Int. J. Multiphase Flow Vol. 9, No. 3, pp. 337–342, 1983
Printed in Great Britain.

0301-9322/83/030337-06$03.00/0
Pergamon Elsevier

BRIEF COMMUNICATION

SETTLING OF ENCAPSULATED DROPLETS AT LOW REYNOLDS NUMBERS

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(*Received* 24 *November* 1981; *in revised form* 22 *July* 1982)

INTRODUCTION

The application of hydrodynamic theory to the motion of particles, solid and fluid, in viscous liquids has received considerable attention in recent years. Important contributions to the understanding of the behaviour of single and multi-particle systems have been made which find application in many fields of technology from geology and atmospheric physics on the one hand to bio-medical engineering on the other.

The study of the motion of particles and droplets arose from the original work of Stokes (1851) who first derived expressions for the terminal settling velocity and the drag coefficient for a single spherical solid particle in a viscous fluid. The extensions to account for translation of a spherical liquid droplet were later proposed by Rybczynski (1911), Hadamard (1911) and Boussinesq (1913). This early theoretical work has formed the basic framework upon which the subject has developed. Many of the important problems associated with the translation of single particles, for example, inertia effects and droplet distortion, have been analysed (Taylor & Acrivos 1964, Pan & Acrivos 1968 and see also Happel & Brenner 1965).

Recently some interesting problems associated with translation of liquid droplets have arisen which have not previously been considered and which have some importance in defining process conditions in their application. The problems relate to translation and settling of encapsulated droplets in viscous liquids and arise in membrane separation processes which are now the subject of much research and development work, Li (1971), Li & Asher (1973), Martin & Davies (1976). In these processes the overall objective is to selectively extract compound(s) from a liquid into a second immiscible liquid. This is accomplished by separating the two miscible phases, donor and receptor, by a membrane through which the solids may diffuse. One such process uses a liquid membrane phase so that there are three fluid phases; the membrane phase has to be immiscible with the other two. Because it is both impractical to produce stable planar membranes and because the overall transfer flux of the solutes depends on the interfacial area between the miscible phases and membrane, in some processes the receptor phase is dispersed as droplets which are encapsulated by the membrane phase, see figure 1. The transfer process using this type of membrane is carried out in a spray or diffusion column, an example of such a column is shown in figure 2. In this case the droplet is formed at the lower phase 1–2 interface and then rises through the phase 3. In this part of the column transfer across the membrane takes place. The performance of the apparatus will depend in part on the rise velocity of the droplets and therefore the calculation of drag coefficient and terminal settling velocity is important. These cannot be calculated from either Stokes or Hadamard–Rybczynski equations since the presence of the membrane phase will influence the flow.

To analyse this consider the three phase system shown in figure 1 in which the droplet assembly, phase 3 + phase 1 is assumed spherical and the membrane phase is assumed to be of uniform thickness. To distinguish between a droplet of single liquid phase and the present case the term globule will be used.

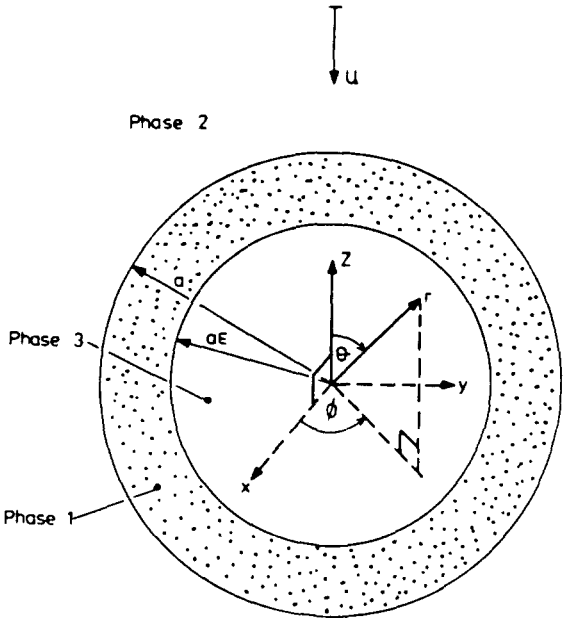

Figure 1. Membrane encapsulated globule.

