# The bulk stress in a suspension of spherical particles of condensed phase in its slightly rarefied vapour gas

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The rheological behaviour of a dilute suspension of spherical particles of condensed phase dispersed in its own slightly rarefied vapour gas is investigated on the basis of suspension theory (Batchelor 1970) and generalized slip-flow theory for a two-phase system of a gas and its condensed phase derived from the Boltzmann equation. The rarefaction of the gas and the phase-change process at the interfaces of the particles have the effect of reducing the Einstein coefficient of $\phi$, volume fraction, in the expression for the effective viscosity in the suspension. In the case in which the pure rarefaction effect alone enters the problem, the coefficient is $\frac{5}{2}(1-2\cdot702\,K)$, where $K$ is the Knudsen number, a rarefaction parameter defined by $K = l/L$, $l$ and $L$ being respectively the mean free path of gas molecules and the radius of a spherical particle. When both the rarefaction and the phase-change process are taken into account, this becomes $\frac{5}{2}(1-3\cdot533\,K)$. These modifications are not small, even at ordinary pressures, when the size of the particles is of the order of microns.

## 1. Introduction

One of the important mechanical properties of the dilute suspension of spherical particles in a fluid of viscosity $\mu$ is the effective viscosity $\mu^*$, which can be expressed as

$$\mu^* = \mu[1 + \beta\phi + O(\phi^2)], \tag{1.1}$$

where $\phi$ is the fraction of the volume occupied by the suspended particles. The coefficient $\beta$ was calculated to be $\frac{5}{2}$ for rigid spherical particles by Einstein (1906, 1911), and to be $(\mu+\frac{5}{2}\mu')/(\mu+\mu')$ for spherical fluid particles of viscosity $\mu'$ by Taylor (1932). These values of $\mu^*$ have been derived on the assumption of no slip and no change of phase at the surface of the particles, in addition to the usual assumptions that the particles are so far apart to be hydrodynamically independent of each other and that the effect of inertia on the motion is negligible. When we think of small particles floating in a gas, we must be careful about rarefaction effects which often induce flows of various kinds that cannot be anticipated within the framework of continuum theory, such as the thermal creep flow due to the non-uniform temperature distribution of a gas over a boundary surface (Kennard 1938, p. 327; Sone 1966), and the thermal stress slip flow due to the non-uniform normal temperature gradient over a surface (Sone 1972). The degree of rarefaction is measured by the Knudsen number $K$, the ratio of the molecular mean free path $l$ to the characteristic length of the system $L$ (e.g. the radius of a typical particle), and it becomes appreciable when $L$ becomes small. Even at ordinary pressures in air we have $K \simeq 0\cdot1$ when $L \simeq 1\ \mu\text{m}$. It may now

be worthwhile to investigate the effects of rarefaction, including the phase change at the interface between the gas and the condensed phase, on the effective viscosity of the suspension as a whole, and this is the purpose of this paper.

For this study, we consider a dilute suspension in which small spherical particles of condensed phase are suspended in its slightly rarefied vapour gas (in the sense that the Knudsen number is small but not negligible) with the same temperature as that of the particles. The appropriate analysis of the motion of slightly rarefied gases must be based on kinetic theory, and we use the asymptotic analysis for small Knudsen numbers for evaporation (or sublimation) and condensation by Sone & Onishi (1978), thus avoiding the trouble of dealing with the kinetic equation as it stands. This analysis gives a generalized slip flow theory for a two-phase system of a gas and its condensed phase derived systematically from the linearized kinetic equation, and gives a uniformly valid description of the steady behaviour of gas coexisting with its own condensed phase of arbitrary shape (with certain smoothness) and configuration. An outline of the theory is given in the next section.

## 2. Outline of the asymptotic theory for a two-phase system

The asymptotic theory by Sone & Onishi (1978) for small Knudsen numbers is a theory which has reduced a kinetic treatment of given problems for small Knudsen numbers to a hydrodynamic one. This is based on the following assumptions: (i) the gas motion is fully described by the Boltzmann–Krook–Welander equation (Bhatnager, Gross & Krook 1954; Welander 1954; Kogan 1958); (ii) the interaction of gas molecules with the condensed phase is of diffusive type characterized by the condition of the saturated (vapour) gas at the same temperature as that of the condensed phase (see for example Pao 1971); (iii) the deviation of the system under consideration from a stationary equilibrium state is so small that the governing equation and the boundary condition may be linearized, i.e. the Mach number (the ratio of the characteristic velocity of the motion of the system, e.g. the suspension, to the sound speed at the reference state) is very small and the second and higher orders are completely neglected since the Mach number is a measure of the deviation in the standard non-dimensional form of the kinetic equation.

