

Apparent Viscosity of a Monodispersed Liquid Aerosol

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This work presents an analytical study of the effective viscosity of a mono-dispersion of liquid aerosol droplets in a gas medium based on a steady-state low-Reynolds number hydrodynamics. The Knudsen and Reynolds numbers are assumed to be small, and thus fluid flows inside and outside the fluid particle can be described using a continuum model with a hydrodynamic slip at the drop-gas interface, while the flow fields are governed by the Stokes equations. Three types of cell models are employed to solve the problem, namely the Simha, Happel and Kuwabara cell models. Regarding the limitations of this study, the analytical expressions of apparent viscosity as functions of the particle volume fraction in a closed form agree with the literature. The bulk viscosity is significantly influenced by the surface properties of the droplet-gas interface and by the internal-to-external viscosity ratio of the droplet. Generally, the influence of the concentration effect of the particles on the apparent viscosity increases with the volume fraction of the dispersed liquid drops.

Key Words: Aerosol, emulsion, liquid droplet, effective viscosity, unit cell model

1. Introduction

The hydrodynamic investigation of particle motion in Newtonian fluids at small Reynolds numbers is significant in various suspension and aerosol problems. Suspension settling velocities, efficiencies of spray scrubber devices for removing particulates from gas streams, and the agglomeration rates of aerosol particles in the atmosphere all depend on the nature of the relative motion of the suspended particles. These suspensions often have complicated rheology properties that differ significantly from those of the suspending fluid, even for a small volume fraction of suspended particles. Studies of suspension

viscosity are important not only for the macroscopic particles involved in many industrial separation and reaction processes, but also in connection with the very small particles commonly known as colloidal, with sizes approaching the molecular dimension of the suspension fluid medium. The same basic variables that characterize sedimentation rates also characterize suspension viscosity, namely: (1) nature of the fluid; (2) nature of the suspended particles; (3) particle concentration; (4) particle and fluid motion- with the shearing field of the latter being the prime distinguishing characteristic. Because of the small size of the particles involved in viscosity problems, other properties, such as internal flexibility and ease of deformation, may also be important.

The theoretical study of the effective viscosity of suspensions originated from the classic work of Einstein (1906) on dilute situations. Einstein's

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formula is generally accepted in the application of extremely dilute dispersions, and thus is said to be accurate to the first order in the volume fraction ϕ of the solid spheres. For a slightly concentrated suspension, the correction terms for ϕ^2 should have been involved by considering the particle-interactions (Happel and Brenner, 1983). Summarizing the hydrodynamic interactions of the two-sphere system in a linear flow field, Batchelor and Green (1972) provided the apparent viscosity to the second order of volume fraction of the solid particles in a colloidal suspension. Batchelor and Green obtained their analytical result by combining two asymptotic analyses, namely the reflection and lubrication theory methods. Yoon and Kim (1987) subsequently conducted a similar investigation that applied a boundary collocation method. These investigations first determined the microscopic model of particle interactions in a dilute dispersion employing both statistical and low Reynolds number hydrodynamic concepts and then obtained the macroscopic ensemble-averaged results. In applying the same mathematic formulation, the parallel problem involving suspensions of immiscible fluid droplets was recently extended by Keh and Chen (2000), whose study also summarizes the current state of knowledge in this area and provides some informative references.

One particularly interesting novel perspective on hydrodynamic treatment, the unit cell model (Simha, 1952; Happel, 1957; Kuwabara, 1959) provides a simpler means of determining the effective viscosity of a mono-dispersed suspension. Based on a homogeneous-environment, the cell models simplify the mono-dispersed colloidal system into a concentric spherical unit cell enclosing a representative particle at its center. Each cell contains suspending fluid in proportion to the fluid-to-solid volume of the suspension, and thus the complicated boundary-value problem for multiple spheres can be reduced to the consideration of a dilational flow within a cell.

Results for the effective viscosity of suspensions of rigid spheres from the cell model were found to be within the range of established experimental data (Simha, 1952; Happel, 1957, 1958).

To solve the Navier-Stokes hydrodynamic problems, there is generally assumed to be no slippage at the solid-fluid and/or the fluid-fluid interfaces. However, this assumption does not accurately reflect the actual transport processes. It has been both experimentally and theoretically confirmed that the adjacent fluid (especially if it is a rarified gas) can cross solid and liquid surfaces (Kennard, 1938; Fulford et al., 1971; Davis, 1972; Beresnev and Chernyak, 1986; Loyalka, 1990; Ying and Peters, 1991; Hutchins et al., 1995; Li and Davis, 1995; Grashchenkov, 1996). Airborne particles also have slippery surfaces. Presumably, any slippage would be proportional to the local velocity gradient next to the particle surface (Basset, 1961; Happel and Brenner, 1983), at least as long as this gradient is small. Theoretical hydrodynamic investigations concerning aerosol suspensions of solid particles and/or liquid drops have attracted less attention than Stokes particles and are more difficult to formulate than Stokes particles because the particles (either solid spheres or fluid droplets) are slippery and an additional flow field inside the particle must be solved to allow the suspended particles to be considered droplets. Recent investigations of the averaged sedimenting velocity of mono-dispersed aerosol systems have used a cell model for solid particles (Chen et al., 1999) and liquid drops (Chen and Yang, 2000).

This study focuses on the bulk rheological properties of a suspension of particles in a Newtonian fluid of uniform viscosity. The following assumptions are made: (1) that the Reynolds number of the relative motion of the fluid near a particle is small compared with 1 and that the Stokes equations describe the fluid motion; (2) that the inertia of a moving particle may be ignored, and

(3) that no external force or couple acts on a particle. These conditions are usually realized in practice by the smallness of the particles involved. It is also assumed that hydrodynamic stresses only influences movement on the particle surface. The analysis ignores the effects of the Brownian motion of particles. Finally, the particles are assumed to be spherical, and of uniform size. (In principles, the calculation is also applicable to non-spherical particles, but the working becomes more complex.) The general analysis supposes the material of the particles to be a Newtonian fluid of viscosity η_i ; the case of rigid particles is then obtained by taking the limit $\eta_i/\eta \rightarrow \infty$, while for gas bubbles in liquid we put $\eta_i/\eta = 0$. A unit cell model (Happel, 1957; Kuwabara, 1959; Zydney, 1995) is applied to predict the viscosity of a suspension of identical spherical droplets. The flow disturbance arising from each droplet is considered to be confined to the cell of fluid surrounding it, which is in turn bounded by virtual envelope. The closed form analytical solutions obtained with this model enable apparent viscosity to be predicted as a function of the volume fraction of the droplets in a wide range. In special cases, our results agree closely with the calculations available in the literature.

2. Analysis

This section considers the effective viscosity of a homogeneous gaseous suspension of identical liquid drops with viscosity η subjected to a deforming motion. The droplets retain their shape with radius a owing to relatively high interfacial tension, and the viscosity of the drops is η_i . The effect of attractive or repulsive interaction potentials between the particles is not considered. Additionally, no coalescence occurred in the quasi-steady mono-dispersion though such coalescence is typical in a concentrated suspension. The Reynolds numbers inside and outside the drops

and the Knudsen number are both assumed to be small, and so the fluid flows are described by the Stokes equations. The formulation presented here is analogous to those developed by Simha (1952), Happel (1957), and Kuwabara (1959). This investigation takes a unit cell model in which each particle is envisaged to be surrounded by a concentric spherical shell of ambient fluid with an outer radius b such that the cell contains the same volumetric proportion of particles as does the entire suspension (namely, the volume fraction of the particles $\phi = a^3/b^3$). On the outer boundary of the imaginary spherical shell, the Simha model assumes that the fluid velocity is equal to the bulk velocity, the Happel model assumes that the radial velocity and the shear stress are equal to the bulk flow, while the Kuwabara model assumes that the radial velocity and the tangential vorticity are the same as those of the bulk flow. The definition of relative viscosity used in the above models is based on the ratio of dissipation of energy per unit volume of suspension to that of the suspending fluid alone.