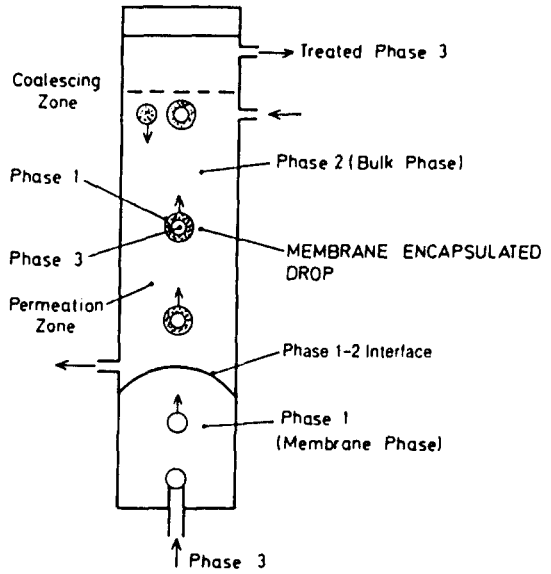

Figure 2. Liquid membrane diffusion column.

2. THEORY

The problem involves the slow motion induced in an incompressible viscous fluid, at rest at infinity, by a fluid globule settling at a constant terminal velocity U. The flows considered are axisymmetric; therefore the Stokes stream function exists. Three regions can be defined, the dispersed phase 3, the membrane phase 1 and the continuous phase 2. The fluid globule, radius a, comprises of a spherical droplet, radius $a\epsilon$ where $0 < \epsilon < 1$, surrounded by a liquid membrane of thickness $a(1-\epsilon)$.

For slow or creeping flow conditions the equations of motion reduce to

$$\frac{1}{\rho}\nabla p = \nu\nabla^2\mathbf{u} \qquad [1]$$

and the continuity equation for incompressible fluids may be written as

$$\nabla . \mathbf{u} = 0 \qquad [2]$$

where $\mathbf{u}$ and p are the local fluid velocity and pressure and ν and ρ the kinetic viscosity and fluid density respectively.

The solution expressed in terms of the stream function ψ_i ($i = 1, 2, 3$) for each flow regime i is:

$$\psi_i = \sin^2\theta \left[\frac{1}{10} A_i r^4 - \frac{1}{2} B_i r + C_i r^2 + \frac{D_i}{r}\right]. \qquad [3]$$

A_i, B_i, C_i and D_i ($i = 1, 2, 3$) are determined from the boundary conditions which express the continuity of velocity and tangential stresses as well as the vanishing of the normal velocity component u_r at each interface. The velocity components must remain finite for all values of r, in particular at $r = 0$, and tend to the free stream velocity, U, as $r \to \infty$. Concentration gradients of solutes or surfactants in the interface are neglected.

The frictional force, F_z in the z-direction opposing the motion of the fluid globule, $r = a$, in a viscous fluid, viscosity μ_2 is:

$$F_z = -4\pi\mu_2 B_2 \qquad [4]$$

or

$$\lambda = \frac{F_z}{-6\pi\mu_2 Ua} = \frac{2}{3}\left[\frac{\mu_{32} + 6\mu_{12}^2 G(\epsilon) + \mu_{12}(2 + 3\mu_{32})F(\epsilon)}{\mu_{32} + 4\mu_{12}^2 G(\epsilon) + 2\mu_{12}(1 + \mu_{32})F(\epsilon)}\right] \qquad [5]$$

where

$$\mu_{12} = \mu_1/\mu_2; \qquad \mu_{32} = \mu_3/\mu_2$$

and

$$F(\epsilon) = \frac{(1+\epsilon)(2\epsilon^2 + \epsilon + 2)}{(1-\epsilon)(4\epsilon^2 + 7\epsilon + 4)}$$

$$G(\epsilon) = \frac{1 - \epsilon^5}{(1-\epsilon)^3(4 + 7\epsilon + 4\epsilon^2)}.$$

$F(\epsilon)$ and $G(\epsilon)$ are monotomically increasing functions in $(0, 1)$ and $F(0) = 1/2$, $G(0) = 1/4$; in addition $F(\epsilon)$ and $G(\epsilon) \to \infty$ as $\epsilon \to 1$.