According to the theory, the desired solution can be obtained in terms of the Hilbert part (subscript $H$) and the Knudsen-layer part (subscript $K$), i.e.

$$f = f_H + f_K, \qquad (2.1)$$

where $f$ represents any one of the perturbations of hydrodynamic quantities (such as gas velocity, pressure, temperature, stress, etc.) from a certain equilibrium reference state. The Hilbert part $f_H$ has a length scale of variation of the order of the characteristic length of the system $L$ (e.g. the radius of a representative particle) which is much larger than the mean free path $l$ of gas molecules and dominates over the whole flow region except in general a thin layer of the thickness of the order of the mean free path adjacent to the interface between the gas and the condensed phase. The Knudsen-layer part $f_K$ has a length scale of variation of the order of the mean free path and is appreciable only in the thin layer, called the Knudsen layer, expressing the Knudsen-layer correction to the Hilbert part in the immediate vicinity of the interface. $f_K$ is assumed to vanish sufficiently rapidly with the distance from the interface (e.g. a

few mean-free paths away from it). Each part is obtained in a power series of a quantity of the order of the Knudsen number, namely

$$f_H = f_H^{(0)} + k f_H^{(1)} + k^2 f_H^{(2)} + \dots, \tag{2.2}$$

$$f_K = f_K^{(0)} + k f_K^{(1)} + k^2 f_K^{(2)} + \dots . \tag{2.3}$$

The parameter $k$ is defined in terms of the Knudsen number $K$ by

$$k = \frac{\sqrt{\pi}}{2} \frac{l}{L} = \frac{\sqrt{\pi}}{2} K \tag{2.4}$$

and the mean free path $l$ is related to the viscosity $\mu$ of the gas as

$$\mu = P_0 (8RT_0/\pi)^{-\frac{1}{2}} l, \tag{2.5}$$

where $R$ is the gas constant, $P_0$ a reference pressure and $T_0$ a reference temperature.

The Hilbert part of the velocity, pressure and temperature satisfies the following Stokes system of hydrodynamic equations at any order of the Knudsen number:

$$\nabla p_H^{(0)} = 0, \quad \nabla \cdot \mathbf{u}_H^{(m)} = 0, \quad \nabla p_H^{(m+1)} - \nabla^2 \mathbf{u}_H^{(m)} = 0, \qquad (2.6), (2.7), (2.8)$$

$$\nabla^2 \tau_H^{(m)} = 0, \quad \rho_H^{(m)} = p_H^{(m)} - \tau_H^{(m)}, \qquad (2.9), (2.10)$$

where $m = 0, 1, 2, \dots$, $P_0(1+p)$, $(2RT_0)^{\frac{1}{2}} \mathbf{u}$, $T_0(1+\tau)$ and $\rho_0(1+\rho)$ are the gas pressure, velocity vector, temperature and density, respectively, and $\rho_0$ is the reference density defined by $\rho_0 = P_0(RT_0)^{-1}$. $\nabla$ is the gradient operator with respect to $\mathbf{x}$, $L\mathbf{x}$ being the rectangular co-ordinates. Equation (2.10) is the linearized form of the equation of state for an ideal gas. It is noted that $(2RT_0)^{\frac{1}{2}}$ is taken as the reference velocity which is of the order of the sound speed at temperature $T_0$. This system of equations governs the whole flow field except the thin Knudsen layer attached to the interface.

The boundary conditions at the interface appropriate for this system of equations (2.6)–(2.9) are obtained by the Knudsen-layer analysis and are given in the form of the generalized slip conditions in appendix A [(A 1), (A 4), (A 5), (A 8), (A 11) and (A 12)]. Together with the boundary conditions, is also listed in appendix A the Knudsen-layer part of the solution which is expressed in terms of the universal functions and the values of the Hilbert part evaluated at the interface surface. Furthermore two other important quantities, i.e. the stress tensor and heat-flow vector, and their Knudsen-layer parts are also given there.

Incidentally, a brief mention is made here of the local stress [denoted by $P_0(\mathbf{I} + \mathbf{P})$ in appendix A] which will be needed in section 4. This has the following parts:

$$P_0(\mathbf{I} + \mathbf{P}) = P_0(\mathbf{I} + \mathbf{P}_H + \mathbf{P}_K), \tag{2.11}$$

where $\mathbf{I}$ is the unit tensor, and $\mathbf{P}$ and $\mathbf{P}_H$ both satisfy the following equations

$$\nabla \cdot \mathbf{P} = 0, \quad \nabla \cdot \mathbf{P}_H = 0. \tag{2.12}$$

The fact that the divergence of the stress is zero is easily shown from the original linearized Boltzmann–Krook–Welander equation, and the second equation is none other than the equation (2.8). If the points outside the thin Knudsen layer at the interfaces of the particles are considered, the contribution to the stress from the Knudsen-layer part is negligible and the stress (2.11) can be expressed only by the Hilbert part. Substituting the expression for $\mathbf{P}_H$ from (A 17) in appendix A, we have

$$P_0(\mathbf{I} + \mathbf{P}) = P_0(1 + p_H^{(0)}) \mathbf{I} - k P_0 \mathbf{\Pi}, \tag{2.13}$$

where $\boldsymbol{\Pi}$, to the order of $k$, is given by

$$\boldsymbol{\Pi} = -(p_H^{(1)} + kp_H^{(2)})\mathbf{I} + \boldsymbol{\epsilon}_0 + k(\boldsymbol{\epsilon}_1 - \nabla\nabla\tau_H^{(0)}). \tag{2.14}$$

The first term on the right-hand side of (2.13) depends on the choice of the reference pressure and is of no particular importance. $kP_0$ can be replaced by the viscosity $\mu$ of the gas using the relations (2.4) and (2.5), i.e. $kP_0 = \mu(2RT_0)^{\frac{1}{2}}L^{-1}$. It is noted that from (2.13) and (2.14) when the temperature field in the continuum limit is uniform, i.e. $\tau_H^{(0)} \equiv 0$, the stress tensor for slightly rarefied gases at points outside the Knudsen layer has exactly the same form as that for Newtonian fluids.

