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We derive averaged equations for large Reynolds number laminar flows of gas–liquid dispersions accounting for slowly varying spatial and temporal fields. In particular, we obtain an exact expression for the dispersed-phase stress tensor to be used in the force balance equation for gas bubbles and illustrate its application by evaluating the stress tensor for a few special cases. It is shown that the dispersed-phase stress tensor gradient with respect to the mean relative motion or the void fraction for the uniformly random bubbly liquids under conditions of large Reynolds number laminar flows is negative and thus has a destabilizing influence on the dynamics of void fraction waves in bubbly liquids.

1. Introduction

We consider the problem of deriving averaged equations for flows of liquids containing spherical gas bubbles when the Reynolds number based on the radius and the characteristic velocity of the bubbles is large compared to unity. This problem has been examined previously by a number of investigators (see, for example, Ishii 1975; Nigmatulin 1979; Prosperetti & Jones 1984; Biesheuvel & van Wijngaarden 1984; Geurst 1986; and van Wijngaarden & Kapteyn 1990). Biesheuvel & van Wijngaarden (1984) used a combination of volume and ensemble averaging techniques to derive the averaged equations for the mixture. This results in averaged quantities such as the Reynolds stress and the overall stress in the medium which depend on the magnitude of the relative motion between the two phases. To calculate this quantity, Biesheuvel & van Wijngaarden proposed that an additional relation derived from the force balance on the dispersed phase must be used. By examining the nature of forces that act on a single bubble under unsteady flow conditions, they proposed such a relation, and this, together with the averaged equations for the overall medium, then forms the complete set of equations for analysing various macroscopic flows. Since this relation for the force balance on a single bubble was derived from a volume averaging procedure, it has the right form as far as the various unsteady terms are concerned. However, as pointed out by these investigators, it lacks the terms that are important when there are spatial variations in the velocity and volume fractions of the individual phases.

Equations of motion taking account of spatial variations in volume fraction and velocity have been developed in the theory of rapid granular flows (Jenkins & Richman 1985) and gas–solid suspensions under conditions of small Reynolds and large Stokes numbers (Koch 1990). These theories contain particle or dispersed-phase

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stress terms which describe the transport of momentum by the random translation of the solid particles and by solid-body collisions. Batchelor (1988) noted that, in addition, there should be a stress contribution from fluid dynamic interparticle forces in a fluidized bed. However, he did not specify the means of calculating this stress from a detailed study of the particle interactions.

For gas–liquid dispersions, van Wijngaarden & Kapteyn (1990) arrived at a term in the force balance on a test bubble which can be interpreted as the divergence of the dispersed-phase stress tensor. These investigators analysed explicitly the case of dilute dispersion only, and the stress tensor they derived is related to the average of the fluctuations in the impulse multiplied by the fluctuations in the velocity of the bubbles. The impulse associated with a bubble is defined through (26) and it plays a role similar to the momentum of a particle in gas–solid suspensions. Thus, the stress tensor that these investigators have derived accounts for the translational stress but not the fluid dynamic interactions and collisions, if any. Biesheuvel & Gorissen (1990) used an approach similar to that used in the statistical mechanics theory of pressure in dense gases or liquids (Rice & Gray 1965) to obtain a formal expression for the stress tensor for bubbly liquids. Their expression accounts for both the translational and the fluid dynamic interparticle interactions. They assumed that the interparticle interactions can be expressed as a sum of pair interactions but did not show how the interactions among many bubbles can be decomposed into pair interactions. They also did not carry out any specific calculations for estimating the contribution due to these fluid dynamic interactions.

With the development of efficient numerical methods for solving rigorously the problem of multiparticle interactions, it has become possible to carry out detailed dynamic simulations of the motion of many bubbles under conditions of large Reynolds number laminar flow, as can be seen, for example, in a companion study, Sangani & Didwania (1993, to be referred to herein as I). With the help of these simulations, it is possible to compute various dynamic quantities, and thus it is important to establish the precise definitions of various average quantities that must be evaluated via such simulations. While both Batchelor's and van Wijngaarden & Kapteyn's studies give a good physical picture of the origin of the term associated with the interparticle stress, they do not give the recipes for determining them from the detailed knowledge of the motion of particle or bubbles. The aim of the present investigation therefore is to derive the averaged equations in a systematic manner and to give formulae for determining various averaged quantities from the details of the bubble motion in non-dilute dispersions, i.e. dispersions in which the volume fraction β of the dispersed phase is not small. The main emphasis is on determining the correct expression for the interparticle or the dispersed-phase stress tensor. This quantity is believed to play an important role in stabilizing small-amplitude disturbances of β in bubbly liquids and fluidized beds (see, for example, van Wijngaarden & Kapteyn 1990; Biesheuvel & Gorissen 1990; Batchelor 1988).

To illustrate the application of this expression for the stress tensor, we consider three special cases. In the first case, the centres of the bubbles coincide with the lattice of a periodic array. For this situation, it is possible to derive an analytical expression that is valid when β is small. In order that the waves in β in the direction of the mean relative flow are stabilized by the dispersed-phase stress tensor, the gradient of this quantity in the direction of the mean flow must be positive. This condition is satisfied when the bubbles are arranged on a simple cubic lattice but not when the bubbles are arranged on the body-centred cubic lattice. Thus the latter arrangement of bubbles is destabilized by the dispersed phase. The dispersed-phase

stress plays a stabilizing role in the case of a simple cubic arrangement provided that the disturbance in β is produced by changing only the spacing between the bubbles in the direction of gravity. We should emphasize here that the actual criterion for the stability of bubbly liquids will also depend on the relative magnitude of the other terms in the averaged equations. Our goal in the present study is not to carry out the complete stability analysis. Rather, we are only interested in determining under what circumstances the dispersed-phase stress will play a stabilizing (or destabilizing) role.

The second case we consider is that of a random arrangement of bubbles rising under the influence of buoyancy forces. The velocity of all bubbles is taken to be the same at time $t = 0$. We find that the gradient of the stress is negative in this case also, indicating its destabilizing influence. Indeed, as shown in I via dynamic simulations, the uniform random state of a bubbly liquid is unstable under these conditions. The magnitude of the fluid dynamic interparticle interaction term is greater than the combined contribution of the translational and collisional components of the stress. van Wijngaarden & Kapteyn (1990) determined the dispersed-phase stress for dilute bubbly liquids by accounting only for the kinetic contribution, and, therefore, the stress they calculated played the stabilizing role. Thus, what we find is that accounting for fluid dynamic interactions actually *reverses* the role played by the dispersed-phase stress in the stability of void fraction waves. We should add here that Biesheuvel & Gorissen (1990) had actually suggested that the potential contribution to the stress is likely to be negative even though they did not actually account for it in their numerical analysis of the stability of the void fraction waves.

The third case we consider is that of an initially random arrangement of bubbles with large fluctuations in their velocities. We take the magnitudes of the relative motion, gravity, and viscosity all to be vanishingly small. This situation may be applicable to flows of bubbly liquids under turbulent flow conditions or to laminar flow of bubbly liquids undergoing shear, as in the case of flows inside pipes. The latter may be thought of as an analogous situation to rapid granular flow of slightly inelastic particles for which it is known that the leading-order velocity distribution is isotropic Maxwellian with the mean shear causing a small perturbation. In this case, the dispersed-phase stress is a function of the Reynolds stress and β, and we determine this dependence by carrying out dynamic simulations. The magnitude of the translational and collisional contributions is much larger than that of the fluid dynamic interaction and, as a consequence, the dispersed-phase stress has a positive gradient. Thus, we see that if there is a mechanism for inducing large variance in the velocity, e.g. by turbulence or shear, the state of uniform bubbly liquids can be made stable.

In addition to the continuity and the force balance condition, the theory of rapid granular flows also includes an energy equation for the dispersed phase. For complete analysis of the flows of bubbly liquids at large Reynolds number, we also require such an energy equation for the dispersed phase. Analogous to the dispersed-phase stress term in the momentum balance, this equation contains an energy flux term. We have been unable to derive an exact expression for this term, and therefore, an approximate expression is given for it. The accuracy of this expression is tested with the results obtained from numerical simulations. The results for this quantity for the special cases described above are also presented.

2. Ensemble-averaged equations for the overall medium

We consider an infinite medium consisting of gas bubbles dispersed in a liquid. The Reynolds number of the flow based on the characteristic velocity and size of the gas bubbles is large compared to unity. The interfacial tension is also large enough to maintain the bubbles' approximate spherical shape. We wish to derive the averaged equations when spatial gradients of various macroscopic quantities, such as the average velocity or the volume fraction of bubbles, are small, and hence it will suffice to obtain averaged equations that are valid to first order in the spatial derivatives on a macroscopic lengthscale. We also wish to derive the averaged equations in a form such that all the quantities that are needed in the analysis of the flows that are slowly varying in time and space can be estimated to the leading order from dynamic simulations of homogeneous flows, as they are the simplest kind of flow to simulate. By homogeneous flows, we mean flows in which there are no macroscopic variations in velocity or volume fraction. Since the density of most gases is negligibly small compared to that of liquids, we treat the bubbles as massless. Finally, we also restrict ourselves to the case when both phases may be regarded as essentially incompressible. For acoustic applications, of course, the compressibility of the phases are important, and therefore the equations to be derived here cannot be used for such applications. The averaged equations for such applications have been derived by a number of investigators including the most recent work by Sangani (1991). In that study, the focus was mainly on problems in which the temporal variations in the averaged quantities were of prime importance, whereas in the present study, the interest is in correct modelling of the spatial variations.