When the cell contains no suspended fluid sphere, the flow field is assumed to comprise a simple shearing motion with a constant velocity gradient 2κ in the zx plane, where (x,y,z) are the rectangular coordinates. Since only spherical particles are considered, the rotational part of the field is not disturbed by the presence of a sphere at the origin of the coordinate system. Thus, the bulk velocity can be expressed as

$$\mathbf{v}_\infty = \kappa(z\mathbf{e}_x + x\mathbf{e}_z), \quad (1)$$

where \mathbf{e}_x and \mathbf{e}_z denote the unit vectors in the x and z directions, respectively.

In spherical coordinates (r, θ, ϕ) , one can employ Lamb's general solution (Happel and Brenner, 1983) of the Stokes equations to determine the velocity fields for the flow inside the fluid sphere $\mathbf{v}_i(\mathbf{x})$ and for the external flow $\mathbf{v}(\mathbf{x})$, respectively, and also to determine the

corresponding pressure distributions $p_i(\mathbf{x})$ and $p(\mathbf{x})$. The solution takes the form

$$v_r = \kappa(r + 6Cr^3 + 2Dr + 6Er^{-2} - 3Fr^{-4})\sin 2\theta \cos \phi, \quad (2a)$$

$$v_\theta = 2\kappa(\frac{1}{2}r + 5Cr^3 + Dr + Fr^{-4})\cos 2\theta \cos \phi, \quad (2b)$$

$$v_\phi = -2\kappa(\frac{1}{2}r + 5Cr^3 + Dr + Fr^{-4})\cos \theta \sin \phi, \quad (2c)$$

$$p = \kappa\eta(42Cr^2 + 12Er^{-3})\sin 2\theta \cos \phi, \quad (2d)$$

for $a \leq r \leq b$, and

$$v_{ri} = \kappa(6C_i r^3 + 2D_i r)\sin 2\theta \cos \phi, \quad (3a)$$

$$v_{\theta i} = 2\kappa(5C_i r^3 + D_i r)\cos 2\theta \cos \phi, \quad (3b)$$

$$v_{\phi i} = -2\kappa(5C_i r^3 + D_i r)\cos \theta \sin \phi, \quad (3c)$$

$$p_i = \frac{2\gamma}{a} + \kappa\eta(42C_i r^2)\sin 2\theta \cos \phi, \quad (3d)$$

for $0 \leq r \leq a$. Here, γ denotes the interfacial tension for the fluid sphere, and the coefficients C , D , E , F , C_i , and D_i are determined from the boundary conditions at surfaces $r = a$ and $r = b$. Since the fluid sphere is assumed to remain spherical, the capillary number $\kappa\eta a/\gamma$ must be sufficiently small for the sphericity and the solution given by Eqs. (2) and (3) satisfies the Young-Laplace equation (Hunter, 1986) for the normal stresses at the particle surface.

Considering the discontinuity of the tangential velocity between the internal and external fluids, which is proportional to the tangential shear stress, as well as the continuity of the tangential shear stress, the boundary conditions for the velocity fields at the particle surface are

$$r = a \quad \mathbf{e}_r \cdot \mathbf{v} = 0, \quad (4a)$$

$$\mathbf{e}_r \cdot (\mathbf{v} - \mathbf{v}_i) = 0, \quad (4b)$$

$$(\mathbf{I} - \mathbf{e}_r \mathbf{e}_r) \cdot (\mathbf{v} - \mathbf{v}_i) = \frac{C_m l}{\eta} (\mathbf{I} - \mathbf{e}_r \mathbf{e}_r) \mathbf{e}_r : \boldsymbol{\tau}, \quad (4c)$$

$$(\mathbf{I} - \mathbf{e}_r \mathbf{e}_r) \mathbf{e}_r : (\boldsymbol{\tau} - \boldsymbol{\tau}_i) = 0, \quad (4d)$$

where $(\mathbf{e}_r, \mathbf{e}_\theta, \mathbf{e}_\phi)$ denote the unit vectors in the spherical coordinates, \mathbf{I} represents the unit dyadic, l is the mean free path of the surrounding fluid, and $\boldsymbol{\tau}$ ($=\eta[\nabla\mathbf{v}+(\nabla\mathbf{v})^T]$) and $\boldsymbol{\tau}_i$ ($=\eta_i[\nabla\mathbf{v}_i+(\nabla\mathbf{v}_i)^T]$) denote the deviatoric stress tensors for the external and internal flows, respectively. For boundary condition (4c), C_m represents the dimensionless frictional slip coefficient, which is semi-empirically related to the momentum accommodation coefficient α at the particle-gas interface by $C_m = (2-\alpha)/\alpha$ (Kennard, 1938). Although C_m clearly depends upon the surface characteristic, examination of the theoretical and experimental data suggests it will range between 1.0-1.5 (Davis, 1972; Talbot et al., 1980; Loyalka, 1990; Takata and Sone, 1995). Substituting the general solutions (2) and (3) into boundary conditions (4) produces four linear algebraic relative formulas

$$6Ca^2 + 2D + 6Ea^{-3} - 3Fa^{-5} = -1, \quad (5a)$$

$$3C_i a^2 + D_i = 0, \quad (5b)$$

$$\begin{aligned} & -2(5 - 16C_m^*)Ca^2 - 2(1 - 2C_m^*)D + \\ & 12C_m^*Ea^{-3} - 2(1 + 8C_m^*)Fa^{-5} \\ & + 10C_i a^2 + 2D_i = 1 - 2C_m^*, \end{aligned} \quad (5c)$$

$$\begin{aligned} & 16Ca^2 + 2D + 6Ea^{-3} - 8Fa^{-5} - \\ & 16\eta^*C_i a^2 - 2\eta^*D_i = -1 \end{aligned}, \quad (5d)$$

where

$$C_m^* = C_m Kn, \quad (6a)$$

$$\eta^* = \frac{\eta_i}{\eta}, \quad (6b)$$

with $Kn = l/a$, the Knudsen number of the aerosol

suspension. To determine the six unknowns C , D , E , F , C_i and D_i , the other two relations must first be formulated, namely the relations resulting from the boundary conditions at the surface of the cell wall ($r=b$).

The energy dissipated per unit time within a cell may be calculated using the surface integral (Happel and Brenner, 1983)

$$\Theta = \int_S \mathbf{n} \cdot \boldsymbol{\sigma} \cdot \mathbf{v} dS = 4\kappa^2 V_c \eta_c, \quad (7)$$

where $\boldsymbol{\sigma} = -p\mathbf{I} + \boldsymbol{\tau}$, $V_c (= \frac{4}{3}\pi b^3)$ is the volume of a unit cell and $4\kappa^2 V_c \eta_c$ is the energy dissipation for the unperturbed flow in the cell. The integration in Eq. (7) needs to be extended over both the outer surface ($r=b$) and inner surface ($r=a$). Evaluation of the integral by using Eq. (3) yields the following expression for the effective viscosity of the suspension

$$\eta_c = \eta(1 + \zeta\varphi), \quad (8)$$

where

$$\begin{aligned} \zeta\varphi = & \frac{2}{5}(42Cb^2 + 10D - 3Eb^{-3}) \\ & - \frac{2}{5}(3Cb^2 - 2D + 18Eb^{-3} - 12Fb^{-5}) \\ & \times (6Cb^2 + 2D + 6Eb^{-3} - 3Fb^{-5}) \\ & + \frac{12}{5}(8Cb^2 + D + 3Eb^{-3} - 4Fb^{-5})(5Cb^2 + D + Fb^{-5}) \\ & + 24\eta^* \varphi (C_i a^2)^2 \\ & + \frac{4}{5}\eta^* \varphi (27C_i a^2 + 5D_i)(3C_i a^2 + D_i). \end{aligned} \quad (9)$$

Notably, Eqs. (8) and (9) are the most general formulas, which satisfy all kinds of cell models in discussing the apparent viscosity of a mono-dispersion of colloidal particles.