In the next section we study a fundamental problem for a suspension, namely the motion of a slightly rarefied gas around a particle of the condensed phase immersed in a pure straining motion, using the results of this section.

## 3. Velocity and pressure fields around a spherical condensed phase in a pure straining motion

Consider the following problem: a spherical condensed phase of radius $L$ maintained at a constant temperature $T_0$ is placed in a uniform pure straining motion of its own gas whose temperature and pressure at infinity are $T_0$ and $P_0$, respectively, $P_0$ being chosen as the saturated gas pressure corresponding to $T_0$ for simplicity. We assume that the ratio of the viscosity of the gas to that of the condensed phase (liquid or solid) is so small that the internal flow of the condensed phase can be neglected.† The uniform pure straining motion is specified by the constant symmetric rate-of-strain tensor $\mathbf{E}$ with the trace $(\mathbf{E}) = 0$.

Let $L$, $T_0$, and $P_0$ be taken as the reference length, temperature and pressure, respectively, and also the origin of the co-ordinate system be at the centre of the condensed phase. Then from the choice of the reference state, $p_w$ and $\tau_w$ are all zero‡ which appear in the generalized slip conditions in appendix A, and also $\mathbf{u}_w = 0$ from the symmetry of the problem. Furthermore, we have the following conditions to be satisfied at infinity,

$$\left.\begin{array}{l} \mathbf{u}_H \to \mathbf{e}.\mathbf{x}, \quad p_H \to 0 \\ \tau_H \to 0 \end{array}\right\} \quad \text{as} \quad r \to \infty, \tag{3.1}$$

where $r = (\mathbf{x}.\mathbf{x})^{\frac{1}{2}}$ and $\mathbf{e} = \mathbf{e}^{(0)} + k\mathbf{e}^{(1)} + \dots$, $\mathbf{e}$ being the non-dimensional rate-of-strain tensor defined by $\mathbf{e} = (2RT_0)^{-\frac{1}{2}}L\mathbf{E}$.

From (2.6), we immediately obtain the solution for $p_H^{(0)}$ subject to (3.1) at infinity as§

$$p^{(0)}(\mathbf{x}) = 0, \tag{3.2}$$

which gives from (A 4) the normal velocity at the interface of the condensed phase

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† Although the viscosity ratio may not be sufficiently small in the case of gas–liquid systems (e.g. the ratio $\simeq 7 \times 10^{-3}$ for $H_2O$ at ordinary conditions), we believe that the qualitative nature of the problem is retained under this assumption.

‡ The effect of surface tension on this pressure is small in ordinary conditions (see appendix B), and will be neglected. On the other hand the particles may be considered to be held spherical by the surface tension $\alpha$, which is equivalent to the assumption that $\alpha(\mu LE)^{-1} \gg 1$, $E$ being a representative magnitude of $\mathbf{E}$.

§ We shall henceforth omit the subscript $H$, on the understanding that the hydrodynamic quantities that appear in this section and the following refer to the Hilbert part unless otherwise stated explicitly.

(evaporation or condensation rate). This normal velocity, together with (A 1), constitutes the boundary condition for the velocity $\mathbf{u}^{(0)}$ and also provides the boundary condition for the temperature $\tau^{(0)}$ from (A 5). Namely we have

$$\left.\begin{aligned} \mathbf{u}^{(0)}.\mathbf{t} = \mathbf{u}^{(0)}.\mathbf{n} = 0 \\ \tau^{(0)} = 0 \end{aligned}\right\} \quad \text{at} \quad r = 1, \tag{3.3}$$

and also at infinity ($r \to \infty$),

$$\mathbf{u}^{(0)} \to \mathbf{e}^{(0)}.\mathbf{x}, \quad p^{(1)} \to 0, \quad \tau^{(0)} \to 0. \tag{3.4}$$

Equation (3.3) shows that at the present stage of approximation (continuum limit), no evaporation and condensation process occurs at the interface. The solution to (2.7)–(2.9) with $m = 0$ subject to these conditions (3.3) and (3.4) is easily obtainable (Landau & Lifshitz 1966, § 22; Batchelor 1967, § 4.11), and it is given as

$$\left.\begin{aligned} \mathbf{u}^{(0)} &= \left(1 + \frac{b_0}{r^5}\right)\mathbf{e}^{(0)}.\mathbf{x} + \left(\frac{a_0}{2r^5} - \frac{5b_0}{2r^7}\right)\mathbf{x}.\mathbf{e}^{(0)}.\mathbf{xx}, \\ p^{(1)} &= \frac{a_0}{r^5}\mathbf{x}.\mathbf{e}^{(0)}.\mathbf{x}, \quad \tau^{(0)} = 0, \end{aligned}\right\} \tag{3.5}$$

with

$$a_0 = -5, \quad b_0 = -1. \tag{3.6}$$

The Knudsen-layer part does not exist in this stage of approximation because $\mathbf{u}^{(0)}.\mathbf{n} = 0$ at $r = 1$ [see (A 2), (A 3), (A 6), (A 7) and (A 15)].