The equations of motion for an incompressible liquid are

$$\nabla \cdot \boldsymbol{u}^{\mathrm{L}} = 0, \tag{1}$$

$$\rho \left[\frac{\partial \boldsymbol{u}^{\mathrm{L}}}{\partial t} + \nabla \cdot (\boldsymbol{u}^{\mathrm{L}} \boldsymbol{u}^{\mathrm{L}}) \right] = -\nabla p^{\mathrm{L}} + \rho \boldsymbol{g} + \nabla \cdot \boldsymbol{\tau}_{\mathrm{v}}^{\mathrm{L}}, \tag{2}$$

where ρ is the density of the liquid, $\boldsymbol{u}^{\mathrm{L}}$ and p^{L} are the velocity and pressure fields for the liquid phase, $\boldsymbol{g}$ is the gravitational acceleration, and $\boldsymbol{\tau}_{\mathrm{v}}^{\mathrm{L}}$ is the viscous stress tensor defined by

$$\boldsymbol{\tau}_{\mathrm{v}}^{\mathrm{L}} = \mu[\nabla \boldsymbol{u}^{\mathrm{L}} + (\nabla \boldsymbol{u}^{\mathrm{L}})^{+}], \tag{3}$$

μ being the viscosity of the liquid.

The density and viscosity of the gas phase are taken to be vanishingly small and therefore the momentum equation for the gas phase is simply

$$\nabla p^{\mathrm{G}} = 0, \tag{4}$$

which implies that pressure inside any bubble is uniform.

To obtain the ensemble-averaged equations for the overall medium, we follow the approach taken in Sangani (1991) and introduce an indicator function χ defined to be unity for a point $\boldsymbol{x}$ lying in the liquid phase and zero for the gas phase. Since there is no mass transfer at the gas–liquid interface, χ is convected along with the fluid, and therefore we have

$$\frac{\partial \chi}{\partial t} + \boldsymbol{u} \cdot \nabla \chi = 0 \tag{5}$$

at all points in the medium. It may be noted that χ has a jump discontinuity at the gas–liquid interface, and therefore $\nabla \chi$ equals a delta function situated at the

interface multiplied by a unit normal vector at the interface pointing into the liquid phase. Similarly, $\partial\chi/\partial t$ is also singular at the interface and can be determined from $\nabla\chi$ and (5).

The ensemble-averaged equations for the overall medium can now be determined by multiplying the equations of motion for the liquid phase by χ and those for the gas phase by $1-\chi$, adding them, and taking ensemble averages of the resulting equations. Thus, since both phases have been assumed to be incompressible, the continuity equation for the overall medium is given by

$$\langle\chi\nabla\cdot\boldsymbol{u}^L+(1-\chi)\,\nabla\cdot\boldsymbol{u}^G\rangle = 0, \tag{6}$$

where the angular brackets denote the operation of ensemble averaging. Taking the divergence outside the averaging procedure produces

$$\nabla\cdot\langle\chi\boldsymbol{u}^L+(1-\chi)\,\boldsymbol{u}^G\rangle+\langle\nabla\chi\cdot(\boldsymbol{u}^G-\boldsymbol{u}^L)\rangle = 0. \tag{7}$$

The second term on the left-hand side of the above equation vanishes identically on account of the kinematic condition at the interface, and thus we obtain

$$\nabla\cdot\boldsymbol{U} = 0 \tag{8}$$

with the ensemble-averaged velocity $\boldsymbol{U}$ of the mixture given by

$$\boldsymbol{U} = \langle\chi\boldsymbol{u}^L+(1-\chi)\,\boldsymbol{u}^G\rangle. \tag{9}$$

The result (8) is rather obvious since we expect the overall medium to be incompressible when both of the constituent phases are incompressible.

We now proceed in a similar manner to derive the averaged momentum equation for the overall medium. Combining (2) and (4) we obtain

$$\rho\left[\frac{\partial}{\partial t}\langle\chi\boldsymbol{u}^L\rangle+\nabla\cdot\langle\chi\boldsymbol{u}^L\boldsymbol{u}^L\rangle-\left\langle\left(\frac{\partial\chi}{\partial t}+\nabla\chi\cdot\boldsymbol{u}^L\right)\boldsymbol{u}^L\right\rangle\right]$$
$$= -\nabla P+(1-\beta)\,\boldsymbol{g}+\nabla\cdot\langle\chi\boldsymbol{\tau}_v^L\rangle+\langle\nabla\chi\cdot(\boldsymbol{\sigma}^G-\boldsymbol{\sigma}^L)\rangle, \tag{10}$$

where $\boldsymbol{\sigma}$ is the stress tensor, i.e. $\sigma_{ij}\equiv-p\delta_{ij}+\tau_{v_{ij}}$. The third term on the left-hand side of (10) vanishes identically due to (5), whereas the last term on the right-hand side can be expressed as

$$\left\langle\frac{\partial\chi}{\partial x_j}(\sigma_{ij}^G-\sigma_{ij}^L)\right\rangle = \int_{|\boldsymbol{x}-\boldsymbol{x}_1|=a}\langle n_j(\sigma_{ij}^G-\sigma_{ij}^L)\rangle_1\,(\boldsymbol{x}\,|\,\boldsymbol{x}_1)\,P_1(\boldsymbol{x}_1)\,\mathrm{d}A_1, \tag{11}$$

where $P_1(\boldsymbol{x}_1)$ is the probability of finding a bubble at $\boldsymbol{x}_1$ such that point $\boldsymbol{x}$ lies on the surface of this bubble, and $\langle\;\rangle_1(\boldsymbol{x}\,|\,\boldsymbol{x}_1)$ denotes the conditional average of a quantity evaluated at $\boldsymbol{x}$ given a bubble at $\boldsymbol{x}_1$. Here we have assumed, for simplicity, that the bubbles are monodisperse with radius a. The above integral involves evaluating the integrand for different bubbles whose centres lie on the surface $|\boldsymbol{x}-\boldsymbol{x}_1| = a$. It is convenient instead to compute integrals on the surface of a single bubble located at, say, $\boldsymbol{x}$. To accomplish this, we note that a conditional-averaged quantity such as $\langle f\rangle_1(\boldsymbol{x}\,|\,\boldsymbol{x}_1)$ may be treated as a function of the position of a point relative to the centre of the test bubble, i.e. $\boldsymbol{x}-\boldsymbol{x}_1$, and the centre of the bubble, i.e. $\boldsymbol{x}_1$. The variation of $\langle f\rangle_1(\boldsymbol{x}-\boldsymbol{x}_1,\boldsymbol{x}_1)$ with respect to the first argument is rapid while that with the second is slow. Thus, we first write

$$\int_{|\boldsymbol{x}-\boldsymbol{x}_1|=a}\langle(\sigma_{ij}^G-\sigma_{ij}^L)\,n_j\rangle_1\,(\boldsymbol{x}-\boldsymbol{x}_1,\boldsymbol{x}_1)\,P_1(\boldsymbol{x}_1)\,\mathrm{d}A_1$$

$$= \int_{s=a}\langle(\sigma_{ij}^G-\sigma_{ij}^L)\,n_j\rangle_1\,(\boldsymbol{s},\boldsymbol{x}-\boldsymbol{s})\,P_1(\boldsymbol{x}-\boldsymbol{s})\,\mathrm{d}A_s, \tag{12}$$

in which $s = x - x_1$ and s is the magnitude of the vector s. Next, when the average quantities vary slowly in space, the use of a Taylor series expansion of the integrand in the above equation near x yields

$$\int_{s=a} \langle (\sigma_{ij}^{\mathrm{G}} - \sigma_{ij}^{\mathrm{L}}) n_j \rangle_1 (s, x-s) P_1(x-s) \, \mathrm{d}A_s = P_1(x) \int_{s=a} \langle (\sigma_{ij}^{\mathrm{G}} - \sigma_{ij}^{\mathrm{L}}) n_j \rangle_1 (s, x) \, \mathrm{d}A_s$$

$$- \frac{\partial}{\partial x_j} \left\{ P_1(x) \int_{s=a} \langle (\sigma_{ik}^{\mathrm{G}} - \sigma_{ik}^{\mathrm{L}}) n_k s_j \rangle_1 (s, x) \, \mathrm{d}A_s \right\} + \dots . \quad (13)$$

Now, since $\sigma_{ij}^{\mathrm{G}} = -p^{\mathrm{G}} \delta_{ij}$, and since the pressure inside any gas bubble is uniform, the integral of $\sigma_{ij}^{\mathrm{G}} n_j$ in the first term on the right-hand side of (13) vanishes. The remainder in that term integrates to the total force exerted by the liquid on the bubble at x, and this also vanishes as the bubbles are massless. Substituting (13) for the last term in (10), and rearranging, we obtain the averaged momentum equation for the overall medium as

$$\rho \left[\frac{\partial}{\partial t} (1-\beta) U_i^{\mathrm{L}} + \frac{\partial}{\partial x_j} (1-\beta) U_i^{\mathrm{L}} U_j^{\mathrm{L}} \right] = -\frac{\partial P}{\partial x_i} + \rho(1-\beta) g_i - \frac{\partial}{\partial x_j} \beta \Sigma_{ij}, \quad (14)$$

where U_i^{L} is the average velocity of the liquid phase defined by

$$(1-\beta) U_i^{\mathrm{L}} = \langle \chi u_i^{\mathrm{L}} \rangle, \quad (15)$$

β is the volume fraction of the gas bubbles, i.e.

$$\beta = \langle 1 - \chi \rangle, \quad (16)$$

and Σ_{ij} is the stress tensor for the overall medium and equals

$$\beta \Sigma_{ij} \equiv \beta \Sigma_{ij}^{\mathrm{R}} + \beta \Sigma_{ij}^{\mathrm{S}} = \langle \chi \rho (u_i^{\mathrm{L}} - U_i^{\mathrm{L}}) (u_j^{\mathrm{L}} - U_j^{\mathrm{L}}) \rangle$$

$$+ \left[\frac{\beta}{v_{\mathrm{b}}} \int_{s=a} \langle ((p^{\mathrm{L}} - p^{\mathrm{G}}) \delta_{ik} - \tau_{\mathrm{v}ik}^{\mathrm{L}}) n_k s_j \rangle_1 \, \mathrm{d}A_s - \langle \chi \tau_{\mathrm{v}ij}^{\mathrm{L}} \rangle \right], \quad (17)$$

where we have made use of the relation $P_1(x) = \beta(x)/v_{\mathrm{b}}, v_{\mathrm{b}} = \frac{4}{3}\pi a^3$ being the volume of the bubble. The averaged momentum equation (14) is the same as that derived by Biesheuvel & van Wijngaarden (1984) who considered the slightly more general case of compressible bubbles and used a somewhat different procedure for deriving these equations.