Simha-Type Cell Model

Employing the Simha model for the boundary conditions on the outer (virtual) envelope of the cell produces

$$r = b \quad \mathbf{v} - \mathbf{v}_\infty = \mathbf{0}. \quad (10)$$

Substituting the general solution (3) into boundary condition (10) then produces two linear algebraic relative formulas

$$6Cb^2 + 2D + 6Eb^{-3} - 3Fb^{-5} = 0, \quad (11a)$$

$$5Cb^2 + D + Fb^{-5} = 0. \quad (11b)$$

Thus, the unknowns C , D , E , F , C_i and D_i can be formulated by Eqs. (5) and (11). The procedure is straightforward and produces

$$C = -\frac{1}{2a^2\delta_s} \left[(2 + 5\eta^* + 10\eta^* C_m^*) \varphi^{5/3} - 5\eta^* \varphi^{7/3} \right], \quad (12a)$$

$$D = \frac{1}{2\delta_s} \left[5(2 + 5\eta^* + 10\eta^* C_m^*) \varphi - 21\eta^* \varphi^{5/3} + 4(1 - \eta^* + 5\eta^* C_m^*) \varphi^{10/3} \right], \quad (12b)$$

$$E = -\frac{a^3}{3\delta_s} \left[(2 + 5\eta^* + 10\eta^* C_m^*) + 5(1 - \eta^* + 5\eta^* C_m^*) \varphi^{7/3} \right], \quad (12c)$$

$$F = -\frac{2a^5}{\delta_s} \left[\eta^* + (1 - \eta^* + 5\eta^* C_m^*) \varphi^{5/3} \right], \quad (12d)$$

$$C_i = \frac{1}{2a^2\delta_s} (2 - 7\varphi^{5/3} + 5\varphi^{7/3}), \quad (12e)$$

$$D_i = -\frac{3}{2\delta_s} (2 - 7\varphi^{5/3} + 5\varphi^{7/3}), \quad (12f)$$

where

$$\delta_s = 4(1 + \eta^* + 5\eta^* C_m^*) - 5(2 + 5\eta^* + 10\eta^* C_m^*) \varphi + 42\eta^* \varphi^{5/3} + 5(2 - 5\eta^* + 10\eta^* C_m^*) \varphi^{7/3} - 4(1 - \eta^* + 5\eta^* C_m^*) \varphi^{10/3} \quad (13)$$

Applying the relatives (5) and (11), Eq. (9) reduces to

$$\zeta_s \varphi = 21Cb^2 + 5D + 24\eta^* \varphi (C_i a^2)^2. \quad (14)$$

Combining Eqs. (12) and (14) produces

$$\zeta_s = \frac{2}{\delta_s} \left[(2 + 5\eta^* + 10\eta^* C_m^*) + 5(1 - \eta^* + 5\eta^* C_m^*) \varphi^{7/3} \right] + \frac{6\eta^*}{\delta_s^2} (2 - 7\varphi^{5/3} + 5\varphi^{7/3})^2. \quad (15)$$

When $C_m^* = 0$, the above equation reduces to the result obtained by Keh and Chen (2000) for a suspension of identical liquid droplets, and this result can be further simplified to the Simha's result (1957) for a monodispersion of no-slip solid particles by setting $\eta^* \rightarrow \infty$. On the other hand, since $\eta^* \rightarrow \infty$, Eq. (15) condenses to describe the behavior of a mono-dispersed solid aerosol, in which a tangential slip velocity occurs at the gas-solid interfaces.

Happel-Type Cell Model

When the Happel cell model for the boundary conditions on the outer envelope of the cell is applied, Eq. (10) is replaced by

$$r = b \quad \mathbf{e}_r \cdot (\mathbf{v} - \mathbf{v}_\infty) = 0, \quad (16a)$$

$$(\mathbf{I} - \mathbf{e}_r \mathbf{e}_r) \mathbf{e}_r : (\boldsymbol{\tau} - \boldsymbol{\tau}_\infty) = \mathbf{0}, \quad (16b)$$

where $\boldsymbol{\tau}_\infty = (\eta[(\nabla \mathbf{v}_\infty) + (\nabla \mathbf{v}_\infty)^T])$ is the deviatoric stress tensor for the undisturbed flow. Following this change, the two linear algebraic relative formulas are Eq. (11a) and

$$8Cb^2 + D + 3Eb^{-3} - 4Fb^{-5} = 0. \quad (17)$$

Additionally, the coefficients in Eqs. (2) and (3) are

$$C = -\frac{1}{2a^2 \delta_h} \eta^* \varphi^{7/3}, \quad (18a)$$

$$D = \frac{1}{2\delta_h} \left[(2 + 5\eta^* + 10\eta^* C_m^*) \varphi - 2(1 - \eta^* + 5\eta^* C_m^*) \varphi^{10/3} \right], \quad (18b)$$

$$E = -\frac{a^3}{6\delta_h} \left[(2 + 5\eta^* + 10\eta^* C_m^*) - 2(1 - \eta^* + 5\eta^* C_m^*) \varphi^{7/3} \right], \quad (18c)$$

$$F = -\frac{a^5}{\delta_h} \eta^*, \quad (18d)$$

$$C_i = \frac{1}{2a^2 \delta_h} (1 - \varphi^{7/3}), \quad (18e)$$

$$D_i = -\frac{3}{2\delta_h} (1 - \varphi^{7/3}), \quad (18f)$$

where

$$\delta_h = 2(1 + \eta^* + 5\eta^* C_m^*) - (2 + 5\eta^* + 10\eta^* C_m^*) \varphi - (2 - 5\eta^* + 10\eta^* C_m^*) \varphi^{7/3} + 2(1 - \eta^* + 5\eta^* C_m^*) \varphi^{10/3} \quad (19)$$

Applying Eqs. (5) and (17) allows Eq. (9) to be re-formulated as

$$\zeta_h \varphi = \frac{2}{5} (42Cb^2 + 11D) + 24\eta^* \varphi (C_i a^2)^2. \quad (20)$$

Substituting Eq. (18) into Eq. (20) produces

$$\zeta_h = \frac{1}{5\delta_h} \left[11(2 + 5\eta^* + 10\eta^* C_m^*) - 42\eta^* \varphi^{2/3} \right] - 22(1 - \eta^* + 5\eta^* C_m^*) \varphi^{7/3} + \frac{6\eta^*}{\delta_h^2} (1 - \varphi^{7/3})^2. \quad (21)$$

In this model, the result of the effective viscosity (by setting $C_m^* = 0$) is consistent with the formula of a mono-dispersion of fluid spheres (Keh and Chen, 2000). Meanwhile, the simplified case of Eq. (21) with $C_m^* = 0$ and $\eta^* \rightarrow \infty$ agrees with the work of Happel (1957).

