peclet number, we may use the general short range diffusion equations developed by Baird & Hamielec (1962) and by Lochiel & Calderbank (1964).

The thin concentration boundary layer approximation leads to

$$\mathrm{Sh}/\mathrm{Pe}^{1/3} = 0.641\left\{\int_0^\pi \left(\frac{u'_\theta}{U}\sin\theta\right)^{1/2}\sin\theta\, d\theta\right\}^{2/3}. \qquad [1]$$

where u'_θ is the gradient of the tangential velocity component at the surface Sh is the Sherwood number ($2ak_c/D$), Pe is the Peclet number ($2aU/D$), D is the diffusivity, k_c is the mass transfer coefficient, U is the velocity of the uniform stream, θ is the spherical coordinate, a is the radius of solid sphere. If the expression for u'_θ is known, then [1] can be integrated to obtain the mass transfer coefficient for a solid sphere moving in viscoelastic fluids at high Peclet numbers.

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The starting point of our analysis is the stream functions obtained by Leslie (1961) for the creeping flow of viscoelastic fluid around a solid sphere. To portray the viscoelastic behavior of the ambient liquid, we have chosen the 3-parameter Oldroyd equation

$$\tau + \lambda_1 \frac{\delta \tau}{\delta t} = \eta_0 \left[E + \lambda_2 \frac{\delta E}{\delta t} \right] \qquad [2]$$

where E is the rate of deformation tensor, τ is the stress tensor, λ_1 and λ_2 are relaxation and retardation times, and η_0 is the viscosity at zero shear. The operator, $\delta/\delta t$, signifies the convected derivative of a contravariant tensor. The stream function for a 3-constant Oldroyd fluid is given in the appendix. At small distances from the surface of the sphere ($y' = \xi - 1$, $y' \ll 1$), the stream function reduces to

$$\begin{aligned}\psi^* = & -\tfrac{3}{4} y'^2 \sin^2\theta - \mathrm{De}^2\, y'^2 \{(0.009619 \sin^4\theta \\ & -0.009788 \sin^2\theta)(1-\epsilon)^2 + 0.01114 \sin^2\theta\,\epsilon(1-\epsilon) \\ & + (-0.1243 \sin^4\theta + 0.1190 \sin^2\theta)(1-\epsilon)\},\end{aligned} \qquad [3]$$

where De is the Deborah number ($\lambda_1 U/a$), ϵ is a dimensionless time parameter (λ_2/λ_1), and y' is a dimensionless radial distance $[(r-a)/a]$, with r being the radial distance from the solid sphere center, so that the dimensionless tangential velocity component, u_θ, is then given by

$$\begin{aligned} u_\theta = \left(\frac{y' u'_\theta}{U}\right) = & -\frac{1}{\sin\theta}\frac{\partial \psi^*}{\partial y'} = \tfrac{3}{2} y' \sin\theta \\ & + 2y'\,\mathrm{De}^2\{(0.009619 \sin^3\theta - 0.009788 \sin\theta)(1-\epsilon)^2 \\ & + 0.01114 \sin\theta\,\epsilon(1-\epsilon) + (-0.1243 \sin^3\theta + 0.1190 \sin\theta)(1-\epsilon)\}. \end{aligned} \qquad [4]$$

The gradient at the surface of the tangential velocity component, u'_θ, obtained from [4] can now be substituted into [1] to obtain

$$\begin{aligned} \mathrm{Sh}/\mathrm{Pe}^{1/3} = 0.641 \Bigg[\int_0^\pi & [1 + \tfrac{4}{3}\mathrm{De}^2\{(0.009619 \sin^2\theta \\ & - 0.009788)(1-\epsilon)^2 + 0.01114\,\epsilon(1-\epsilon) + (-0.1243 \sin^2\theta \\ & + 0.1190)(1-\epsilon)\}^{1/2}](\tfrac{3}{2})^{1/2} \sin^2\theta \, d\theta \Bigg]^{2/3}. \end{aligned} \qquad [5]$$