It may be noted that the expression (17) for the overall stress is actually valid for all Reynolds numbers. The first term on the right-hand side of (17) corresponds to the Reynolds stress, while the deviatoric part of the second term is related to the stresslet that is required in the evaluation of the effective viscosity of suspensions under low Reynolds number conditions. In the case of large Reynolds number flows, we may neglect $\tau_{\mathrm{v}}^{\mathrm{L}}$ in the above expression, and thus this part of the overall stress is related to a particular distribution of the inviscid pressure at the interface. Thus, we write

$$\beta \Sigma_{ij}^{\mathrm{S}} = \frac{\beta}{4\pi a^2} \left[\int_{s=a} \langle p^{\mathrm{L}} (3 n_i n_j - \delta_{ij}) \rangle_1 \, \mathrm{d}A_s + \delta_{ij} \int_{s=a} \langle p^{\mathrm{L}} - p^{\mathrm{G}} \rangle_1 \, \mathrm{d}A_s \right]. \quad (18)$$

Here we have made use of the fact that the pressure inside the bubble is uniform and therefore the surface integration of $p^{\mathrm{G}}(3 n_i n_j - \delta_{ij})$ vanishes. Now, the second term on the right-hand side of (18) can be shown to equal $8\pi a T \delta_{ij}$ plus a term which is of $O(\rho a T V^2 / P_{\mathrm{a}})$. Here, T is the interfacial tension, V is the characteristic velocity of the

bubbles, and P_a is the pressure in the liquid in the absence of flow. The correction term arises because of a small change in the radius of the bubbles due to variations in the dynamic pressure resulting from the flow, and its magnitude is negligible in most practical applications.

In the limit of small β, if the interactions among the bubbles may be neglected, the above quantities can be readily related to the velocity of the bubbles relative to the liquid (or the mixture) by

$$\Sigma_{ij}^{R} = \frac{\rho}{20} V_i V_j + \frac{3\rho}{20} V^2 \delta_{ij}, \tag{19}$$

$$\Sigma_{ij}^{S} = \frac{2T}{a} \delta_{ij} + \frac{9\rho}{20} V_i V_j - \frac{3\rho}{20} V^2 \delta_{ij}, \tag{20}$$

which are in agreement with the expressions derived by Biesheuvel & van Wijngaarden (1984). Here $V_i = U_i^{G} - U_i$ is the velocity of the gas phase relative to the mixture. It is important to note that the dynamic part of the stresslet term represented by the last two terms in (20) has a zero trace, and thus it contributes only to the deviatoric stress of the overall medium. In situations where the gradient of β is not zero, the most significant contribution to the overall stress arises from the surface tension term since the dynamic quantities are $O(We)$ compared to it, $We = \rho a V^2/T$ being the Weber number.

The continuity and momentum equations for the overall medium (cf. (8) and (14)) together constitute a total of four scalar relations among a total of eight variables: U, U^{L}, P, and β, and therefore, it is clear that more relations are needed for a unique determination of these quantities.

As mentioned in the Introduction, Biesheuvel & van Wijngaarden (1984) proposed using an additional relation based on the force balance on a single bubble to close the system of equations. This relation is

$$\langle F \rangle = 0 = \rho v_{b} \left[\frac{\partial U}{\partial t} - \frac{1}{2} \frac{\partial V}{\partial t} \right] - 12\pi\mu a V - \rho v_{b} g, \tag{21}$$

where $\frac{1}{2}\rho v_{b}$ is the virtual or the added mass associated with the relative motion of a massless bubble in the mixture. At steady state, the magnitude of the relative motion is determined from the balance of gravitational and viscous forces, and an acceleration in the mean flow causes this relative motion to change as given by (21) whenever interactions among the bubbles and spatial derivatives of β or mean flow are negligible.

Our aim, as mentioned in the Introduction, is to systematically derive the terms that are necessary to account for spatial variations, and thereby to obtain expressions for various averaged quantities that should be evaluated from dynamic simulations. This requires deriving another set of averaged equations corresponding to the relative motion of the dispersed phase.

3. Averaged equations for the dispersed phase

The continuity equation for the dispersed phase can be obtained simply by taking the ensemble average of the continuity equation for the gas phase multiplied by its indicator function $1 - \chi$. This yields

$$\langle (1-\chi) \nabla \cdot u^{G} \rangle = 0 \quad \text{or} \quad \nabla \cdot (\beta U^{G}) + \langle \nabla\chi \cdot u^{G} \rangle = 0. \tag{22}$$

The second term on the left-hand side of (22) equals $\langle -\partial \chi / \partial t \rangle$ (cf. (5)), and thus, we obtain

$$\nabla \cdot (\beta U^{\mathrm{G}}) + \frac{\partial \beta}{\partial t} = 0. \tag{23}$$

The main difficulty is in deriving the momentum equation for the dispersed phase, as this yields

$$\langle (1 - \chi) \, \nabla p^{\mathrm{G}} \rangle = 0, \tag{24}$$

from which no further progress can be made to obtain a relation for the force acting on the dispersed phase in terms of various different physical phenomena, such as the added mass or the viscous effects, as was done, for example, in writing down (21). Therefore, we need to follow a different approach to derive the averaged momentum equations for the dispersed phase.

At large Reynolds numbers, the force balance on a massless bubble α in the midst of a dispersion containing many other bubbles, yields (cf. I)

$$\frac{\mathrm{d}I_i^\alpha}{\mathrm{d}t} = \rho \int_{S^\alpha} (\tfrac{1}{2} u_j^{\mathrm{L}} u_j^{\mathrm{L}} - u_j^{\mathrm{L}} v_j^\alpha) \, n_i \, \mathrm{d}A + F_{i,\mathrm{v}}^\alpha + F_{i,\mathrm{g}}^\alpha, \tag{25}$$

where $F_{i,\mathrm{v}}^\alpha$ and $F_{i,\mathrm{g}}^\alpha$ are, respectively, the viscous and gravitational forces on the bubble α, v^α is the velocity of the bubble, S^α is the surface enclosing the bubble, and I_i^α is the impulse associated with the bubble as defined by

$$I_i^\alpha = -\rho \int_{S^\alpha} \phi n_i \, \mathrm{d}A, \tag{26}$$

with ϕ being the velocity potential. As discussed in more detail in I, the velocity field can be reasonably well approximated in the large Reynolds number limit by the inviscid, irrotational flow approximation, for which it is possible to write $u^{\mathrm{L}} = \nabla \phi$. The integral in (25) is thus to be evaluated from the potential flow approximation. The viscous force evaluation is also discussed at length in I. Finally, the time derivative in (25) is to be evaluated following the motion of the bubble α.

For a single bubble moving with velocity U^{G} in a liquid, whose velocity at infinity is U, it is easy to show that the instantaneous velocity potential is given by

$$\phi = U \cdot x + \frac{a^3}{2r^3} (U - U^{\mathrm{G}}) \cdot x, \tag{27}$$

in which r and x are measured from the centre of the bubble, and a is the radius of the bubble. The impulse associated with such a bubble is given by

$$I_i = \rho v_{\mathrm{b}} [\tfrac{1}{2} V_i - U_i], \tag{28}$$

with $V_i = U_i^{\mathrm{G}} - U_i$. Also, for the case of a single bubble, it is easy to show that the integral on the right-hand side of (25) vanishes on account of symmetry of the flow around an isolated single bubble. Thus, we see that (21) is obtained simply by averaging (25) without the integral term in that equation.

When spatial variations in β or V are non-zero, a contribution will arise from the first term on the right-hand side of (25), and we wish to determine it. For this purpose, we find it most convenient to use an approach similar to that taken by Biesheuvel & Gorissen (1990).

3.1. *Derivation of the momentum equation for the dispersed phase*

We assume that there are N bubbles within a unit cell and that the entire space is filled with replicas of this cell. Let x^α and v^α, $\alpha = 1, 2, \ldots, N$, denote the position and velocities of these bubbles at some instant and let x_L denote the lattice vectors for

the periodic arrangement. The velocity and spatial distribution of the bubbles is thus unchanged upon a translation by x_L. The reason for creating such an artificial periodicity condition in our formulation is simply the need to properly account for the long-range hydrodynamic interactions among bubbles in a definitive manner.