We now proceed to the next approximation. Since $p^{(1)}$ is already determined in (3.5), we can easily find the boundary conditions for the velocity $\mathbf{u}^{(1)}$ and the temperature $\tau^{(1)}$ from (A 8), (A 11) and (A 12), i.e.

$$\left.\begin{aligned} \mathbf{u}^{(1)}.\mathbf{t} &= -5k_0\mathbf{n}.\mathbf{e}^{(0)}.\mathbf{t}, \quad \mathbf{u}^{(1)}.\mathbf{n} = -\frac{5}{C_4^*}\mathbf{n}.\mathbf{e}^{(0)}.\mathbf{n}, \\ \tau^{(1)} &= -5\frac{d_4^*}{C_4^*}\mathbf{n}.\mathbf{e}^{(0)}.\mathbf{n}, \\ k_0 &= -1\cdot016, \quad C_4^* = -2\cdot132, \quad d_4^* = -0\cdot4467, \end{aligned}\right\} \tag{3.7}$$

at $r = 1$, and also from (3.1)

$$\mathbf{u}^{(1)} \to \mathbf{e}^{(1)}.\mathbf{x}, \quad p^{(2)} \to 0, \quad \tau^{(1)} \to 0, \tag{3.8}$$

as $r \to \infty$.

It should be noted that the normal velocity in (3.7) arises solely from the pressure difference from the equilibrium pressure and is responsible for the evaporation (or sublimation) and condensation process, while the tangential velocity comes from the pure rarefaction effect regardless of whether or not the phase-change process occurs.

The solution to (2.7)–(2.9) with $m = 1$ subject to (3.7) and (3.8) is

$$\left.\begin{aligned} \mathbf{u}^{(1)} &= \frac{b_1}{r^5}\mathbf{e}^{(0)}.\mathbf{x} + \left(\frac{a_1}{2r^5} - \frac{5b_1}{2r^7}\right)\mathbf{x}.\mathbf{e}^{(0)}.\mathbf{xx} \\ &\quad + \left(1 + \frac{b_0}{r^5}\right)\mathbf{e}^{(1)}.\mathbf{x} + \left(\frac{a_0}{2r^5} - \frac{5b_0}{2r^7}\right)\mathbf{x}.\mathbf{e}^{(1)}.\mathbf{xx}, \\ p^{(2)} &= \frac{a_1}{r^5}\mathbf{x}.\mathbf{e}^{(0)}.\mathbf{x} + \frac{a_0}{r^5}\mathbf{x}.\mathbf{e}^{(1)}.\mathbf{x}, \\ \tau^{(1)} &= -5\frac{d_4^*}{C_4^*}\frac{1}{r^5}\mathbf{x}.\mathbf{e}^{(0)}.\mathbf{x} \end{aligned}\right\} \tag{3.9}$$
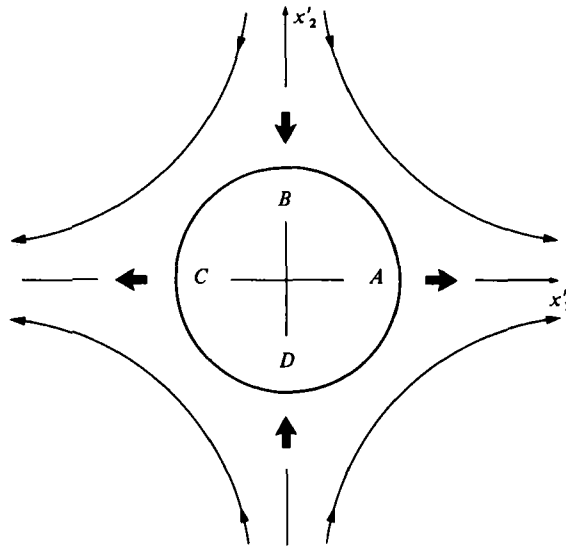
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FIGURE 1. $\mathbf{x}'$: the principal axes. The thick arrows show the direction of mass flow at the interface of the condensed phase.

and

$$a_1 = -5\left(\frac{2}{C_4^*} + 3k_0\right), \quad b_1 = -5k_0. \tag{3.10}$$

The Knudsen-layer part for the velocity, pressure and temperature can be obtained immediately from (A 9), (A 10), (A 13), (A 14) and (A 15) in appendix A, but is not written down here. We note, however, that the Knudsen-layer parts of the pressure and temperature appear due to the occurrence of the evaporation and condensation process at the interface, while the Knudsen-layer part of the tangential velocity, which is the only non-zero component, arises from the pure rarefaction effect of the gas.

The force $\mathbf{F}$ acting on the condensed phase is shown to be

$$\mathbf{F} = -\int_A P_0[\mathbf{I} + \mathbf{P}] \cdot \mathbf{n} \, dA = 0,$$

where $\mathbf{n}$ is the unit vector outward normal to the (dimensional) surface $A$ which encloses the condensed phase and lies entirely in the gas outside the Knudsen layer. The couple acting on it is also zero. These results are clear from the symmetry of the problem. It may be interesting to note that when the spherical condensed phase is placed in its own uniform gas flow $\mathbf{U}$, the force acting on it is found to be

$$\mathbf{F} = 6\pi\mu L\mathbf{U}\left[1 + \frac{\sqrt{\pi}}{2}\left(\frac{1}{2C_4^*} + k_0\right)K\right], \tag{3.11}$$

where $(\tfrac{1}{2}\sqrt{\pi})\,[(2C_4^*)^{-1} + k_0] = -1\cdot108$ (Onishi 1977$b$, where $k$ is used instead of $K$).