Kuwabara-Type Cell Model

When the Kuwabara cell model is used as the outer boundary of the cell, Eq. (16b) becomes

$$r = b \quad (\mathbf{I} - \mathbf{e}_r \mathbf{e}_r) \cdot \nabla \times (\mathbf{v} - \mathbf{v}_\infty) = \mathbf{0}. \quad (22)$$

Thus, the two corresponding formulas according to the boundary conditions at the cell surface are Eq. (11a) and

$$7Cb^2 - 3Eb^{-3} = 0. \quad (23)$$

Following the above change, the coefficients in Eqs. (2) and (3) can be obtained as

$$C = -\frac{1}{4a^2\delta_k}(2 + 5\eta^* + 10\eta^*C_m^*)\varphi^{5/3}, \quad (24a)$$

$$D = \frac{1}{4\delta_k} \left[\frac{10(2 + 5\eta^* + 10\eta^*C_m^*)\varphi - 21\eta^*\varphi^{5/3}}{-6(1 - \eta^* + 5\eta^*C_m^*)\varphi^{10/3}} \right], \quad (24b)$$

$$E = -\frac{7a^3}{12\delta_k}(2 + 5\eta^* + 10\eta^*C_m^*), \quad (24c)$$

$$F = -\frac{a^5}{2\delta_k}[7\eta^* + 2(1 - \eta^* + 5\eta^*C_m^*)\varphi^{5/3}], \quad (24d)$$

$$C_i = \frac{7}{4a^2\delta_k}(1 - \varphi^{5/3}), \quad (24e)$$

$$D_i = -\frac{21}{4\delta_k}(1 - \varphi^{5/3}), \quad (24f)$$

where

$$\delta_k = 7(1 + \eta^* + 5\eta^*C_m^*) - 5(2 + 5\eta^* + 10\eta^*C_m^*)\varphi + 21\eta^*\varphi^{5/3} + 3(1 - \eta^* + 5\eta^*C_m^*)\varphi^{10/3}. \quad (25)$$

Evaluation of the integral in Eq. (7) by using Eqs. (5), (17a), and Eq. (23) again produces the apparent viscosity

$$\zeta_k\varphi = 14Cb^2 + 4D + \frac{12}{5}(15Cb^2 + D - 4Fb^{-5}) + (5Cb^2 + D + Fb^{-5}) + 24\eta^*\varphi(C_i a^2)^2 \quad (26)$$

Substituting Eq. (24) into Eq. (26) produces

$$\zeta_k = \frac{1}{2\delta_k} \left[\frac{13(2 + 5\eta^* + 10\eta^*C_m^*) - 42\eta^*\varphi^{2/3}}{-12(1 - \eta^* + 5\eta^*C_m^*)\varphi^{7/3}} \right]$$

$$-\frac{15}{4\delta_k^2} \left[\frac{(2 + 5\eta^* + 10\eta^*C_m^*) - 7\eta^*\varphi^{2/3}}{-2(1 - \eta^* + 5\eta^*C_m^*)\varphi^{7/3}} \right]^2 + \frac{147\eta^*}{2\delta_k^2} (1 - \varphi^{5/3})^2. \quad (27)$$

In this model, the result of the apparent viscosity (by setting $C_m^* = 0$) is consistent with the formula of a mono-dispersion of fluid spheres (Keh and Chen, 2000). When $C_m^* = 0$ and $\eta^* \rightarrow \infty$, Eq. (27) agrees with the original article by Kuwabara (1959) describing the problem of apparent viscosity in a mono-dispersion of no-slip solid particles.

3. Results and Discussion

Tables 1 and 2 list the relative viscosity results η_e/η of a suspension of identical aerosol droplets for $C_m^* = 0.01$ and $\eta^* = 100$, respectively, obtained from the Simha, Happel and Kuwabara cell models in the previous section. The volume fractions of the suspended particles are considered for situations ranging from extremely dilute ($\varphi \rightarrow 0$) to very concentrated ($\varphi = 0.6$). Although the situations listed in Table 1 for an internal-to-external viscosity ratio of 0, 0.1 and even 1 almost never exist in actual aerosol suspensions, these data provide a good comparative reference and can also be applied to discuss a dispersed problem of hydrosol owing to our general formulation. Assuming the limiting values of $C_m^* = 0$ and $\eta^* \rightarrow \infty$, the calculations made herein using the three models agree with those presented in the classic works by Simha (1952), Happel (1957) and Kuwabara (1959). The appendix lists some limiting cases of the effective viscosity simplified by the three cell models.

Tables 1 and 2 clearly indicate the relative viscosity of the system increases monotonically with increasing particle volume fraction regardless of the physical properties specified. For a dilute aerosol dispersion (say, $\varphi \leq 0.2$), the relative

Table 1. The relative viscosity η_e/η for an aerosol suspension of identical liquid drops as a function of the volume fraction ϕ of the droplets with $C_m^* = 0.01$.

η^*	ϕ	η_e/η		
		Simha Model	Happel model	Kuwabara model
0	0	1.0000	1.0000	1.0000
	0.1	1.1329	1.2444	1.1749
	0.2	1.3823	1.5500	1.3991
	0.3	1.9023	1.9429	1.6934
	0.4	3.0919	2.4667	2.0921
	0.5	6.0938	3.2000	2.6571
	0.6	14.7236	4.3000	3.5120
0.1	0	1.0000	1.0000	1.0000
	0.1	1.1734	1.2881	1.2105
	0.2	1.5019	1.6522	1.4904
	0.3	2.1662	2.1334	1.8738
	0.4	3.6186	2.8021	2.4216
	0.5	7.1203	3.7932	3.2527
	0.6	16.7382	5.4027	4.6316
1	0	1.0000	1.0000	1.0000
	0.1	1.3287	1.4571	1.3329
	0.2	2.0265	2.0788	1.8462
	0.3	3.4597	3.0001	2.6623
	0.4	6.4663	4.4707	4.0245
	0.5	13.1800	7.0246	6.4592
	0.6	29.9586	11.9773	11.2688
10	0	1.0000	1.0000	1.0000
	0.1	1.4123	1.5695	1.3743
	0.2	2.3891	2.3862	2.0119
	0.3	4.5872	3.7030	3.1449
	0.4	9.5758	6.0494	5.2960
	0.5	21.4737	10.7138	9.7539
	0.6	52.7178	21.3363	20.1767
∞	0	1.0000	1.0000	1.0000
	0.1	1.4175	1.5850	1.3693
	0.2	2.4079	2.4244	2.0007
	0.3	4.6600	3.7829	3.1282
	0.4	9.8294	6.2203	5.2887
	0.5	22.3463	11.1231	9.8362
	0.6	55.8963	22.5124	20.7336

viscosity predicted by Happel cell model generally exceeds that predicted by the other two models. However, the Simha cell model replaces this primacy of Happel cell as the particle concentration increases. In a concentrated mono-dispersed aerosol, the Simha cell model results in markedly higher apparent viscosity than the Happel and Kuwabara cells, while the differences in relative viscosity between the Kuwabara and Happel cell models are fairly small. Generally, the apparent viscosity predicted by the Kuwabara cell model is

always smaller than that predicted by the Simha and Happel cell models, meaning that the energy dissipation in the particle motion with the Kuwabara cell model is less than with the other two models. Given a constant particle concentration, the relative viscosity of the aerosol suspension increases with increasing η^* and/or decreasing C_m^* , which explain why the energy dissipation during motion of the aerosol suspension increases with increasing droplet viscosity or decreasing slippage on the particle surface.

Table 2. The relative viscosity η_e/η for an aerosol suspension of identical liquid drops as a function of the volume fraction ϕ of the droplets with $\eta^* = 100$.