We further assume that the velocity field everywhere in the liquid can be approximated in terms of a velocity potential and that the viscous force on bubbles depends only on the velocities of the bubbles at a given instant and not on their past history. In this case, the complete state is determined if the velocity and position of each bubble is specified. Let $f_N(t, C_N, \dot{C}_N)$ denote the N-bubble probability distribution function for finding bubbles in the neighbourhood of configurations $C_N \equiv (x^1, x^2, \ldots, x^N)$ and $\dot{C}_N \equiv (v^1, v^2, \ldots, v^N)$. Since the unit cell always contains N bubbles, f_N satisfies the normalization condition

$$\int f_N(t, C_N, \dot{C}_N) \, \mathrm{d}C_N \, \mathrm{d}\dot{C}_N = N! \tag{29}$$

Also, since f_N is conserved in the phase space, it satisfies

$$\frac{\partial f_N}{\partial t} + \sum_{\gamma=1}^{N} \boldsymbol{\nabla}_{x^\gamma} \cdot (v^\gamma f_N) + \boldsymbol{\nabla}_{v^\gamma} \cdot (\dot{v}^\gamma f_N) = 0. \tag{30}$$

To obtain averaged continuity or momentum equations for the dispersed phase, we multiply the above equation by an appropriate dynamical variable and then integrate the resulting equation over the phase space of positions and velocities of the bubbles. The dynamical variable can be chosen to be any function of x, C_N, and $\dot{C}_N$ but not of t. For example, the continuity equation for the dispersed phase derived earlier (cf. (23)) can also be obtained if we multiply (30) by the dynamical variable

$$v_b \sum_{\alpha=1}^{N} \sum_L \delta(x - x_L - x^\alpha). \tag{31}$$

It may be noted that integration of the above quantity multiplied by f_N is related to volume fraction β of the gas bubbles by

$$\beta(x, t) = \frac{v_b}{N!} \int \sum_{\alpha=1}^{N} \sum_L \delta(x - x^\alpha - x_L) f_N \, \mathrm{d}C_N \, \mathrm{d}\dot{C}_N. \tag{32}$$

Actually, β computed in this manner is somewhat different from our previous definition (cf. (16)) due to the finite size of the bubbles. The definition (32) assigns all of the volume of the bubble to be located at the centre of the bubble, which is only approximately correct. The difference in the two definitions is significant, however, only when the second-order gradients of β are to be included in the calculations. Since in the present study we wish to derive the averaged momentum equation correct only to the first spatial derivative in β or the velocity field, this distinction between the two definitions of β is not important.

Now multiplying (30) by (31) and integrating over the phase space, we obtain

$$\frac{\partial}{\partial t} \frac{v_b}{N!} \int \sum_{\alpha=1}^{N} \sum_L \delta(x - x^\alpha - x_L) f_N \, \mathrm{d}C_N \, \mathrm{d}\dot{C}_N$$

$$= -\frac{v_b}{N!} \int \sum_{\gamma=1}^{N} [\boldsymbol{\nabla}_{x^\gamma} \cdot (v^\gamma f_N) + \boldsymbol{\nabla}_{v^\gamma} \cdot (\dot{v}^\gamma f_N)] \sum_{\alpha=1}^{N} \sum_L \delta(x - x^\alpha - x_L) \, \mathrm{d}C_N \, \mathrm{d}\dot{C}_N. \tag{33}$$

The right-hand side of the above equation can be simplified with the use of the divergence theorem to yield

$$\frac{v_{\mathrm{b}}}{N!}\int f_N \,\mathrm{d}C_N \,\mathrm{d}\dot{C}_N \sum_{\gamma=1}^{N}\left[\boldsymbol{v}^{\gamma}\cdot\boldsymbol{\nabla}_{x^{\gamma}}+\dot{\boldsymbol{v}}^{\gamma}\cdot\boldsymbol{\nabla}_{v^{\gamma}}\right]\sum_{\alpha=1}^{N}\sum_{L}\delta(\boldsymbol{x}-\boldsymbol{x}^{\alpha}-\boldsymbol{x}_L) \tag{34}$$

plus an integral on the surface of the phase boundary which can be shown to vanish because of the periodicity of the probability density function and the velocity fields. Now, we note that in (34) only the derivative of the delta function with respect to $\boldsymbol{x}^{\gamma}$ with $\gamma=\alpha$ is non-zero, and upon using $\boldsymbol{\nabla}_{x^{\alpha}}\delta(\boldsymbol{x}-\boldsymbol{x}^{\alpha}-\boldsymbol{x}_L)=-\boldsymbol{\nabla}_x\delta(\boldsymbol{x}-\boldsymbol{x}^{\alpha}-\boldsymbol{x}_L)$, we obtain the continuity equation for the dispersed phase (23) with

$$\beta U^{\mathrm{G}}\equiv\frac{v_{\mathrm{b}}}{N!}\int\sum_{\alpha=1}^{N}\sum_{L}\boldsymbol{v}^{\alpha}\delta(\boldsymbol{x}-\boldsymbol{x}_L-\boldsymbol{x}^{\alpha})f_N\,\mathrm{d}C_N\,\mathrm{d}\dot{C}_N. \tag{35}$$

The same procedure can be repeated now for determining the momentum (or impulse) equation for the dispersed phase. For this, we use the dynamical variable

$$v_{\mathrm{b}}\sum_{\alpha=1}^{N}\sum_{L}I_i^{\alpha}\,\delta(\boldsymbol{x}-\boldsymbol{x}_L-\boldsymbol{x}^{\alpha}). \tag{36}$$

Defining an average impulse via

$$\beta I_i\equiv\frac{v_{\mathrm{b}}}{N!}\int\sum_{\alpha=1}^{N}\sum_{L}I_i^{\alpha}\,\delta(\boldsymbol{x}-\boldsymbol{x}^{\alpha}-\boldsymbol{x}_L)f_N\,\mathrm{d}C_N\,\mathrm{d}\dot{C}_N, \tag{37}$$

we obtain

$$\frac{\partial}{\partial t}\beta I_i=-\frac{\partial}{\partial x_j}\frac{v_{\mathrm{b}}}{N!}\int\mathrm{d}C_N\,\mathrm{d}\dot{C}_N f_N\sum_{\alpha=1}^{N}\sum_{L}v_j^{\alpha}I_i^{\alpha}\,\delta(\boldsymbol{x}-\boldsymbol{x}^{\alpha}-\boldsymbol{x}_L)$$
$$+\frac{v_{\mathrm{b}}}{N!}\int\mathrm{d}C_N\,\mathrm{d}\dot{C}_N f_N\sum_{\alpha=1}^{N}\sum_{L}\dot{I}_i^{\alpha}\,\delta(\boldsymbol{x}-\boldsymbol{x}_L-\boldsymbol{x}^{\alpha}), \tag{38}$$

where $\dot{I}_i^{\alpha}$ is defined as

$$\dot{I}_i^{\alpha}=\sum_{\gamma=1}^{N}\left(\boldsymbol{v}^{\gamma}\cdot\boldsymbol{\nabla}_{x^{\gamma}}+\dot{\boldsymbol{v}}^{\gamma}\cdot\boldsymbol{\nabla}_{v^{\gamma}}\right)I_i^{\alpha} \tag{39}$$

and equals the time derivative of the impulse associated with the bubble α following its motion in the dispersion (cf. (25)). We therefore substitute for $\dot{I}_i^{\alpha}$ from (25) into (38) and then average separately each of the three terms on the right-hand side of (25). The viscous and gravitational forces can be averaged in a straightforward manner. The main difficulty is in determining the average of the first term on the right-hand side of (25), which we shall refer to as $\dot{I}_{\mathrm{p}_l}^{\alpha}$. This is the force due to potential flow interactions among the bubbles. As shown in I, the sum of this term over all bubbles in the unit cell vanishes, i.e.

$$\sum_{\alpha=1}^{N}\dot{I}_{\mathrm{p}_l}^{\alpha}=\sum_{\alpha=1}^{N}\rho\int_{S^{\alpha}}(\tfrac{1}{2}u_j^{\mathrm{L}}u_j^{\mathrm{L}}-u_j^{\mathrm{L}}v_j^{\alpha})\,n_i\,\mathrm{d}A=0 \tag{40}$$

when the velocity of the liquid is derived from a scalar potential. Thus, the average of this term over all bubbles in a unit cell is zero for homogeneous flow conditions. In the presence of spatial variations, the last term on the right-hand side of (38) is expressed in terms of a fluid dynamic interaction potential by means of

$$-\frac{\partial}{\partial x_j}(\beta\tau_{ij}^{\mathrm{p}})\equiv\frac{v_{\mathrm{b}}}{N!}\int\mathrm{d}C_N\,\mathrm{d}\dot{C}_N f_N\sum_{\alpha=1}^{N}\sum_{L}\dot{I}_i^{\alpha}\,\delta(\boldsymbol{x}-\boldsymbol{x}^{\alpha}-\boldsymbol{x}_L), \tag{41}$$

where τ_{ij}^{p} is to be determined. Substituting the above result into (38), making use of the continuity relation for the dispersed phase (23), and rearranging, we finally arrive at

$$\frac{\partial \boldsymbol{I}}{\partial t} + \boldsymbol{U}^{\mathrm{G}} \cdot \boldsymbol{\nabla} \boldsymbol{I} = -\frac{1}{\beta} \boldsymbol{\nabla} \cdot (\beta \boldsymbol{\tau}) + \boldsymbol{f}_{\mathrm{v}} + \boldsymbol{f}_{\mathrm{g}}, \tag{42}$$

where

$$\boldsymbol{f}_{\mathrm{v}} = \frac{v_{\mathrm{b}}}{\beta N!} \int \mathrm{d}C_N \, \mathrm{d}\dot{C}_N f_N \sum_{\alpha=1}^{N} \boldsymbol{F}_{\mathrm{v}}^{\alpha} \, \delta(\boldsymbol{x} - \boldsymbol{x}^{\alpha}), \tag{43}$$

$$\boldsymbol{f}_{\mathrm{g}} = \frac{v_{\mathrm{b}}}{\beta N!} \int \mathrm{d}C_N \, \mathrm{d}\dot{C}_N f_N \sum_{\alpha=1}^{N} \boldsymbol{F}_{\mathrm{g}}^{\alpha} \, \delta(\boldsymbol{x} - \boldsymbol{x}^{\alpha}), \tag{44}$$

$$\boldsymbol{\tau} = \boldsymbol{\tau}^{\mathrm{k}} + \boldsymbol{\tau}^{\mathrm{p}} \tag{45}$$

with the kinetic or translational part of the dispersed-phase stress $\boldsymbol{\tau}^{\mathrm{k}}$ defined via

$$\beta \tau_{ij}^{\mathrm{k}} = \frac{v_{\mathrm{b}}}{N!} \int \mathrm{d}C_N \, \mathrm{d}\dot{C}_N f_N \sum_{\alpha=1}^{N} (v_j^{\alpha} I_i^{\alpha} - U_j^{\mathrm{G}} I_i) \, \delta(\boldsymbol{x} - \boldsymbol{x}^{\alpha}). \tag{46}$$

We now proceed to determine τ_{ij}^{p}.