The total mass flow $M$ from the interface of the condensed phase is given by

$$M = \int_{A_0} \rho_0(2RT_0)^{\frac{1}{2}}\, \mathbf{u} \cdot \mathbf{n}\, dA,$$

where $\mathbf{u}$ is the total velocity (Hilbert part + Knudsen-layer part) and $A_0$ is the (dimen-

sional) area of the interface. Taking into account (3.3), (3.7), (A 3), (A 10) and the fact that $\mathbf{e}:\mathbf{l} = 0$, we easily find

$$M = \rho_0 (2RT_0)^{\frac{1}{2}} k \int_{A_0} \mathbf{u}^{(1)}.\mathbf{n}\,dA = 0,$$

which means that the total mass of the condensed phase is always conserved. This is also true for the case of the uniform streaming problem (Onishi 1977$b$). This seems to be quite natural in view of the symmetry of the problem. Since $\mathbf{e}$ is a symmetric tensor, we can find the principal values ($e_1', e_2', e_3'$) and the corresponding principal axes ($x_1', x_2', x_3'$), namely

$$\mathbf{x}.\mathbf{e}.\mathbf{x} = e_1' x_1'^2 + e_2' x_2'^2 + e_3' x_3'^2.$$

Suppose that $e_1'$ is positive and $e_3'$ is zero, then $e_2'$ must be negative and $e_2' = -e_1'$ because of the condition $\mathbf{e}:\mathbf{l} = 0$. Since $\mathbf{u}^{(1)}.\mathbf{n}$ is proportional to $\mathbf{x}.\mathbf{e}.\mathbf{x}$ (the coefficient is positive) from (3.7), $\mathbf{u}^{(1)}.\mathbf{n}$ is positive about points $A$ and $C$ and negative about points $B$ and $D$ (see figure 1). These outward fluxes about $A$ and $C$ are completely balanced by the inward fluxes about $B$ and $D$.

## 4. Bulk stress and effective viscosity of a dilute suspension in a slightly rarefied gas

Consider a suspension of randomly dispersed particles in a slightly rarefied gas. The inertia forces of the particles are assumed to be negligible and no external force and couple act on them. Following the suspension theory by Batchelor (1970) (see also Batchelor & Green 1972), we define the deviatoric part of the bulk stress $\boldsymbol{\Sigma}$ for the suspension in a slightly rarefied gas by

$$\boldsymbol{\Sigma} - \tfrac{1}{3}\mathbf{l}(\boldsymbol{\Sigma}:\mathbf{l}) = \frac{1}{V}\int_V P_0[\boldsymbol{\sigma} - \tfrac{1}{3}\mathbf{l}(\boldsymbol{\sigma}:\mathbf{l})]\,dV$$

$$= \frac{1}{V}\int_{V-\Sigma V_0} P_0[\boldsymbol{\sigma} - \tfrac{1}{3}\mathbf{l}(\boldsymbol{\sigma}:\mathbf{l})]\,dV + \frac{1}{V}\Sigma\int_{V_0} P_0[\boldsymbol{\sigma} - \tfrac{1}{3}\mathbf{l}(\boldsymbol{\sigma}:\mathbf{l})]\,dV, \quad (4.1)$$

where $P_0\,\boldsymbol{\sigma}$ is the local stress[†] and $V$ is the (dimensional) volume which contains many particles and whose linear dimension is large compared to the average distance between the particles. $V_0$ is the (dimensional) volume such that within it only one particle and the Knudsen layer around it are included; the summation extends over all such volumes in $V$. Since $V - \Sigma V_0$ occupies the whole gas region outside the Knudsen layers attached to the particles, the integrand of the first term on the right-hand side of (4.1) may be replaced by the corresponding Hilbert part, i.e. by (2.13) with (2.14), giving

$$\frac{1}{V}\int_{V-\Sigma V_0} P_0[\boldsymbol{\sigma} - \tfrac{1}{3}\mathbf{l}(\boldsymbol{\sigma}:\mathbf{l})]\,dV$$

$$= k P_0\{\langle\boldsymbol{\epsilon}_0\rangle + k\langle\boldsymbol{\epsilon}_1 - \nabla\nabla\tau^{(0)}\rangle\} + \frac{1}{V}\Sigma\int_{A_0} k P_0 L\{(\mathbf{u}^{(0)}\mathbf{n}$$

$$+ \mathbf{n}\mathbf{u}^{(0)}) + k(\mathbf{u}^{(1)}\mathbf{n} + \mathbf{n}\mathbf{u}^{(1)} - \mathbf{n}\nabla\tau^{(0)})\}\,dA, \quad (4.2)$$

where $A_0$ is the (dimensional) surface area of the volume $V_0$ which involves the Knudsen

[†] In the gas phase, this local stress is minus the stress tensor (Hilbert part + Knudsen-layer part) defined in appendix A.

layer completely, and $\mathbf{n}$ is the unit outward normal to $A_0$. The symbol $\langle \rangle$ denotes the volume average over $V$.