C_m^*	ϕ	η_e/η		
		Simha Model	Happel model	Kuwabara model
0	0	1.0000	1.0000	1.0000
	0.1	1.4345	1.6034	1.3791
	0.2	2.4945	2.4822	2.0401
	0.3	4.9547	3.9336	3.2479
	0.4	10.7459	6.6076	5.6282
	0.5	25.2099	12.1768	10.8206
	0.6	65.5982	25.7458	23.8774
0.1	0	1.0000	1.0000	1.0000
	0.1	1.3141	1.4708	1.3140
	0.2	1.9843	2.0976	1.7952
	0.3	3.3748	3.0152	2.5591
	0.4	6.3256	4.4718	3.8409
	0.5	12.9876	7.0035	6.1580
	0.6	29.7964	11.9462	10.8125
1	0	1.0000	1.0000	1.0000
	0.1	1.1724	1.2968	1.2101
	0.2	1.4997	1.6662	1.4876
	0.3	2.1625	2.1510	1.8664
	0.4	3.6135	2.8220	2.4069
	0.5	7.1141	3.8147	3.2271
	0.6	16.7776	5.4256	4.5898
10	0	1.0000	1.0000	1.0000
	0.1	1.1374	1.2505	1.1790
	0.2	1.3952	1.5631	1.4092
	0.3	1.9305	1.9658	1.7126
	0.4	3.1475	2.5052	2.1263
	0.5	6.2010	3.2655	2.7179
	0.6	14.9367	4.4181	3.6252
∞	0	1.0000	1.0000	1.0000
	0.1	1.1329	1.2444	1.1749
	0.2	1.3823	1.5500	1.3991
	0.3	1.9023	1.9429	1.6934
	0.4	3.0919	2.4667	2.0921
	0.5	6.0938	3.2000	2.6571
	0.6	14.7236	4.3000	3.5120

Taking Taylor expansion about $\phi = 0$, the apparent viscosity can be represented in the form of an increasing power series of $\phi^{1/3}$ as

$$\left(\frac{\eta_e}{\eta}\right)_s = 1 + \left[\frac{2 + 5\eta^* + 10\eta^* C_m^*}{2(1 + \eta^* + 5\eta^* C_m^*)} + \frac{3\eta^*}{2(1 + \eta^* + 5\eta^* C_m^*)^2} \right] \phi + \left[\frac{5(2 + 5\eta^* + 10\eta^* C_m^*)^2}{8(1 + \eta^* + 5\eta^* C_m^*)^2} + \frac{15\eta^*(2 + 5\eta^* + 10\eta^* C_m^*)}{4(1 + \eta^* + 5\eta^* C_m^*)^3} \right] \phi^2 + \left[\frac{105\eta^*(\eta^* + 2\eta^* C_m^*)}{4(1 + \eta^* + 5\eta^* C_m^*)^2} - \frac{63\eta^{*2}}{2(1 + \eta^* + 5\eta^* C_m^*)^3} \right] \phi^{8/3} + O(\phi^3) \quad (28a)$$

for the Simha cell model,

$$\left(\frac{\eta_e}{\eta}\right)_h = 1 + \left[\frac{11(2 + 5\eta^* + 10\eta^* C_m^*)}{10(1 + \eta^* + 5\eta^* C_m^*)} + \frac{3\eta^*}{2(1 + \eta^* + 5\eta^* C_m^*)^2} \right] \phi - \frac{21\eta^*}{5(1 + \eta^* + 5\eta^* C_m^*)} \phi^{5/3} + \left[\frac{11(2 + 5\eta^* + 10\eta^* C_m^*)^2}{20(1 + \eta^* + 5\eta^* C_m^*)^2} + \frac{3\eta^*(2 + 5\eta^* + 10\eta^* C_m^*)}{2(1 + \eta^* + 5\eta^* C_m^*)^3} \right] \phi^2 - \frac{21\eta^*(2 + 5\eta^* + 10\eta^* C_m^*)}{10(1 + \eta^* + 5\eta^* C_m^*)^2} \phi^{8/3} + O(\phi^3) \quad (28b)$$

for the Happel cell model, and

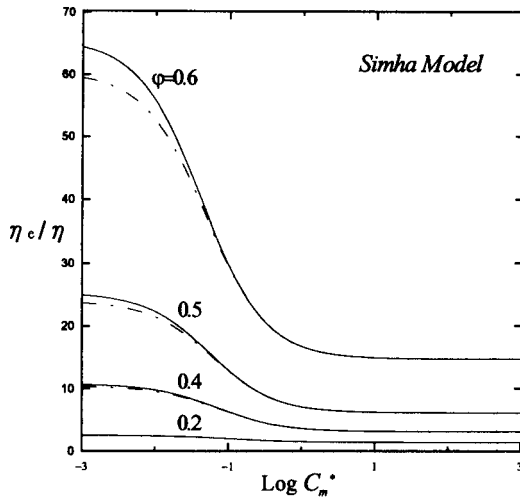


Fig. 1a

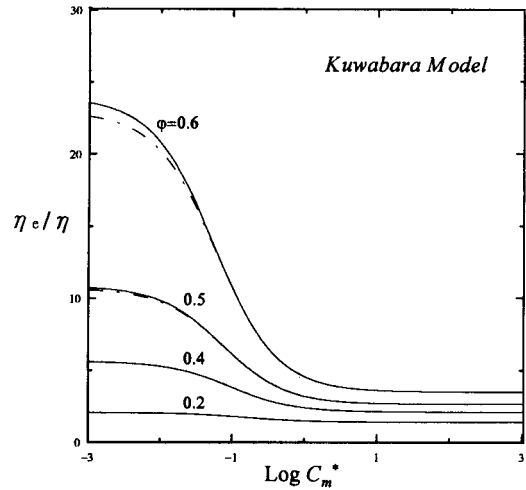


Fig. 1c

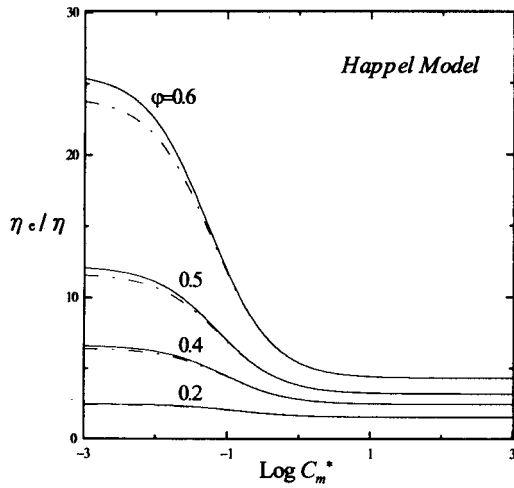


Fig. 1b

$$\left[\frac{15\eta^*(12 + 23\eta^* + 46\eta^*C_m^*)}{14(1 + \eta^* + 5\eta^*C_m^*)^2} + \frac{9\eta^{*2}}{(1 + \eta^* + 5\eta^*C_m^*)^3} - \frac{195\eta^*(2 + 5\eta^* + 10\eta^*C_m^*)^2}{98(1 + \eta^* + 5\eta^*C_m^*)^3} \right] \varphi^{8/3} + O(\varphi^3) \tag{28c}$$

Fig. 1. Plots of the relative viscosity η_e/η for a mono-dispersed aerosol of liquid drops as a function of C_m^* with the volume fraction φ as a parameter : solid line for $\eta^* = 100$ and dash line for $\eta^* = 10$.