3.1.1. *The potential contribution to the dispersed-phase stress*

The velocity potential can be expressed in terms of multipoles at the centre of each bubble as (cf. I)

$$\phi = \boldsymbol{G} \cdot \boldsymbol{x} + \sum_{\alpha=1}^{N} \sum_{n=1}^{\infty} \boldsymbol{A}_{(n)}^{\alpha} (\cdot)^n \boldsymbol{\nabla}^{(n)} S_1(\boldsymbol{x} - \boldsymbol{x}^{\alpha}), \tag{47}$$

where S_1 is the periodic Green's function for the Laplace equation (Hasimoto 1959; Sangani & Acrivos 1983) which satisfies

$$\boldsymbol{\nabla} \cdot \boldsymbol{\nabla} S_1(\boldsymbol{x}) = 4\pi [\mathscr{V}^{-1} - \sum_L \delta(\boldsymbol{x} - \boldsymbol{x}_L)]. \tag{48}$$

$\mathscr{V}$ is the volume of the unit cell of the periodic lattice, and

$$\boldsymbol{A}_{(n)}^{\alpha} (\cdot)^n \boldsymbol{\nabla}^{(n)} \equiv \sum_{m=0}^{n} (A_{nm}^{\alpha} \Delta_m + \tilde{A}_{nm}^{\alpha} \tilde{\Delta}_m) \, \partial_1^{n-m} \tag{49}$$

with

$$\Delta_m = [\partial_\xi^m + \partial_\eta^m], \quad \tilde{\Delta}_m = i \, [\partial_\xi^m - \partial_\eta^m], \tag{50}$$

$\partial_1 \equiv \partial/\partial x_1$, $(\cdot)^n$ denotes an n-fold scalar product, $\boldsymbol{\nabla}^{(n)}$ and nth-order gradient, $\xi = x_2 + ix_3$, and $\eta = x_2 - ix_3$. $\boldsymbol{G}$ in (47) is related to the average velocity of the dispersion as shown in I. The strengths of 2^n-multipoles, $\boldsymbol{A}_{(n)}$, can be determined as described in I from the boundary conditions on the surface of the bubbles, and subsequently the integral in (25) can be evaluated to determine the force acting on the bubble α due to potential flow interactions. To understand how one evaluates the potential stress from the magnitudes of these multipoles, let us first consider a simple case of well-separated dilute random arrays for which the separation between any two bubbles is large compared to their radii.

Well-separated dilute random arrays. For such arrays, the velocity potential can be adequately approximated by keeping only the leading-order term with $n = 1$ in (47). This amounts to treating the bubbles as point dipoles located at their centres. The magnitude of these dipoles can be determined by first expanding ϕ near the surface of bubble α as

$$\phi = \boldsymbol{C}^{\alpha} \cdot \boldsymbol{s} - \frac{\boldsymbol{A}^{\alpha} \cdot \boldsymbol{s}}{s^3} + \dots, \tag{51}$$

where $A^\alpha \equiv A^\alpha_{(1)}$, $s = x - x^\alpha$, and C^α is related to the regular part of ϕ near x^α by

$$C^\alpha = \nabla\phi^{\mathrm{r}}(x^\alpha). \tag{52}$$

The singular part of ϕ near x^α corresponds to the derivative of the term $1/s$ in the expansion of $S_1(x - x^\alpha)$ near its singularity (cf. (48), Hasimoto 1959), and therefore, the regular part of ϕ is obtained by subtracting this singularity from S_1, i.e.

$$C^\alpha = G + \sum_{\gamma=1}^{N} A^\gamma \cdot \nabla\nabla S_1^{\mathrm{r}}(x^\alpha - x^\gamma), \tag{53}$$

where $S_1^{\mathrm{r}} = S_1 - 1/s$ for $\alpha = \gamma$ and $S_1^{\mathrm{r}} = S_1$ otherwise. The impulse associated with the bubble α (cf. (26)) can be readily evaluated from (51) to be given by

$$I^\alpha = m(A^\alpha a^{-3} - C^\alpha), \tag{54}$$

and the velocity of the bubble by

$$v^\alpha = 2A^\alpha a^{-3} + C^\alpha, \tag{55}$$

where $m = \rho v_{\mathrm{b}}$. Let us now consider the case in which the pressure gradient across the dispersion is held constant. In this case, G is independent of time, and, since we expect the dispersed-phase stress to be independent of the magnitude of the average velocity of the mixture, we may choose it be zero without loss of generality. The force on bubble α due to potential flow interactions can be evaluated from (25) by substituting $u^{\mathrm{L}} = \nabla\phi$. However, a simple calculation will show that the integral in (25) with point dipoles alone used in the local expansion of ϕ (cf. (51)) vanishes. In order to obtain a non-zero estimate, we shall also need to keep the quadrupole $A^\alpha_{(2)}$ term in the expansion. The leading-order estimate, however, can be obtained from the dipole approximation alone by using an alternative expression for the force as derived from the Langrangian (cf. Biesheuvel & Gorissen 1990). This expression is

$$\dot{I}^\alpha_{\mathrm{p}} = \nabla_{x^\alpha} K, \tag{56}$$

where K is the kinetic energy per unit cell which, for $G = 0$, can be evaluated from (cf. I)

$$K = \frac{1}{2}\sum_{\gamma=1}^{N} I^\gamma \cdot v^\gamma. \tag{57}$$

Substituting for I^α and v^α from (54) and (55) into (57), we obtain

$$K = m\sum_{\alpha=1}^{N}[A^\alpha \cdot A^\alpha a^{-6} - \tfrac{1}{2}A^\alpha \cdot C^\alpha a^{-3} - \tfrac{1}{2}C^\alpha \cdot C^\alpha]. \tag{58}$$

In the point-bubble approximation, we regard a as a small quantity, and the inter-bubble distances as $O(1)$. To the leading-order approximation, therefore,

$$A^\alpha = \tfrac{1}{2}a^3 v^\alpha. \tag{59}$$

Now using (53) to evaluate C^α and taking the gradient of K with the position of the bubble yields the following leading-order estimate of the potential interaction force on the bubble α:

$$\dot{I}^\alpha_{\mathrm{p}} = -\pi\rho a^6 \sum_{\gamma=1}^{N} v^\alpha v^\gamma (\,\cdot\,)^2 \nabla^{(3)} S_1^{\mathrm{r}}(x^\alpha - x^\gamma), \tag{60}$$

where the gradients are to be evaluated with respect to $x^\alpha - x^\gamma$. Expressed in this

manner, we see that the potential interaction force on bubble α, i.e. $\dot{I}^{\alpha}_{\mathrm{p}}$, may be thought of as a sum of forces acted upon by each bubble γ in the dispersion with a pair potential between the bubbles α and γ being, roughly speaking, proportional to $\nabla\nabla S^{\mathrm{r}}_1(x^{\alpha}-x^{\gamma})$. The total potential can then be obtained by adding over all the pairs in the dispersion. This simple scheme, however, needs some modification since $\nabla\nabla S^{\mathrm{r}}_1(x^{\alpha}-x^{\gamma})$ behaves like $1/|x^{\alpha}-x^{\gamma}|^3$ for distances that are small compared to the unit cell size, and hence this sum may not converge for infinitely large unit cells. In other words, we need to proceed with some caution.

As pointed out in Sangani & Acrivos (1983), the use of the periodic Green's function S_1 in the analysis avoids the difficulties associated with the non-convergence of long-range interactions. It is easy to show, using the Green's identity, and the Green's function for the Laplace equation in an infinite space $(1/|x|)$, that S_1 has the following representation:

$$S_1(x) = \sum_L \varphi(x-x_L) \equiv \sum_L \left[\frac{1}{|x-x_L|} - \frac{1}{\mathscr{V}} \int_{\mathscr{V}} \frac{\mathrm{d}x'}{|x-x_L-x'|} \right]. \tag{61}$$

For large $|x-x_L|$, the integrand in the above expression can be expanded in a Taylor series to show that for cubic unit cells φ decays as $1/|x-x_L|^5$. Thus, although the summation of each term inside the square bracket in (61) diverges as $|x_L| \to \infty$, the overall expression for S_1 converges and the value of S_1 at a point within the basic unit cell does not depend on the manner in which we take the limit $|x_L| \to \infty$. Now we follow Biesheuvel & Gorissen (1990) and determine the expression for the potential contribution to the stress tensor using the particular form of pairwise decomposition we have obtained in (60). In the following manipulations, we take bubble α to be in the basic unit cell and write the total force on it as a sum of forces due to each bubble γ and its periodic images. Thus

$$\int f_N \, \mathrm{d}C_N \, \mathrm{d}\dot{C}_N \sum_{\alpha=1}^{N} \dot{I}^{\alpha}_{\mathrm{p}_i} \delta(x-x^{\alpha}) = \int f_N \, \mathrm{d}C_N \, \mathrm{d}\dot{C}_N \sum_{\gamma=1}^{N} \sum_L \dot{I}^{\gamma}_{\mathrm{p}_i} \delta(x-x_L-x^{\gamma}) \tag{62a}$$

$$= -\pi\rho a^6 \int f_N \, \mathrm{d}C_N \, \mathrm{d}\dot{C}_N \sum_{\alpha=1}^{N} \sum_{\gamma=1}^{N} \sum_L v^{\alpha}_j v^{\gamma}_k \partial^3_{ijk}[\varphi(R-x_L)] \, \delta(x-x^{\alpha}) \tag{62b}$$

$$= \pi\rho a^6 \int f_N \, \mathrm{d}C_N \, \mathrm{d}\dot{C}_N \sum_{\gamma=1}^{N} \sum_L \sum_{\alpha=1}^{N} v^{\alpha}_j v^{\gamma}_k \partial^3_{ijk}[\varphi(R-x_L)] \, \delta(x-x_L-x^{\gamma}) \tag{62c}$$

$$= -\tfrac{1}{2}\pi\rho a^6 \int f_N \, \mathrm{d}C_N \, \mathrm{d}\dot{C}_N \sum_{\alpha=1}^{N} \sum_{\gamma=1}^{N} \sum_L v^{\alpha}_j v^{\gamma}_k \partial^3_{ijk}[\varphi(R-x_L)]$$
$$\times (\delta(x-x^{\alpha}) - \delta(x-x_L-x^{\gamma})), \tag{62d}$$

where $R = x^{\alpha}-x^{\gamma}$ and $\partial_i \equiv \partial/\partial R_i$. Equation (62a) is a simple identity in which the summation over α is replaced by the summation over γ; (62b) is obtained from (60); (62c) is obtained by making use of the fact that $\nabla^{(3)}\varphi(R)$ is an odd function of R; and (62d) is obtained by adding the terms on the right-hand sides of (62b) and (62c) and dividing by two.