The second term on the right-hand side of (4.1) can be converted into the surface integral over $A_0$ using the divergence theorem to give

$$\frac{1}{V}\Sigma \int_{V_0} P_0[\boldsymbol{\sigma} - \tfrac{1}{3}\mathbf{I}(\boldsymbol{\sigma}:\mathbf{I})]\, dV = \frac{1}{V}\Sigma \int_{A_0} P_0 L[\mathbf{x}\boldsymbol{\sigma}.\mathbf{n} - \tfrac{1}{3}\mathbf{I}\mathbf{x}.\boldsymbol{\sigma}.\mathbf{n}]\, dA. \qquad (4.3)$$

The integrand of the surface integral is the value (with minus sign) of the Hilbert part of the stress evaluated on the surface $A_0$ outside the Knudsen layer. It is noted that the justification of this conversion in the present case when some components of the stress tensor have very large values within the interfacial layer between the gas and the particles can be given by the same arguments as was done by Batchelor (1970).

On combining (4.2) and (4.3), we obtain

$$\boldsymbol{\Sigma} - \tfrac{1}{3}\mathbf{I}(\boldsymbol{\Sigma}:\mathbf{I}) = kP_0\{\langle\boldsymbol{\epsilon}_0\rangle + k\langle\boldsymbol{\epsilon}_1 - \nabla\nabla\tau^{(0)}\rangle\} + \boldsymbol{\Sigma}^{(p)}. \qquad (4.4)$$

The first term on the right-hand side is the deviatoric stress that would be generated in the ambient gas in the absence of the particles. It is noted that $kP_0$ is related to the viscosity $\mu$ as $kP_0 = \mu(2RT_0)^{\frac{1}{2}} L^{-1}$. The second term $\boldsymbol{\Sigma}^{(p)}$ is the particle stress due to the presence of the particles and is given by

$$\boldsymbol{\Sigma}^{(p)} = \frac{1}{V}\Sigma\mathbf{S}, \qquad (4.5)$$

$$\mathbf{S} = \int_{A_0} L\{P_0[\mathbf{x}\boldsymbol{\sigma}.\mathbf{n} - \tfrac{1}{3}\mathbf{I}\mathbf{x}.\boldsymbol{\sigma}.\mathbf{n}] - kP_0[\mathbf{u}^{(0)}\mathbf{n} + \mathbf{n}\mathbf{u}^{(0)} $$
$$+ k(\mathbf{u}^{(1)}\mathbf{n} + \mathbf{n}\mathbf{u}^{(1)} - \mathbf{n}\nabla\tau^{(0)})]\}\, dA, \qquad (4.6)$$

where $\mathbf{S}$ is the force dipole strength or stresslet strength resulting from the replacement of ambient gas by the particle and depends on the size, shape, constitution of the particle and on the relative positions of the other particles, as well as on the bulk motion imposed on the suspension. Again note that the integrand of (4.6) is expressed by the Hilbert part of the quantities and is evaluated on $A_0$ outside the Knudsen layer.

When the suspension is so dilute that the gas motion near one particle is virtually independent of the presence of the others, and also the temperature field in the continuum limit is uniform everywhere (i.e. $\tau^{(0)} \equiv 0$), the calculation of $\mathbf{S}$ to the first order of the volume fraction $\phi$ is reduced to solving the gas motion around a typical particle immersed in a uniform pure straining motion and finding the symmetric part of the constant traceless tensor corresponding to the force doublet of the disturbance motion caused by the presence of the particles (see Batchelor 1970).

Since we are considering here a dilute suspension whose temperature is uniform in the continuum limit, it is a simple matter to find the term which corresponds to the force doublet of the disturbance motion due to a single particle in the expressions for hydrodynamic quantities, e.g. pressure. Consequently using the results in the previous section, we have for $\mathbf{S}$

$$\mathbf{S} = 4\pi L^3 kP_0[-\tfrac{1}{3}(a_0 + ka_1)\,\mathbf{e}]. \qquad (4.7)$$

The bracketed term is the contribution from the force doublet and is correct to order $k$.

Substituting (3.6) and (3.10) into (4.7) and rewriting it in terms of the Knudsen number $K$ instead of the expansion parameter $k$, we obtain

$$\mathbf{S} = \frac{4\pi}{3} L^3 2\mu \mathbf{E} \frac{5}{2} \left[ 1 + \frac{\sqrt{\pi}}{2} \left( \frac{2}{C_4^*} + 3k_0 \right) K \right],$$ (4.8)

and

$$\frac{\sqrt{\pi}}{2} \left( \frac{2}{C_4^*} + 3k_0 \right) = -3\cdot533.$$

Hence for a dilute suspension of spherical particles of condensed phase in its slightly rarefied gas, the particle stress is

$$\Sigma^{(p)} = 2\mu \mathbf{E} \left\{ \frac{5}{2} \left[ 1 + \frac{\sqrt{\pi}}{2} \left( \frac{2}{C_4^*} + 3k_0 \right) K \right] \right\} \phi$$ (4.9)

to order $\phi$, where $\phi$ is the volume fraction defined by $V\phi = \Sigma \frac{4}{3}\pi L^3$. Thus the coefficient $\beta$ in the expression (1.1) for the effective viscosity will be $\frac{5}{2}(1-3\cdot533\,K)$. When the effect of rarefaction alone enters the problem, the particle stress to order $\phi$ is given by

$$\Sigma^{(p)} = 2\mu \mathbf{E} \{ \frac{5}{2}[1 + \frac{3}{2}\sqrt{\pi}\,k_0 K] \} \phi,$$ (4.10)

where $\frac{3}{2}\sqrt{\pi}\,k_0 = -2\cdot702$.

Finally, it may be interesting to compare (4.9) with (3.11) where the apparent viscosity of the gas would be considered to be $\mu(1-1\cdot108\,K)$.