$$\left(\frac{\eta_e}{\eta} \right)_k = 1 + \left[\frac{13(2 + 5\eta^* + 10\eta^*C_m^*)}{14(1 + \eta^* + 5\eta^*C_m^*)} + \frac{3\eta^*}{2(1 + \eta^* + 5\eta^*C_m^*)^2} - \frac{15(2 + 5\eta^* + 10\eta^*C_m^*)^2}{196(1 + \eta^* + 5\eta^*C_m^*)^2} \right] \varphi - \left[\frac{3\eta^*}{1 + \eta^* + 5\eta^*C_m^*} - \frac{15\eta^*(2 + 5\eta^* + 10\eta^*C_m^*)}{14(1 + \eta^* + 5\eta^*C_m^*)^2} \right] \varphi^{5/3} + \left[\frac{65(2 + 5\eta^* + 10\eta^*C_m^*)^2}{98(1 + \eta^* + 5\eta^*C_m^*)^2} + \frac{15\eta^*(2 + 5\eta^* + 10\eta^*C_m^*)}{7(1 + \eta^* + 5\eta^*C_m^*)^3} - \frac{75(2 + 5\eta^* + 10\eta^*C_m^*)^3}{686(1 + \eta^* + 5\eta^*C_m^*)^3} \right] \varphi^2 - \frac{15\eta^{*2}}{4(1 + \eta^* + 5\eta^*C_m^*)^2} \varphi^{7/3}$$

for the Kuwabara cell model. The above result shows that the first three leading orders of the effect of particle concentration on the aerosol viscosity are $(\varphi, \varphi^2, \varphi^{8/3})$ for Simha cell and $(\varphi, \varphi^{5/3}, \varphi^2)$ for Happel cell and Kuwabara cell, respectively. For a dilute or moderately concentrated aerosol comprising identical particles, Eq. (28) accurately predicts the apparent system viscosity.

In Figures 1 and 2, the relative viscosity of a mono-dispersed aerosol is plotted as a function of C_m^* and η^* , respectively, for the Simha, Happel and Kuwabara cell models. These figures verifies that the existence of particles will make the bulk viscosity of the suspension stickier than the surrounding medium, regardless of the droplet viscosity and/or the surface slippage. This viscosity-increase becomes significant with increasing volume fraction of suspended particles. Again, the effective viscosity of the aerosol system decreases with increasing slip coefficient and Knudsen number, meaning less energy is consumed in the suspension flow. However, the bulk viscosity of the mono-dispersed aerosol is a weak function of C_m^* when $C_m^* > 10$; that is, when

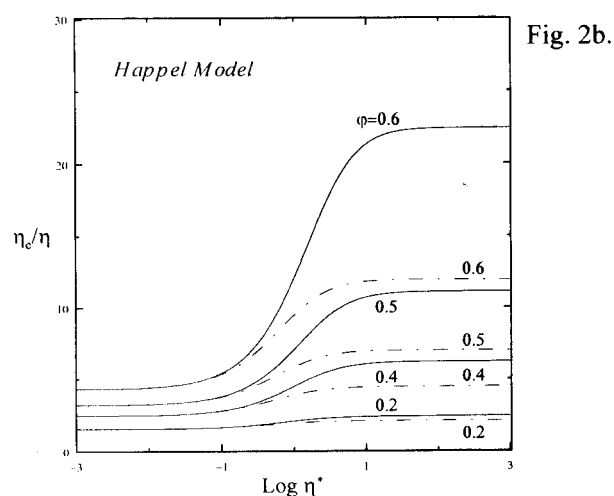
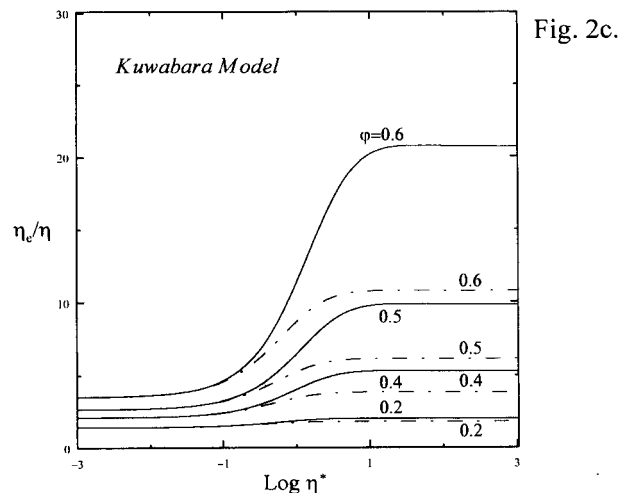
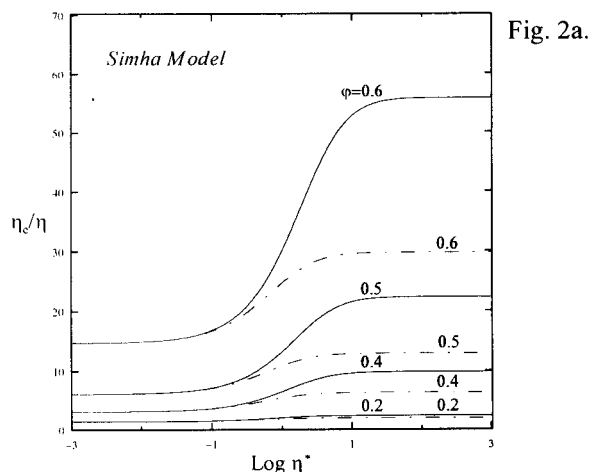


Fig. 2. Plots of the relative viscosity η_c/η for a mono-dispersed aerosol of liquid drops as a function of η^* with the volume fraction ϕ as a parameter: solid line for $C_m^* = 0.01$ and dash line for $C_m^* = 0.1$.

$C_m^* > 10$, the changes of C_m^* will only slightly influence the apparent viscosity. Meanwhile, bulk viscosity increases with increasing droplet viscosity, as displayed in Fig. 2. The increase of the viscosity affects the shear stress at the droplet-gas interface and thus causes the phenomenon of the increased energy expenditure in transport.

An analogous phenomenon to the physical property of C_m^* , the apparent viscosity of liquid aerosol is a weak function of η^* because $\eta^* < 0.1$. Figures 1 and 2 reveal that changes in C_m^* and/or η^* significantly influence effective viscosity in concentrated suspensions, while in dilute aerosols the effective viscosity is a weak function of C_m^* and η^* . In general, the tendency of the bulk

viscosity influenced by the physical properties (C_m^* and η^*) and volume fraction of the droplets is similar for the three cell models, though their numerical evaluations do not all agree.

Figures 3 and 4 illustrate the plots of effective viscosity as a function of C_m^* and η^* for given a constant volume fraction of the droplet ($\phi = 0.6$). Interestingly, the apparent viscosity evaluated by the three cell models is not influenced by variations in η^* when $C_m^* > 10$ or by variations in C_m^* when $\eta^* < 0.01$. However, this phenomenon does not mean that predictions of bulk viscosity with the three cell models have the same limitations. These limited bulk viscosities are 14.7, 4.3 and 3.5 for the Simha, Happel and Kuwabara cell models, respectively. From the perspective of hydrodynamics, these limitations are identical to those for a mono-dispersion of gas bubbles in a liquid medium, though they are not applicable to aerosol samples. Conversely, the apparent viscosity is a sensitive function of η^* when C_m^* is small, and a sensitive function of C_m^* when η^* is large. In the concentrated aerosol illustrated in Figs. 3 and 4, the Simha cell again reveals much higher energy dissipation in the hydrodynamic motion of the particle relative to the surrounding gas than the other models do, resulting in higher aerosol viscosity.

4. Conclusion

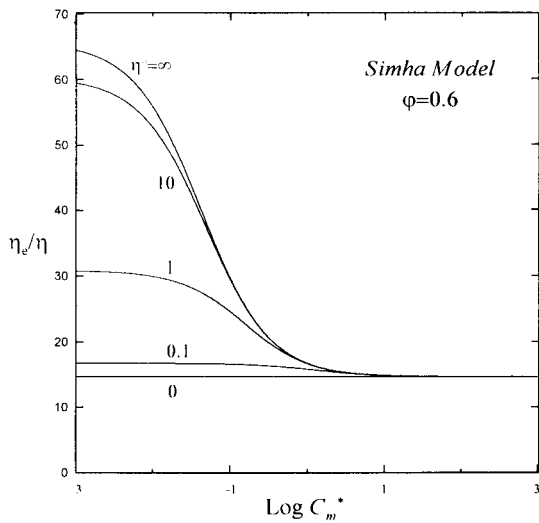


Fig. 3a.