Now to express the above quantity as the divergence of a stress tensor, we use the usual procedure adopted in the statistical mechanics literature (see, for example, Rice & Gray 1965) and expand the delta function in the Taylor series

$$\delta(x-x_L-x^{\gamma}) = \delta(x-x^{\alpha}) + (R-x_L)_j \frac{\partial}{\partial x_j} \delta(x-x^{\alpha}) + \dots. \tag{63}$$

Since $\partial^3_{ijk}\varphi$ decays as $1/|x-x_L|^8$, the use of Taylor series expansion of the delta function is justified even for large $x-x_L$. Substituting the above expansion in (62), and comparing the resulting expression with (41), we obtain the expression for the potential flow interaction contribution to the dispersed-phase stress as

$$\beta\tau^{\mathrm{p}}_{ij} = \frac{v_{\mathrm{b}}}{2N!}\int f_N\,\mathrm{d}C_N\,\mathrm{d}\dot{C}_N\sum_{\alpha=1}^{N}\sum_{\gamma=1}^{N}(-\pi\rho a^6)\,v^\alpha_m v^\gamma_n\,\Psi_{imnj}(R)\,\delta(x-x^\alpha), \qquad (64)$$

where
$$\Psi_{imnj}(R) = \sum_L (R-x_L)_j\,\partial^3_{imn}\varphi(R-x_L). \qquad (65)$$

It is easy to verify that the above quantity converges as $|x_L|\to\infty$. For the purpose of numerical evaluation of the stress, it is useful to express this quantity in terms of periodic functions S_1 and S_2 as Ewald's summation technique described in Hasimoto (1959) can then be readily used. Here, the function S_2 satisfies $\nabla^2 S_2 = S_1$ and is given by

$$S_2(R) = \sum_L\left[\frac{|R-x_L|}{2}-\frac{1}{\mathscr{V}}\int_{\mathscr{V}}\frac{|R-x_L-x'|}{2}\,\mathrm{d}x'\right]. \qquad (66)$$

The summation in this case does not converge even though S_2 itself is finite. Thus, the above representation should not be used for evaluating S_2. In the present study, however, we shall only be interested in $\nabla^{(n)}S_2$ with $n\geqslant 4$, and it can be seen that such derivatives of S_2 can be evaluated from (66) as the sum is then convergent.

To recast (65) in terms of derivatives of S_1 and S_2, we use the identity

$$R_i\,\partial^n_{i_2\ldots i_{n+1}}R^{-1} = \partial^{n+1}_{i_1\ldots i_{n+1}}R-(\delta_{i_1 i_2}\partial^{n-1}_{i_3\ldots i_{n+1}}+\delta_{i_1 i_3}\partial^{n-1}_{i_2\ldots i_{n+1}}+\ldots n\text{ terms})R^{-1}. \qquad (67)$$

With $n=3$ in the above identity, and summing over x_L, we obtain

$$\Psi_{imnj} = 2\partial^4_{ijmn}S^{\mathrm{r}}_2-(\delta_{ij}\partial^2_{mn}+\delta_{jm}\partial^2_{in}+\delta_{jn}\partial^2_{in})S^{\mathrm{r}}_1+\psi^{\mathrm{ren}}_{imnj}, \qquad (68)$$

where

$$\Psi^{\mathrm{ren}}_{imnj} = \frac{1}{\mathscr{V}}\sum_L\int_{\mathscr{V}}\left\{\partial^4_{ijmn}|R-x_L-x'|-(\delta_{ij}\partial^2_{mn}+\delta_{jm}\partial^2_{in}+\delta_{jn}\partial^2_{im})\frac{1}{|R-x_L-x'|}\right.$$
$$\left.-(R-x_L)_j\,\partial^3_{imn}\frac{1}{|R-x_L-x'|}\right\}\mathrm{d}x' = -\frac{1}{\mathscr{V}}\sum_L\int_{\mathscr{V}}x'_j\,\partial^3_{imn}\frac{1}{|R-x_L-x'|}\,\mathrm{d}x'. \qquad (69)$$

It can be shown that, for a simple cubic unit cell, the above quantity is independent of R and equals

$$\Psi^{\mathrm{ren}}_{imnj} = \frac{4\pi}{\mathscr{V}}\delta_{imnj}, \qquad (70)$$

where δ_{imnj} equals unity for $i=j=m=n$ and zero otherwise.

Non-dilute arrays. We now return to the case of non-dilute arrays. The key step in the previous derivation was the decomposition of the total potential interaction force on bubble α in terms of a sum of the forces exerted by the other bubbles. For the general case of non-dilute dispersions, such a decomposition may not always be possible. Indeed, a straightforward calculation of the total kinetic energy of the system in terms of higher-order reflections would quickly involve interactions among three or more bubbles. Fortunately though, such a decomposition is possible in the present case as we shall now show.

The formula (56) for evaluating the potential interaction force from the gradient of the kinetic energy is not suitable for non-dilute arrays, as the explicit dependence of the total kinetic energy on the position of the bubbles is not known. Instead, it proves more convenient to evaluate the integral in (25) directly from the local expansion of ϕ near the surface of bubble α. Defining $\phi^* = \phi - v^\alpha \cdot (x - x^\alpha)$, we have

$$\dot{I}_{\mathrm{p}}^\alpha = \rho \int_{S^\alpha} (\tfrac{1}{2} u^{\mathrm{L}} \cdot u^{\mathrm{L}} - u^{\mathrm{L}} \cdot v^\alpha) \, n \, \mathrm{d}A = \rho \int_{S^\alpha} \tfrac{1}{2} \nabla \phi^* \cdot \nabla \phi^* n \, \mathrm{d}A. \tag{71}$$

Next, let the expansion of ϕ^* near the surface of bubble α in spherical harmonics be given by (cf. I)

$$\phi^* = \sum_{n=0}^{\infty} \sum_{m=0}^{n} [f_{nm}^\alpha(s) \, Y_{nm} + \tilde{f}_{nm}^\alpha(s) \, \tilde{Y}_{nm}], \tag{72}$$

with

$$f_{nm}^\alpha(s) = C_{nm}^\alpha \, s^n + D_{nm}^\alpha \, s^{-n-1} \tag{73}$$

and $s = x - x^\alpha$. Now the boundary conditions at the surface of the bubble give the relationship

$$C_{nm}^\alpha a^n = \frac{n+1}{n} D_{nm}^\alpha a^{-n-1}. \tag{74}$$

Similar expressions apply to $\tilde{f}_{nm}^\alpha$. Substituting (72) into (71), and simplifying, we obtain a relatively simple expression for $\dot{I}_{\mathrm{p}}^\alpha$. For example (cf. I),

$$\dot{I}_{\mathrm{p}_1}^\alpha = \sum_{n=1}^{\infty} \sum_{m=0}^{n} H_{nm} [f_{nm}^\alpha f_{n+1,m}^\alpha + \tilde{f}_{nm}^\alpha \tilde{f}_{n+1,m}^\alpha], \tag{75}$$

where the functions f_{nm}^α etc. are to be evaluated at $s = a$, and H_{nm} is given by

$$H_{nm} = \frac{2\pi\rho n(n+2)(n+m+1)!}{(2n+1)(2n+3)(n-m)!}(1+\delta_{m0}). \tag{76}$$

Now we make use of (73) and (74) to obtain

$$f_{nm}^\alpha(a) = \frac{2n+1}{n} D_{nm}^\alpha a^{-n-1} = \frac{2n+1}{n+1} C_{nm}^\alpha a^n. \tag{77}$$

Substituting for f_{nm}^α and $f_{n+1,m}^\alpha$ in (75) in terms of D_{nm}^α and $C_{n+1,m}^\alpha$ respectively, we obtain

$$\dot{I}_{\mathrm{p}_1}^\alpha = \sum_{n=1}^{\infty} \sum_{m=0}^{n} 2\pi\rho(1+\delta_{m0})\frac{(n+m+1)!}{(n-m)!}[D_{nm}^\alpha C_{n+1,m}^\alpha + \tilde{D}_{nm}^\alpha \tilde{C}_{n+1,m}^\alpha]. \tag{78}$$

Now we make use of the fact that $C_{n+1,m}^\alpha$ must be related to the regular part of ϕ near x^α (cf. (13) in I) by

$$C_{n+1,m}^\alpha = \frac{1}{(n+m+1)!}\frac{(-2)^m}{(1+\delta_{m0})}\left[\left(\frac{\partial}{\partial x_1}\right)^{n-m+1} \Delta_m \phi^{\mathrm{r}}\right]_{x=x^\alpha}. \tag{79}$$