## Appendix A

(1) The general slip boundary condition for hydrodynamic quantities at the interface between the condensed phase and its gas phase, and the Knudsen-layer part which is the correction to the Hilbert part near the interface:

$$(\mathbf{u}_H^{(0)} - \mathbf{u}_w^{(0)}).\mathbf{t} = 0,$$ (A 1)

$$\mathbf{u}_K^{(0)}.\mathbf{t} = 0, \quad \mathbf{u}_K^{(0)}.\mathbf{n} = 0,$$ (A 2), (A 3)

$$\begin{bmatrix} p_H^{(0)} - p_w^{(0)} \\ \tau_H^{(0)} - \tau_w^{(0)} \\ \rho_K^{(0)} \\ \tau_K^{(0)} \end{bmatrix} = \mathbf{u}_H^{(0)}.\mathbf{n} \begin{bmatrix} C_4^* \\ d_4^* \\ \Omega_4^*(\eta) \\ \Theta_4^*(\eta) \end{bmatrix},$$
(A 4)
(A 5)
(A 6)
(A 7)

$$\begin{bmatrix} (\mathbf{u}_H^{(1)} - \mathbf{u}_w^{(1)}).\mathbf{t} \\ \mathbf{u}_K^{(1)}.\mathbf{t} \end{bmatrix} = -\mathbf{n}.\boldsymbol{\epsilon}_0.\mathbf{t} \begin{bmatrix} k_0 \\ Y_0 \end{bmatrix} - \mathbf{t}.\nabla\tau_H^{(0)} \begin{bmatrix} K_1 \\ \frac{1}{2}Y_1 \end{bmatrix}$$

$$+ \mathbf{t}.\nabla(\mathbf{u}_H^{(0)}.\mathbf{n}) \begin{bmatrix} K_2 \\ 2Y_0 + \frac{1}{2}d_4^* Y_1 \end{bmatrix},$$
(A 8)
(A 9)

$$\mathbf{u}_K^{(1)} \cdot \mathbf{n} = 0, \tag{A 10}$$

$$\begin{bmatrix} p_H^{(1)} - p_w^{(1)} \\ \tau_H^{(1)} - \tau_w^{(1)} \\ \rho_K^{(1)} \\ \tau_K^{(1)} \end{bmatrix} = \mathbf{u}_H^{(1)} \cdot \mathbf{n} \begin{bmatrix} C_4^* \\ d_4^* \\ \Omega_4^* \\ \Theta_4^* \end{bmatrix} + \mathbf{n} \cdot \nabla \tau_H^{(0)} \begin{bmatrix} C_1 \\ d_1 \\ \Omega_1 \\ \Theta_1 \end{bmatrix}$$

$$+ \mathbf{n} \cdot \boldsymbol{\epsilon}_0 \cdot \mathbf{n} \begin{bmatrix} C_6 \\ d_6 \\ \Omega_6 \\ \Theta_6 \end{bmatrix} - 2\kappa \mathbf{u}_H^{(0)} \cdot \mathbf{n} \begin{bmatrix} C_7 \\ d_7 \\ \Omega_7 \\ \Theta_7 \end{bmatrix}, \qquad \begin{matrix} \text{(A 11)} \\ \text{(A 12)} \\ \text{(A 13)} \\ \text{(A 14)} \end{matrix}$$

$$p_K^{(m)} = \rho_K^{(m)} + \tau_K^{(m)} \quad (m = 0, 1), \tag{A 15}$$

$$C_4^* = -2 \cdot 132\,039, \quad d_4^* = -0 \cdot 446\,749,$$

$$C_1 = 0 \cdot 558\,437, \quad d_1 = 1 \cdot 302\,716,$$

$$C_6 = 0 \cdot 820\,853, \quad d_6 = 0 \cdot 330\,345,$$

$$C_7 = -0 \cdot 380\,569, \quad d_7 = -0 \cdot 131\,574,$$

$$k_0 = -1 \cdot 016\,191, \quad K_1 = -0 \cdot 383\,161,$$

$$K_2 = -0 \cdot 795\,186,$$

where $\boldsymbol{\epsilon}_m$ $(m = 0, 1)$ is the rate-of-strain tensor defined by

$$\boldsymbol{\epsilon}_m(\mathbf{x}) = \nabla \mathbf{u}_H^{(m)} + (\nabla \mathbf{u}_H^{(m)})^T \tag{A 16}$$

but here it is evaluated at the interface. $(2RT_0)^{\frac{1}{2}} \mathbf{u}_w$, and $T_0(1 + \tau_w)$ are the velocity (with $\mathbf{u}_w \cdot \mathbf{n} = 0$) and the temperature of the condensed phase, respectively, and $P_0(1 + p_w)$ is the saturated gas pressure corresponding to $T_0(1 + \tau_w)$ (the effect of surface tension on this pressure is considered in appendix B and found to be small) and is uniquely related to the temperature (see Onishi 1977a), but the explicit functional form is not necessary here. These parameters $\mathbf{u}_w$, $\tau_w$ and $p_w$ are also expanded in terms of $k$ as in (2.2). $\mathbf{n}$ and $\mathbf{t}$ are the unit outward normal vector and any tangential unit vector to the interface, respectively, and $2\kappa = (\kappa_1 + \kappa_2)$, $L^{-1}\kappa_1$ and $L^{-1}\kappa_2$ being the principal curvatures taken negative when the corresponding centre of curvature lies in the gas phase. The functions $\Omega_4^*, \Theta_4^*, \Omega_1, \Theta_1, \Omega_6, \Theta_6, \Omega_7, \Theta_7, Y_0$, and $Y_1$ are the universal functions of $\eta$ only, a stretched co-ordinate normal to the interface defined by the relation