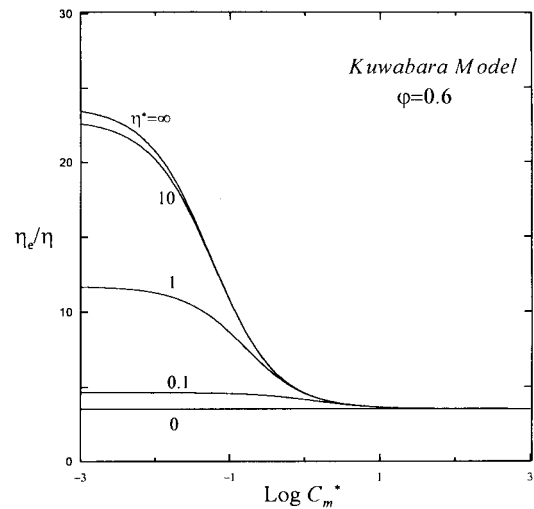


Fig. 3c.

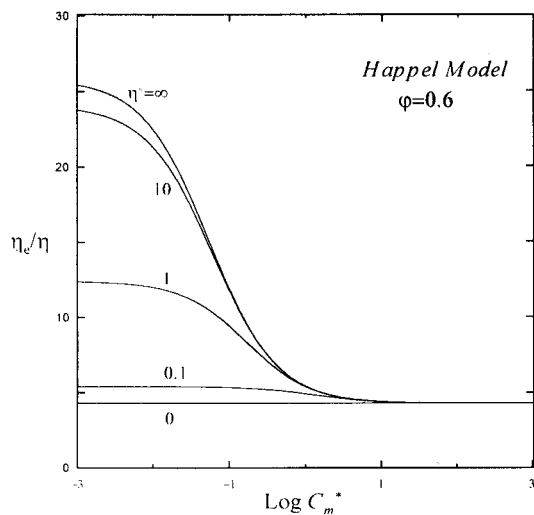


Fig. 3b.

Fig. 3. Plots of the relative viscosity η_c/η for a mono-dispersed aerosol of liquid drops as a function of C_m^* with η^* as a parameter ($\phi = 0.6$).

This study analytically formulates the effective viscosity of a mono-dispersed aerosol of liquid droplets by using three types of cell models: (a) Simha cell model, (b) Happel cell model and (c) Kuwabara cell model. Increased droplet viscosity and/or decreased slippage feature at the droplet-gas interface are both demonstrated to increase the apparent viscosity of the mono-dispersed aerosol, implying a corresponding increase in energy dissipation during aerosol transport. Although the discussions in the previous section focus on the liquid aerosol suspension, the general results of

Eqs. (15), (21) and (27) can be simplified to some classic investigations of the effective viscosity of colloidal systems, such as no-slip solid particles in a fluid medium (gas or liquid), solid aerosol particles in the gas phase, gas bubbles in the liquid phase, macro-molecule suspension, and so on. Notably the cell model can be viewed as an art methodology. Researchers who use experimental studies can always find suitable cell models to predict their data, at least in some specified concentrations they treated. Consequently, it is extremely difficult to determine which cell model is better. This work merely presents a pre-experimental theoretical study to those who are interested.

Acknowledgement

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Appendix

Some Limiting Cases of on Effective Viscosity in the Simha-Type Cell Model

Case I: Monodispersion of Liquid Drops ($C_m^* = 0$)

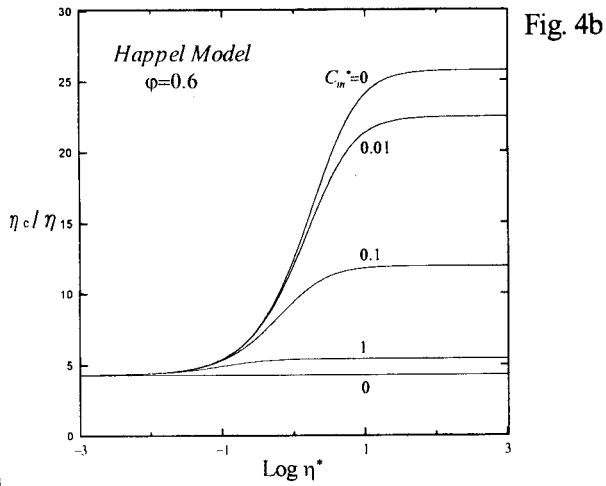
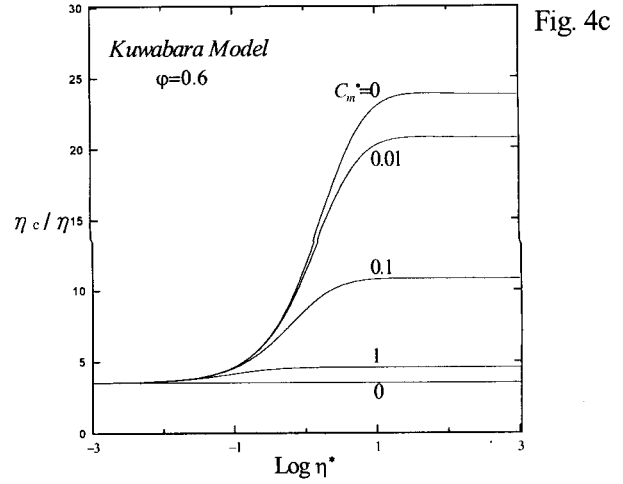
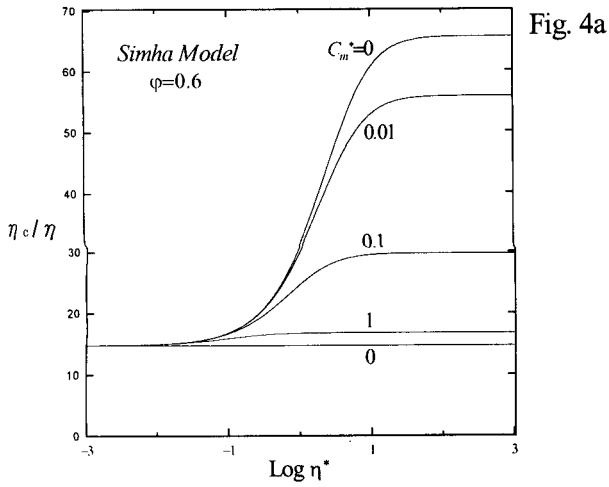


Fig. 4. Plots of the relative viscosity η_e/η for a mono-dispersed aerosol of liquid drops as a function of η^* with C_m^* as a parameter ($\phi = 0.6$).

$$\zeta_s = \frac{2}{\delta_s} [(2 + 5\eta^*) + 5(1 - \eta^*)\phi^{7/3}] + \frac{6\eta^*}{\delta_s^2} (2 - 7\phi^{5/3} + 5\phi^{7/3})^2 \quad (A1a)$$

$$\delta_s = 4(1 + \eta^*) - 5(2 + 5\eta^*)\phi + 42\eta^*\phi^{5/3} + 5(2 - 5\eta^*)\phi^{7/3} - 4(1 - \eta^*)\phi^{10/3} \quad (A1b)$$