Substituting for the regular part of ϕ from (47), using the relations between D_{nm}^α and A_{nm}^α (cf. (12) in I), combining (78) with (79), and rearranging we obtain

$$\dot{I}_{\mathrm{p}_1}^\alpha = \sum_{n=1}^{\infty} \sum_{k=1}^{\infty} \sum_{\gamma=1}^{N} 4\pi\rho(-1)^n[A_{(n)}^\alpha A_{(k)}^\gamma (\cdot)^{k+n} \nabla^{(k+n)}]\partial_1 S_1^{\mathrm{r}}(R) \tag{80}$$

in which $R = x^\alpha - x^\gamma$. Similar calculations can be made for the x_2- and x_3-components of $\dot{I}_p^\alpha$, and the final result is

$$\dot{I}_p^\alpha = \sum_{n=1}^\infty \sum_{k=1}^\infty \sum_{\gamma=1}^N 4\pi\rho(-1)^n A_{(n)}^\alpha A_{(k)}^\gamma (\cdot)^{n+k} \boldsymbol{\nabla}^{(n+k+1)} S_1^r(\boldsymbol{R}). \tag{81}$$

It may be noted that this result is valid for any $\boldsymbol{G}$ (cf. (47)) since evaluation of $C_{n+1,m}^{l\alpha}$ requires second- and higher-order derivatives of ϕ for $n \geqslant 1$ and these derivatives of $\boldsymbol{G}\cdot\boldsymbol{x}$ vanish identically.

For the point-bubble approximation, $A_{(1)}^\alpha = \frac{1}{2}a^3 v^\alpha$, and thus we recover the expression (60) by keeping only the terms with $k = n = 1$ in (81). For non-dilute random dispersions, the multipoles $A_{(n)}^\alpha$ indeed depend on the interactions among many bubbles, but, as far as the derivation of the potential flow stress is considered, these multipoles may be regarded as constants, just as the velocities of the bubbles were treated as constants in our derivation of the stress tensor for the well-separated dilute random arrays. It may be noted that although (81) has some similarity to the expression based on the kinetic energy (cf. (56)), there is no direct relation between the terms appearing in it to those in the evaluation of the kinetic energy. Since the constants that appear in the expression for K are functions of the positions of all the bubbles, it is unlikely that the result (81) can be derived starting from (56).

The result (81) gives us an *exact* formula for decomposing the potential flow interaction force ($\dot{I}_p^\alpha$) into the sum of forces exerted by all other bubbles in the dispersion. Now, it is straightforward to derive the exact expression for the stress tensor. If we take the mean relative motion to coincide with the x_1-axis, it will suffice to evaluate τ_{ii}^p and τ_{11}^p because of the isotropy of the microstructure in the plane transverse to the mean relative motion. Substituting (49) into (80), making use of (61) and (67), and simplifying we obtain

$$\tau_{11}^p = \frac{3\rho v_b}{2Na^3}[\Sigma(-1)^n(A_{nm}^\alpha \Delta_m + \tilde{A}_{nm}^\alpha \tilde{\Delta}_m)(A_{kj}^\gamma \Delta_j + \tilde{A}_{kj}^\gamma \tilde{\Delta}_j)\,\partial_1^p\{2\partial_{11}^2 S_2^r - (p+1)S_1^r\} + \tau_{11}^{ren}], \tag{82}$$

$$\tau_{ii}^p = \frac{3\rho v_b}{2Na^3}[\Sigma(-1)^{n+1}(k+n+1)(A_{nm}^\alpha \Delta_m + \tilde{A}_{nm}^\alpha \tilde{\Delta}_m)(A_{kj}^\gamma \Delta_j + \tilde{A}_{kj}^\gamma \tilde{\Delta}_j)\,\partial_1^p S_1^r + \tau_{ii}^{ren}], \tag{83}$$

where the summation is to be carried over n, m, k, j, α, and γ, the functions S_1^r and S_2^r and their derivatives are to be evaluated at $R = x^\alpha - x^\gamma$, $p = n - m + k - j$, and τ_{11}^{ren} and τ_{ii}^{ren} are the contributions that arise from terms similar to ψ_{imnj}^{ren} in (69). Only the dipole terms contribute to these quantities because the integrals such as one on the extreme right of (69) vanish when derivatives of order greater than 3 of $1/|\boldsymbol{R} - \boldsymbol{x}_L - \boldsymbol{x}'|$ are involved. This is not surprising as one would expect the renormalization of S_1, represented by the integral term in (61), to be necessary only for evaluating its derivatives of order less than or equal to 2. Thus, we obtain

$$\tau_{ii}^{ren} = -\sum_{\alpha=1}^N \sum_{\gamma=1}^N \frac{4\pi}{\mathcal{V}}[4A_{10}^\alpha A_{10}^\gamma + A_{11}^\alpha A_{11}^\gamma + \tilde{A}_{11}^\alpha \tilde{A}_{11}^\gamma], \tag{84}$$

$$\tau_{11}^{ren} = -\sum_{\alpha=1}^N \sum_{\gamma=1}^N \frac{16\pi}{\mathcal{V}}A_{10}^\alpha A_{10}^\gamma. \tag{85}$$

The strengths of the multipoles can be determined directly by solving the many-bubble interaction problem as shown in I. The derivatives of S_1 can be evaluated from the formula given in Sangani, Zhang & Prosperetti (1991) which employs

Ewald's summation technique. Similar expression can also be obtained for the derivatives of S_2, and hence (82)–(85) provide us with an exact method for determining the potential flow interaction contribution to the dispersed phase.

Collisional contribution to the dispersed-phase stress. As discussed in detail in I, there is experimental evidence (Kok 1989) to suggest that bubbles do not coalesce when a small amount of surface-active impurity is present in the dispersion. In our simulations, we assume that bubbles collide in such a way as to conserve the kinetic energy and the momentum of the liquid. The timescale for collision is assumed to be small compared to the inertial timescale. It is further assumed that the fluid dynamic interactions in the gap between the colliding pair of bubbles is manifested as an instantaneous force (on the timescale of inertial motion) of collision on the colliding bubbles. Thus, for times close to a collision time t_c, we write

$$\dot{I}_p^1 = m_c F_c \, \delta(t - t_c) = -\dot{I}_p^2; \quad \dot{I}^\alpha = 0 \quad \text{for} \quad \alpha = 3, 4, \dots, \tag{86}$$

where 1–2 is the colliding pair of bubbles, $m_c = (x^1 - x^2)/2a$, and F_c is the magnitude of the impulse acting on the colliding bubbles that is to be determined from the conservation of the kinetic energy of the liquid (cf. I). For evaluating the contribution to the dispersed-phase stress due to collisions, the trial function must strictly be taken as a function of $(C_N, \dot{C}_N)$ and thus no explicit dependence on time is permissible. This can be done by replacing $\delta(t - t_c)$ in the above by $\delta(|x^1 - x^2| - 2a)$. Now we use

$$\sum_{\alpha=1}^{N} \dot{I}_p^\alpha \, \delta(x - x^\alpha) = \tfrac{1}{2}(\dot{I}_p^1 + \dot{I}_p^2)\,(\delta(x - x^1) + \delta(x - x^2)) + \tfrac{1}{2}(\dot{I}_p^1 - \dot{I}_p^2)\,(\delta(x - x^1) - \delta(x - x^2))$$

$$= -\tfrac{1}{4}\nabla \cdot \{(x^1 - x^2)\,(\dot{I}_p^1 - \dot{I}_p^2)\,\delta(x - x^1) + (x^2 - x^1)\,(\dot{I}_p^2 - \dot{I}_p^1)\,\delta(x - x_2)\} \tag{87}$$

to obtain the collision contribution as given by

$$\tau^c = \frac{3v_b}{2Na^2 T'} \sum_c F_c\, m_c\, m_c, \tag{88}$$

where T' is the length of the time interval and the summation is over all collisions occurring during that interval.

3.2. *Remarks on the derivation*

The approach we have taken in deriving the expression for the stress tensor is similar to that used in deriving the expression for pressure in pure liquids from statistical mechanics principles. There the problem is to average out the molecular interactions to arrive at the pressure to be used in the continuum description of liquids. The pressure in a liquid is the sum of two quantities. The first, referred to as the translational or the kinetic part of the pressure, is related to the average of $m'v_i^\alpha v_j^\alpha - m'V_i V_j$, where m' is the mass of a molecule, v_i^α its velocity, and V_l the average velocity. In the case of bubbles dispersed in liquids, the mass of a bubble is, of course, negligible, but there is still a virtual mass associated with its motion under potential flow conditions. The impulse I^α in the dynamics of bubbly liquids plays a role similar to that of momentum $m'v^\alpha$ in molecular dynamics. Thus, (46) simply represents the translational contribution to the stress tensor. For ideal gases in which the interaction among molecules is negligible, the pressure is entirely due to this translational part. For dense gases and liquids, there is a second part of the pressure that arises from the interactions among molecules. In molecular dynamics, it is

customary to represent these intermolecular forces in terms of pair potentials, and thus the second part of the pressure is usually expressed in terms of these pair potentials. Deriving the equivalent form of pair potentials for bubbly liquids is the main contribution of the present study. We showed that while the multiparticle interactions are quite important, it is still possible to interpret the force acting on a representative bubble as a sum of pairwise interaction forces. The intermolecular forces in ordinary liquids are short range and therefore, the pair potentials can be added in a straightforward manner to determine the potential contribution to the pressure. In the statistical mechanics literature for pressure in liquids, the potential part of the pressure is sometimes expressed in terms of a quantity which is the sum of $x_j^\alpha F_i^\alpha$ over all the molecules. Here F_i^α is the force on the molecule α. When the intermolecular forces are short range, this alternative expression provides an efficient method for determining the pressure via simulations. We have followed a different approach here mainly because the interactions in our case are long range.