When the second term in this integral is much smaller than one, the Sherwood number can be approximated by

$$\mathrm{Sh}/\mathrm{Pe}^{1/3} = 0.734 \left\{ \frac{\pi}{2} + \mathrm{De}^2 (0.0243 - 0.0099\,\epsilon - 0.0144 \epsilon^2) \right\}^{2/3} \qquad [6]$$

where the Deborah number De and the time parameter ϵ are the two dimensionless measures of the liquid's viscoelasticity. Equation [6] predicts that, in the creeping flow regime, the mass transfer rate will increase with increasing relaxation time but the extent of this enhancement is so small that it is unlikely to have any significant effect.

In order to assess the predictive value of [6], we will compare its predictions with those derived from the analysis of Moo-Young & Hirose (1972) and those derived by Tiefenbruck & Leal (1980). It will be remembered that both these analyses apply to gas bubbles, i.e. to spheres with mobile interface.

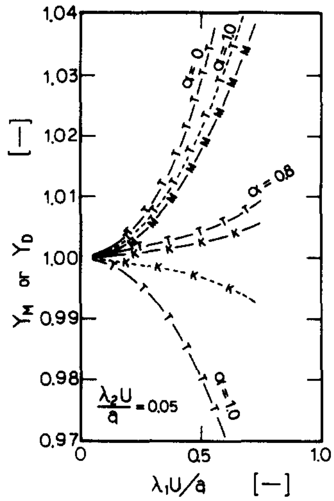

Figure 1. Effect of elasticity of fluids on mass transfer from a solid sphere. Y_m = mass transfer correction factor, Y_D = drag correction factor.

—— T —— Tiefenbruck & Leal (1980) (Bubble).
—— M —— Y_M Moo-Young & Hirose (1972) (Bubble).
—— K —— This work.

- - - - - T - - - - - Y_D Tiefenbruck & Leal (1980) (Bubble).
- - - - - K - - - - - This work.

The rheological equation chosen by Tiefenbruck & Leal (1980) to interpret the viscoelastic behavior of the ambient liquid was the 4-constant Oldroyd equation. Using this, they arrived at a mass transfer expression

$$\mathrm{Sh}/\mathrm{Pe}^{1/2} = 0.65\left[1 + \mathrm{De}^2(1-\epsilon)\left\{\frac{13}{50} + \frac{3\alpha}{70} + \frac{\alpha^2(1-\epsilon)}{21} - \frac{\alpha(1-\epsilon)}{35} - \frac{72\alpha^2}{175}\right\}\right]^{1/2} \qquad (7)$$

where the meaning of the Deborah number De and of the parameter ϵ is the same as given in the appendix. When the third parameter, α, a material constant in the 4-constant Oldroyd model becomes $\alpha = 1$, then this is equivalent to a 4-constant Oldroyd equation reducing to a 3-constant equation, i.e. to [2].

Tiefenbruck & Leal (1980) found that the mass transfer rate may either increase or decrease relative to its value in a Newtonian fluid, depending upon the value of α as shown in figure 1. Although De was too large in the experiments of Zana & Leal (1978) for the analysis presented by Tiefenbruck & Leal (1980) to apply, their data suggested that the mass transfer rate is enhanced in a viscoelastic fluid. The Tiefenbruck & Leal (1980) analysis predicts, as shown in figure 1, that for the case of $\alpha = 1$ a viscoelastic correction factor for mass transfer (Y_M) decreases with the increase of the Deborah number, which is opposite to the results of our analysis, also shown in figure 1. Only when $\alpha = 0.8$, Tiefenbruck's and Leal's analysis yields results close to ours.