Case II: Monodispersion of Solid Aerosol Spheres ($\eta^* \rightarrow \infty$)

$$\zeta_s = \frac{10}{\delta_s} [(1 + 2C_m^*) - (1 - 5C_m^*)\phi^{7/3}] \quad (A2a)$$

$$\delta_s = 4(1 + 5C_m^*) - 25(1 + 2C_m^*)\phi + 42\phi^{5/3}$$

$$- 25(1 - 2C_m^*)\phi^{7/3} + 4(1 - 5C_m^*)\phi^{10/3} \quad (A2b)$$

Case III: Monodispersion of No-slip Solid Particles ($C_m^* = 0, \eta^* \rightarrow \infty$)

$$\zeta_s = \frac{10}{\delta_s} (1 - \phi^{7/3}) \quad (A3a)$$

$$\delta_s = 4 - 25\phi + 42\phi^{5/3} - 25\phi^{7/3} + 4\phi^{10/3} \quad (A3b)$$

Case IV: Monodispersion of Gas Bubbles

($C_m^* = 0^*, \eta^* = 0$ or $C_m^* \rightarrow \infty, \eta^* \rightarrow \infty$)

$$\zeta_s = \frac{1}{\delta_s} (2 + 5\phi^{7/3}) \quad (A4a)$$

$$\delta_s = 2 - 5\phi + 5\phi^{7/3} - 2\phi^{10/3} \quad (A4b)$$

Some Limiting cases of Effective Viscosity in the Happel-Type Cell Model

Case I: Monodispersion of Liquid Drops ($C_m^* = 0$)

$$\zeta_h = \frac{1}{5\delta_h} [11(2 + 5\eta^*) - 42\eta^*\phi^{2/3} - 22(1 - \eta^*)\phi^{7/3}] + \frac{6\eta^*}{\delta_h^2} (1 - \phi^{7/3})^2 \quad (A5a)$$

$$\delta_h = 2(1 + \eta^*) - (2 + 5\eta^*)\phi - (2 - 5\eta^*)\phi^{7/3} + 2(1 - \eta^*)\phi^{10/3} \quad (A5b)$$

Case II: Monodispersion of Solid Aerosol Spheres
($\eta^* \rightarrow \infty$)

$$\zeta_h = \frac{1}{5\delta_h} [55(1 + 2C_m^*) - 42\varphi^{2/3} + 22(1 - 5C_m^*)\varphi^{7/3}] \quad (\text{A6a})$$

$$\delta_h = 2(1 + 5C_m^*) - 5(1 + 2C_m^*)\varphi + 5(1 - 2C_m^*)\varphi^{7/3} - 2(1 - 5C_m^*)\varphi^{10/3} \quad (\text{A6b})$$

Case III: Monodispersion of No-slip Solid Particles
($C_m^* = 0, \eta^* \rightarrow \infty$)

$$\zeta_h = \frac{1}{5\delta_h} (55 - 42\varphi^{2/3} + 22\varphi^{7/3}) \quad (\text{A7a})$$

$$\delta_h = 2 - 5\varphi + 5\varphi^{7/3} - 2\varphi^{10/3} \quad (\text{A7b})$$

Case IV: Monodispersion of Gas Bubbles ($C_m^* = 0, \eta^* = 0$ or $C_m^* \rightarrow \infty, \eta^* \rightarrow \infty$)

$$\zeta_h = \frac{11}{\delta_s} (1 - \varphi^{7/3}) \quad (\text{A8a})$$

$$\delta_h = 1 - \varphi - \varphi^{7/3} + \varphi^{10/3} \quad (\text{A8b})$$

Some Limiting cases of Effective Viscosity in the Kuwabara-Type Cell Model

Case I: Monodispersion of Liquid Drops ($C_m^* = 0$)

$$\zeta_k = \frac{1}{2\delta_k} [13(2 + 5\eta^*) - 42\eta^*\varphi^{2/3} - 12(1 - \eta^*)\varphi^{7/3}] - \frac{15}{4\delta_k^2} [(2 + 5\eta^*) - 7\eta^*\varphi^{2/3} - 2(1 - \eta^*)\varphi^{7/3}]^2 + \frac{147\eta^*}{2\delta_k^2} (1 - \varphi^{5/3})^2 \quad (\text{A9a})$$

$$\delta_k = 7(1 + \eta^*) - 5(2 + 5\eta^*)\varphi + 21\eta^*\varphi^{5/3} + 3(1 - \eta^*)\varphi^{10/3} \quad (\text{A9b})$$

Case II: Monodispersion of Solid Aerosol Spheres
($\eta^* \rightarrow \infty$)

$$\zeta_k = \frac{1}{2\delta_k} [65(1 + 2C_m^*) - 42\varphi^{2/3} + 12(1 - 5C_m^*)\varphi^{7/3}]$$

$$- \frac{15}{4\delta_k^2} [5(1 + 2C_m^*) - 7\varphi^{2/3} + 2(1 - 5C_m^*)\varphi^{7/3}]^2 \quad (\text{A10a})$$

$$\delta_k = 7(1 + 5C_m^*) - 25(1 + 2C_m^*)\varphi + 21\varphi^{5/3} - 3(1 - 5C_m^*)\varphi^{10/3} \quad (\text{A10b})$$

Case III: Monodispersion of No-slip Solid Particles
($C_m^* = 0, \eta^* \rightarrow \infty$)

$$\zeta_k = \frac{1}{5\delta_k} (65 - 42\varphi^{2/3} + 12\varphi^{7/3}) - \frac{152\delta_k}{4\delta_k^2} (5 - 7\varphi^{2/3} + 2\varphi^{7/3})^2 \quad (\text{A11a})$$

$$\delta_k = 7 - 25\varphi + 21\varphi^{5/3} - 3\varphi^{10/3} \quad (\text{A11b})$$

Case IV: Monodispersion of Gas Bubbles ($C_m^* = 0, \eta^* = 0$ or $C_m^* \rightarrow \infty, \eta^* \rightarrow \infty$)

$$\zeta_k = \frac{1}{\delta_k} (13 - 6\varphi^{7/3}) - \frac{15}{2\delta_k^2} (1 - \varphi^{7/3})^2 \quad (\text{A12a})$$

$$\delta_k = 7 - 10\varphi + 3\varphi^{10/3} \quad (\text{A12b})$$

Nomenclature

a	radius of a fluid sphere (m)
b	radius of a unit cell (m)
C, D, E, F, C_i, D_i	coefficients defined by Equation (8)
C_m	frictional slip coefficient
C_m^*	($= C_m l / a$)
$\mathbf{e}_r, \mathbf{e}_\theta, \mathbf{e}_\phi$	unit vectors in the spherical coordinate system
$\mathbf{e}_x, \mathbf{e}_y, \mathbf{e}_z$	unit vectors in the rectangular coordinate system
E^2	Stokes operator defined by Equation (7) (m^{-2})
\mathbf{F}	force exerted on a particle by the external fluid (N)
\mathbf{I}	unit dyadic
Kn	Knudsen number
l	mean free path of the external fluid (m)

p	dynamic pressure, N m^{-2}
(r, θ, ϕ)	the spherical coordinates
\mathbf{v}	velocity field (m s^{-1})
\mathbf{v}_∞	prescribed velocity field (m s^{-1})
\mathbf{x}	position vector (m)
(x, y, z)	the rectangular coordinates

Greek Letters

α	momentum accommodation coefficient
γ	interfacial tension (Nm^{-1})
ζ	coefficient defined in Equation (2.8)
η	fluid viscosity ($\text{kg m}^{-1} \text{s}^{-1}$)
η^*	internal-to-external viscosity ratio of a droplet
κ	absolute constant velocity gradient, s^{-1}
τ	viscous stress dyadic (Nm^{-2})
φ	volume fraction of particles in suspension

Subscripts

i	properties inside the fluid sphere
e	effective properties
h	coefficient related to Happel cell model
k	coefficient related to Kuwabara cell model
s	coefficient related to Simha cell model

Superscripts

*	dimensionless group
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