In [5] λ is the correction factor or dimensionless drag coefficient and is a function of ϵ and the dimensionless viscosities μ_1/μ_2 and μ_3/μ_2. In a gravitational force field λ is the correction which must be applied to Stokes' law. If U_s is the Stokes settling velocity then $\lambda U = U_s$ and, since from the theory $\lambda \leq 1$, a fluid globule should travel with a velocity greater than or equal to the Stokes settling velocity, $U = U_s/\lambda$.

The solution for gravity settling is shown graphically in figure 3. Before examining this some conclusions and corollaries may be deduced from the general solution, to the creeping motion equations [5]. Thus:

(a) When $\mu_1 \gg \mu_2$ and $\mu_1 \gg \mu_3$ the situation of an immobile membrane phase is achieved, an example may be a polymer membrane, where in this case $\lambda \to 1$ and $U \to U_s$. This would be the

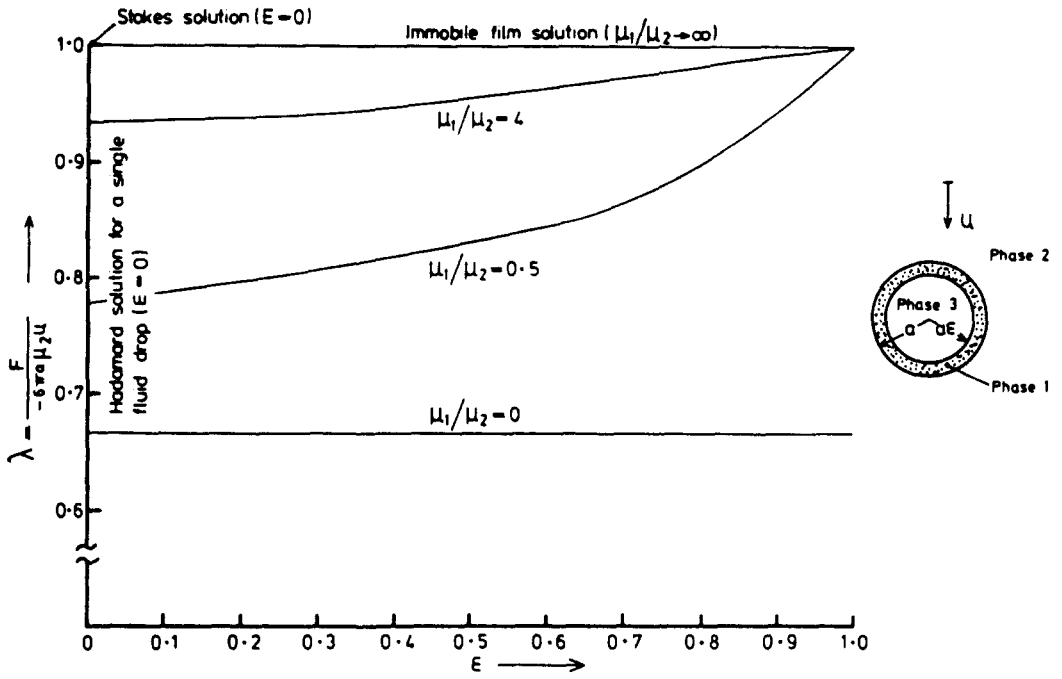

Figure 3. Correction factor, λ, applied to Stokes' law for a single solid particle as a function of radii ratio, ϵ, for various values of μ_1/μ_2 and μ_3/μ_2 of order unity.

anticipated situation since no momentum flux is transferred to the inner phase 3. The terminal settling velocity would then be calculated from Stokes' law using the equivalent density of the globule, phase 1 and phase 3.

(b) If $\mu_3 \gg \mu_1$ and $\mu_3 \gg \mu_2$

$$\lambda \to \frac{2}{3}\frac{(1+3\mu_{12}F(\epsilon))}{(1+2\mu_{12}F(\epsilon))}. \qquad [6]$$

This is the correction factor applicable to a solid particle encapsulated by a membrane.

(c) Limiting cases can be examined, for example for arbitrary values of μ_{12} and μ_{32} as $\epsilon \to 0$ the correction factor reduces to

$$\lambda \to \frac{(2+3\mu_{12})}{3(1+\mu_{12})} = \lambda_f \qquad [7]$$

where λ_f is the correction factor for the Hadamard solution for a single fluid sphere, viscosity μ_1, in an infinite medium, viscosity μ_2.