Another discrepancy comes to light also in the region of fluid drag. According to Tiefenbruck & Leal (1980), for the case of $\alpha = 1$, the viscoelasticity tends to increase the drag coefficient, whilst the opposite is predicted by our analysis. This discrepancy is shown graphically in figure 1 by the dotted line. The only experimental evidence of the influence of viscoelasticity, that we know of, is the work of Chhabra *et al.* (1980). They found that, under the creeping flow conditions, the drag on a sphere in a viscoelastic fluid is reduced as compared with the drag in a Newtonian liquid. We are not aware of any experimental work on mass transfer from rigid spheres moving slowly in viscoelastic liquids.

To complete the comparison, we have also plotted the results of Moo-Young & Hirose's analysis (1972) for the creeping flow past a bubble. They, too, predicted an enhancement of the mass transfer coefficient and they found experimental confirmation (1970) in dissolving bubbles of ethylene in aqueous solutions of CMC and HEC.

The optimistic predictions of viscoelasticity enhancing the mass transfer under creeping flow conditions might not be, however, entirely justified under all circumstances. Marrucci *et al.* (1970) showed the surfactant impurities, which are always present in polymer solutions, tend to accumulate at the interface and thus provide a surface rigidity. Such a fluid sphere will then have its surface immobilized and thus move through the liquid as a rigid body. As a consequence of that, any enhancement of mass transfer is likely to be negligible.

In this context, we may also briefly mention that, at the threshold of the transition, the power of the Schmidt number changes from 1/2 to 1/3. The same is, however, true for a Newtonian inelastic liquid.

HIGH REYNOLDS NUMBER REGION

Unfortunately, no convective fields have been published in the literature concerning the flow past a solid sphere in the high Reynolds number range. However, the photographic evidence presented by Acharya et al. (1976) suggests that there are dramatic differences in the wake region of an inelastic and a viscoelastic fluid in this range. They experimentally found delayed separation and the phenomenon of dual wake formation in viscoelastic fluids. Verma (1977) theoretically showed that the increase in the elasticity of the liquid causes a shift in the point of separation towards the forward stagnation point. While considering multiparticle systems, the differences in the wake regions can be of great importance. Thus, if a two-particle system is considered, then it can be clearly seen that the wake region of a given sphere mixes the fluid and provides fresh fluid with a uniform composition to the subsequent sphere following in the wake. It appears that this mixing mechanism may be eliminated in elastic fluids and this may have a sizable influence on mass transfer properties of dispersions of viscoelastic liquids.

CONCLUSIONS

It is shown that increased elasticity of fluids enhances to some extent the mass transfer from a solid sphere under the creeping flow conditions but it does so to a lesser extent as compared with a sphere with a mobile interface (bubbles and drops). It should be noted that this influence is opposite so that on the drag coefficient. Both Moo-Young & Hirose (1972) as well as Tiefenbruck & Leal (1980) (at least for $\alpha \geq 0.8$) predict that the elasticity increases the mass transfer coefficient from bubbles which move very slowly in a viscoelastic solution. These predictions may be too optimistic if surface active impurities are present which render the surface of the bubble immobile. All of the three analyses discussed in this work are limited to a case where the elastic contribution of the fluids is small. Theoretical understanding of this problem for strong elasticity must await further investigation.

Finally, we have also drawn some conclusions on mass transfer in the high Reynolds region.

NOMENCLATURE

a	Radius of solid sphere
De	Deborah number, $\mathrm{De} = \lambda_1 U/a$
D	diffusivity
E	rate of deformation tensor
F_D	drag force
k_c	mass transfer coefficient
Pe	Peclet number ($= 2a\,U/D$)
r	radial distance from the solid sphere center
Sh	Sherwood number ($= k_c 2a/D$)
t	time

U velocity of uniform stream
u'_θ gradient at the surface of the tangential velocity component
Y_D drag correction factor $(= (F_D)/(F_D)_{\text{Newtonian}})$
Y_M mass transfer correction factor $(=(\text{Sh}/\text{Pe}^{1/3})/(\text{Sh}/\text{Pe}^{1/3})_{\text{Newtonian}}$ or $(\text{Sh}/\text{Pe}^{1/2})/(\text{Sh}/\text{Pe}^{1/2})_{\text{Newtonian}})$
y' dimensionless radial distance $\left(= \dfrac{r-a}{a}\right)$