$$\mathbf{x} = \mathbf{n}k\eta + \mathbf{x}_w(\zeta_1, \zeta_2),$$

where $\mathbf{x}_w$ is the equation of the interface, and $\zeta_1$ and $\zeta_2$ are (unstretched) co-ordinates within a parallel surface $\eta = $ constant. These functions decay rapidly (more rapidly than $\eta^{-N}$ where $N$ is any positive integer) as $\eta \to \infty$. The numerical values of these functions and some of their integrals are given in the original paper (Sone & Onishi 1978).

(2) The stress tensor and heat-flux vector of the gas and their Knudsen-layer parts:

$$\begin{aligned} \mathbf{P}_H^{(0)} &= p_H^{(0)} \mathbf{I}, \quad \mathbf{P}_H^{(1)} = p_H^{(1)} \mathbf{I} - \boldsymbol{\epsilon}_0, \\ \mathbf{P}_H^{(2)} &= p_H^{(2)} \mathbf{I} - \boldsymbol{\epsilon}_1 + \nabla \nabla \tau_H^{(0)}, \end{aligned} \right\} \tag{A 17}$$

$$n.\mathbf{P}_K^{(0)}.n = 0, \quad n.\mathbf{P}_K^{(0)}.t = 0, \\ n.\mathbf{P}_K^{(1)}.n = -3\kappa u_H^{(0)}.n\Psi(\eta), \\ n.\mathbf{P}_K^{(1)}.t = \tfrac{3}{2}t.\nabla(u_H^{(0)}.n)\Psi(\eta),$$

(A 18)

$$Q_H^{(0)} = 0, \quad Q_H^{(1)} = -\tfrac{5}{4}\nabla\tau_H^{(0)}, \\ Q_H^{(2)} = -\tfrac{5}{4}\nabla\tau_H^{(1)} + \tfrac{1}{2}\nabla^2 u_H^{(0)},$$

(A 19)

$$Q_K^{(0)}.n = 0, \quad Q_K^{(1)}.n = 0,$$

(A 20)

and

$$\Psi(\eta) = \int_\eta^\infty [\Omega_4^* + \Theta_4^*] \, d\eta', \quad \Psi(0) = 0.23886,$$

where $P_0(\mathbf{I}+\mathbf{P})$ and $P_0(2RT_0)^{\frac{1}{2}}\mathbf{Q}$ are the stress tensor and heat-flux vector of the gas, respectively, and $\mathbf{I}$ is the dyadic idemfactor. The viscosity $\mu$ of the gas is related to the Knudsen number through (2.4) and (2.5), and the thermal conductivity $\lambda$ is equal to $\tfrac{5}{2}R\mu$ in the Boltzmann–Krook–Welander equation (see Vincenti & Kruger 1965, cha. 10). Note that the conventional stress tensor is minus the stress tensor defined here.

## Appendix B

We consider the effect of surface tension on the saturated gas pressure $P_0(1+p_w)$ introduced in appendix A. Let $p_0$ be the saturated gas pressure corresponding to temperature $T$ when the interface between the condensed phase and its gas is a plane. When the interface is curved because of the surface tension, the equilibrium (or saturated) pressure corresponding to the same temperature $T$ will change to $p_1$ in the gas phase (subscript 1) and to $p_2$ in the condensed phase (subscript 2), but since the phases are in equilibrium with each other, the specific Gibbs-free energy $G$ must be the same in both phases, namely

$$G_1(p_1, T) = G_2(p_2, T),$$

(B 1)

with

$$p_2 - p_1 = \alpha \left(\frac{1}{r} + \frac{1}{r'}\right),$$

(B 2)

where $\alpha$ is the surface tension, and $r$ and $r'$ are the principal radii of curvature of the condensed phase. Expanding both functions around $p_0$ and noting that

$$(\partial G/\partial p)_T = V,$$

where $V$ is the specific volume, we obtain

$$p_1 - p_0 = \alpha \left(\frac{1}{r} + \frac{1}{r'}\right) \frac{V_2}{V_1 - V_2}.$$

(B 3)

Here we have assumed that $p_1 - p_0$ and $p_2 - p_0$ are small. This is a valid assumption owing to the smallness of the surface effects (Landau & Lifshitz 1958, § 141). This $p_1$ corresponds to $P_0(1+p_w)$ in appendix A, but when the temperature is well below the critical temperature, the specific volume $V_1$ of the gas is much larger than that of the condensed phase ($V_2$), and the difference $p_1 - p_0$ will become very small. In the case of $H_2O$ for example, $(p_1 - p_0)/p_0 \sim 0.6 \times 10^{-4}$ for $T = 10\ °C$ and $r = r' = 20\ \mu m$,

and $(p_1 - p_0)/p_0 \sim 10^{-4}$ for $T = 20\,°C$ and $r = r' = 10\,\mu m$. In view of this, we may take the saturated gas pressure over the plane interface, which is given by the Clausius–Clapeyron relation, as $P_0(1 + p_w)$ in most cases (Sone & Onishi 1978).

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