(d) For a very thin membrane $\epsilon \to 1$, then since

$$\underset{\epsilon \to 1}{\text{Lt}}\left[\frac{F(\epsilon)}{G(\epsilon)}\right] = 0 = \underset{\epsilon \to 1}{\text{Lt}}\left[\frac{1}{G(\epsilon)}\right]$$

the correction factor λ approaches unity in agreement with the Stokes correction factor. In other words, a liquid droplet encapsulated by a very thin membrane translates with the Stokesian velocity of a solid particle not with the velocity of an equivalent fluid droplet. This result is at first rather surprising but can be explained by reference to the streamlines induced by the flow, figure 4. The presence of the membrane to give a three phase system with the attendant interface boundary conditions causes streamline patterns to be established as shown

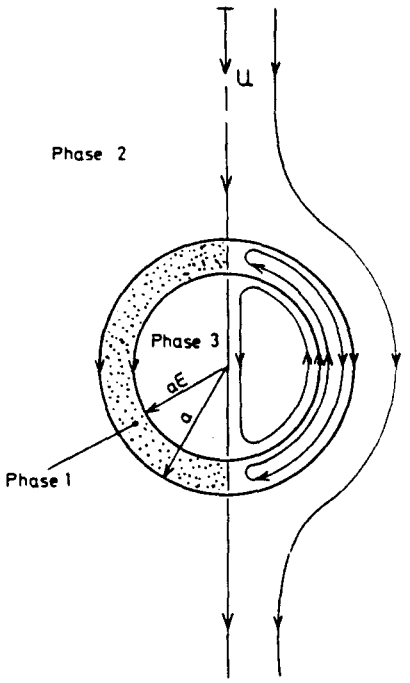

Figure 4. Streamline patterns in an encapsulated globule.

in figure 4. This is a different problem mathematically than that of a single liquid drop but is based on the boundary conditions defined on two interfaces at $r = a$ and $r = a\epsilon$ and the subsequent limit as $\epsilon \to 1$. The direction of the internal calculation streamlines in the inner phase 3 are reversed compared to that in a single fluid droplet. Near $\epsilon = 1$, $(u_\theta)_3$ is in an opposite direction to $(u_\theta)_2$ hence in the limit as $\epsilon \to 1$ the only possible solution satisfying the velocity boundary condition is $(u_\theta) = 0$. Mobility in the membrane causes internal circulation which is suppressed as $\epsilon \to 1$. The net result at this condition is that the globule consists of a stationary fluid droplet encapsulated by an immobile fluid shell. Thus for very thin membranes the drag force coefficient is better represented by Stokes' law than by using the Hadamard correction factor for a fluid sphere.

In practical applications the objective is to use thin membranes, (namely ϵ values near unity), so that the quantity of the membrane phase 1 is minimised and, since mass transfer takes place by diffusion through the membrane, the mass fluxes are kept high. The implication of this analysis on translation has major importance to the mass transfer process. If circulation in the inner phase 3 is suppressed as $\epsilon \to 1$ then mass transfer into or from the inner phase will be by diffusion only. Thus overall mass transfer coefficients will be lower than for liquid droplets having similar transport coefficients.

The graphical solution showing the departure from the Stokes' law correction factor is shown as figure 3. λ is a function of ϵ and the dimensionless viscosities. Data for two values of μ_1/μ_2 are shown. All values of λ lie between the two limiting conditions discussed, $\lambda = 1$ and $\lambda = 2/3$. Thus the settling velocity for the globule U must be such that $U_f > U > U_s$ where the subscripts refer to the Hadamard single fluid drop and Stokes' single solid particle settling velocities respectively.

Membrane techniques have been applied on small scale to systems where phases 2 and 3 are aqueous solutions and phase 1 an organic hydrocarbon film and to the inverse system, that is organic miscible phases 2 and 3 and water membranes. In the first case typical values of μ_1/μ_2 are of the order 4 and in the latter case μ_1/μ_2 lies in the range 0 to 1/2. The examples shown in figure 3 illustrate these two cases. For this theory to be applicable to these systems the radius of

the encapsulated drop must be of the order of 0.5 mm, a range in which equipment of the type shown in figure 2 can operate.

Acknowledgements—This work arose from a research programme on liquid membranes sponsored by the European Economic Community. The authors wish to record their thanks to the E.E.C. for the support of this work.

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