Greek symbols

α material constant in 4-constant Oldroyd model
ϵ dimensionless time parameter $(= \lambda_2/\lambda_1)$
θ spherical coordinate
η_0 viscosity at zero shear
λ_1 relaxation time
λ_2 retardation time
ξ dimensionless radial distance $(= r/a)$
τ stress tensor
ψ stream function
ψ^* dimensionless stream function $(= \psi/a^2 U)$

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APPENDIX

The dimensionless stream function obtained by Leslie (1961) reduced to a 3-constant Oldroyd fluid.

$$
\begin{aligned}
\psi^* = {} & \tfrac{1}{2}(\xi^2 - \tfrac{3}{2}\xi + \tfrac{1}{2}\xi^{-1})\sin^2\theta + \tfrac{3}{8}\mathrm{De}(1-\epsilon)\sin^2\theta\cos\theta(1-\xi^{-1})^3 \\
& + \mathrm{De}^2[\tfrac{27}{8}(1-\epsilon)^2\{\sin^4\theta(0.0629\ \xi^{-1} - 0.4583\ \xi^{-2} + 0.7655\ \xi^{-3} \\
& + 0.2143\ \xi^{-3}\ln\xi - 0.6667\ \xi^{-4} + 0.2980\ \xi^{-5} + 0.0417\ \xi^{-6} - 0.0431\ \xi^{-7}) \\
& + \sin^2\theta(0.0006\ \xi - 0.0374\ \xi^{-1} + 0.3333\ \xi^{-2} - 0.1714\ \xi^{-3}\ln\xi \\
& - 0.6181\ \xi^{-3} + 0.5833\ \xi^{-4} - 0.2352\ \xi^{-5} - 0.0833\ \xi^{-6} + 0.0567\ \xi^{-7})\} \\
& + \tfrac{27}{8}\epsilon(1-\epsilon)\{\sin^4\theta(0.2704\ \xi^{-1} - 1.1250\ \xi^{-2} + 2.1511\ \xi^{-3} \\
& - 0.2143\ \xi^{-3}\ln\xi - 1.8333\ \xi^{-4} + 0.5354\ \xi^{-5} + 0.0417\ \xi^{-6} - 0.0402\ \xi^{-7}) \\
& + \sin^2\theta(-0.0006\ \xi - 0.1849\ \xi^{-1} + 0.0833\ \xi^{-2} + 0.1714\ \xi^{-3}\ln\xi \\
& - 1.7152\ \xi^{-3} + 1.5278\ \theta^{-4} - 0.4315\ \xi^{-5} - 0.833\ \xi^{-6} + 0.0544\ \xi^{-7})\} \\
& + \tfrac{9}{4}(1-\epsilon)\{\sin^4\theta(0.1515\ \xi^{-1} - 0.9375\ \xi^{-2} + 0.3733\ \xi^{-3} \\
& + 0.9643\ \xi^{-3}\ln\xi + 0.5000\ \xi^{-4} - 0.0758\ \xi^{-5} - 0.0208\ \xi^{-6} + 0.0061\ \xi^{-7} \\
& + 0.0032\ \xi^{-9}) + \sin^2\theta(0.0044\ \xi - 0.1363\ \xi^{-1} + 0.7500\ \xi^{-2} \\
& - 0.7714\ \xi^{-3}\ln\xi - 0.2815\ \xi^{-3} - 0.3750\ \xi^{-4} + 0.0011\ \xi^{-5} \\
& + 0.0417\ \xi^{-6} + 0.0012\ \xi^{-7} - 0.0055\ \xi^{-9})\}]
\end{aligned}
\qquad [\mathrm{A1}]
$$

where $\mathrm{De} = U\lambda_1/a$ and $\epsilon = \lambda_2/\lambda_1$.