It may be noted that (42) is equivalent to (5.13) in van Wijngaarden & Kapteyn (1990). A comparison of these two expressions suggests that their dispersed-phase tensor is the same as the translational part in our (46). It may appear at first, by analogy with the theories for pressure in gases at low number densities where the translational part is the most important one, that the expression that van Wijngaarden & Kapteyn obtained must be valid when β is small. This, however, is not the case as there is an important difference between the two systems. In gases at finite temperatures, there is considerable randomness in the motion of molecules, which makes the variance in velocity, and hence the pressure, finite even in the limit of zero number density. In the gas–liquid dispersions that we have examined, the variance in the velocity of the bubbles vanishes in the limit of small β (provided that the bubbles do not form aggregates), and thus both the translational and potential parts of the stress tensor are of comparable magnitude. Indeed, as we shall see in §4, the potential contribution to the stress is two to three times larger than the translational contribution for β as small as 0.1.

3.3. *Energy equation for the dispersed phase*

The momentum and continuity equations for the dispersed phase derived in previous sections (cf. (23) and (42)) constitute a total of four scalar relations among β, $V_i = U_i^G - U_i$, I_i, and the average frictional force $f_{i,v}$. In the limit of large Reynolds number flows, the kinetic energy of the liquid remains approximately constant over a timescale small compared to the viscous relaxation time $a^2\rho/\mu$ and the microstructure of the dispersion depends on the kinetic energy of the liquid. Thus I_i will not only depend on β and V_i but also on the kinetic energy of the dispersion. When the Reynolds number is large but finite, this kinetic energy will slowly change with time, and hence, after a sufficiently long time, the kinetic energy of the liquid, and thereby the microstructure of the dispersion, under homogeneous and equilibrium conditions will depend only on β and V_i. Thus, I_i and $f_{i,v}$ will eventually be functions of β and V_i only. When spatial and temporal fluctuations from this homogeneous state occur, I_i will vary because of the variations in the kinetic energy in addition to those in β and V_i. Thus, for a correct analysis of such flows, we must also include an additional relation corresponding to the changes in kinetic energy. Such equations are also used in the study of granular flows.

The energy equation can be derived in essentially the same manner as the momentum equation. The key is the choice of the proper trial function such as (36) for the momentum equation. In particular, the trial function to be selected must

satisfy a condition such as (40). As discussed in detail in I, which quantities remain invariant depends on the conditions of simulations. If the average velocity of the liquid phase, or the pressure gradient across the dispersion, are maintained constant, then the sum of e^α remains invariant. Here, e^α is defined by

$$e^\alpha = \tfrac{1}{2} I_i^\alpha v_i^\alpha. \tag{89}$$

If, however, the average velocity of the mixture is held constant during simulation, the sum of e^α defined via (57) in I remains invariant. In the present study, we shall determine the average quantities for the former condition, i.e. for the constant average velocity of the liquid phase. Thus, we shall use (89) as our definition of e^α.

Now the derivation of the energy equation is quite straightforward and, for inviscid interactions, yields

$$\frac{\partial e}{\partial t} + U_j^{\mathrm{G}} \frac{\partial e}{\partial x_j} = -\frac{1}{\beta} \frac{\partial}{\partial x_j} (\beta q_j), \tag{90}$$

where

$$e = \frac{v_{\mathrm{b}}}{\beta N!} \int \sum_{\alpha=1}^{N} e^\alpha \delta(\boldsymbol{x} - \boldsymbol{x}^\alpha) f_N \, \mathrm{d}C_N \, \mathrm{d}\dot{C}_N, \tag{91}$$

and q_j is the energy flux vector for the dispersed phase and consists of three terms q_j^{k}, q_j^{p}, and q_j^{c} corresponding, respectively, to the kinetic, potential and collisional contributions. The kinetic part is given by

$$q_j^{\mathrm{k}} = \frac{v_{\mathrm{b}}}{\beta N!} \int \mathrm{d}C_N \, \mathrm{d}\dot{C}_N f_N \sum_{\alpha=1}^{N} (v_j^\alpha e^\alpha - U_j^{\mathrm{G}} e) \tag{92}$$

whereas the potential and collisional contributions are defined via

$$-\frac{\partial}{\partial x_j} \beta(q_j^{\mathrm{p}} + q_j^{c}) = \frac{v_{\mathrm{b}}}{N!} \int f_N \, \mathrm{d}C_N \, \mathrm{d}\dot{C}_N \sum_{\alpha=1}^{N} \dot{e}^\alpha \delta(\boldsymbol{x} - \boldsymbol{x}^\alpha). \tag{93}$$

We have been unable to obtain an exact expression for the time rate of change of energy associated with bubble α, i.e. $\dot{e}^\alpha$, as the sum of pair interaction potential energies. Therefore, an approximate expression based on point bubbles (dilute well-separated arrays) will be presented here. The accuracy of this expression will be tested against the numerical results for $\dot{e}^\alpha$ in the next section.

To determine the point-bubble approximation for $\dot{e}^\alpha$, let us choose $\boldsymbol{G} = 0$. The impulse and velocity of bubble α are related to the dipoles by (54) and (55). Taking the time derivative of the scalar product of these two expressions, we obtain

$$\dot{e}^\alpha = \tfrac{1}{2} m (2 \boldsymbol{A}^\alpha \cdot \dot{\boldsymbol{A}}^\alpha a^{-6} - \dot{\boldsymbol{C}}^\alpha \cdot \boldsymbol{A}^\alpha a^{-3} - \boldsymbol{C}^\alpha \cdot \dot{\boldsymbol{A}}^\alpha a^{-3}). \tag{94}$$

To determine $\dot{e}^\alpha$ to leading order when a is small, we note that $\boldsymbol{A}^\alpha, \dot{\boldsymbol{C}}^\alpha = O(a^3)$, $\dot{\boldsymbol{A}}^\alpha = O(a^6)$. Let us first determine $\dot{\boldsymbol{C}}^\alpha$ by differentiating (53). Taking $\boldsymbol{G} = 0$, the differentiation gives two terms; the first involves a derivative of $\boldsymbol{A}^\gamma$ and the second involves that of $\boldsymbol{x}^\alpha - \boldsymbol{x}^\gamma$, of which the latter is larger. Thus

$$\dot{\boldsymbol{C}}^\alpha = \frac{a^3}{2} \sum_{\gamma=1}^{N} \boldsymbol{v}^\gamma (\boldsymbol{v}^\alpha - \boldsymbol{v}^\gamma) (\cdot)^2 \boldsymbol{\nabla}^{(3)} S_1^{\mathrm{r}}(\boldsymbol{x}^\alpha - \boldsymbol{x}^\gamma), \tag{95}$$

where we used $\boldsymbol{A}^\alpha = \tfrac{1}{2} \boldsymbol{v}^\alpha a^3$. Now using the expression for $\dot{\boldsymbol{I}}^\alpha$ (cf. (60)) and combining it with (95) and the time derivative of (54) we obtain

$$\dot{\boldsymbol{A}}^\alpha = -\frac{a^6}{4} \sum_{\gamma=1}^{N} (\boldsymbol{v}^\alpha + 2\boldsymbol{v}^\gamma) \boldsymbol{v}^\gamma (\cdot)^2 \boldsymbol{\nabla}^{(3)} S_1^{\mathrm{r}}(\boldsymbol{x}^\alpha - \boldsymbol{x}^\gamma). \tag{96}$$

Combining the above leading-order estimates of $\dot{A}^\alpha$ and $\dot{C}^\alpha$ with (94) and noting that, to the leading order, $\frac{1}{2}A^\alpha a^{-3} = v^\alpha = I^\alpha/(2m)$ we obtain three equivalent expressions for $\dot{e}^\alpha$:

$$\dot{e}^\alpha = -\frac{\pi\rho a^6}{2}\sum_{\gamma=1}^{N} \boldsymbol{v}^\alpha \boldsymbol{v}^\gamma (\boldsymbol{v}^\alpha + \boldsymbol{v}^\gamma)\,(\,\cdot\,)^3 \boldsymbol{\nabla}^{(3)} S_1^r(\boldsymbol{x}^\alpha - \boldsymbol{x}^\gamma)$$

$$= -\frac{4\pi\rho}{a^3}\sum_{\gamma=1}^{N} \boldsymbol{A}^\alpha \boldsymbol{A}^\gamma (\boldsymbol{A}^\alpha + \boldsymbol{A}^\gamma)\,(\,\cdot\,)^3 \boldsymbol{\nabla}^{(3)} S_1^r(\boldsymbol{x}^\alpha - \boldsymbol{x}^\gamma)$$

$$= -\frac{4\pi\rho}{a^3 m^3}\sum_{\gamma=1}^{N} \boldsymbol{I}^\alpha \boldsymbol{I}^\gamma (\boldsymbol{I}^\alpha + \boldsymbol{I}^\gamma)\,(\,\cdot\,)^3 \boldsymbol{\nabla}^{(3)} S_1^r(\boldsymbol{x}^\sigma - \boldsymbol{x}^\gamma). \tag{97}$$

It is easy to verify that the sum of $\dot{e}$ over all the bubbles in the dispersion vanishes, as it should for the case of inviscid interactions.

As mentioned earlier, we have been unable to derive an exact decomposition of $\dot{e}^\alpha$ in pair potential energies, and therefore we would like to use the results of the above analysis to obtain an approximate pair decomposition. The three expressions listed above are equivalent for the dilute case, but one of these three may provide better approximation to the actual $\dot{e}^\alpha$ in non-dilute dispersions, which can be evaluated by using a forward time difference formula on the values of e^α obtained in numerical simulations. A comparison of the actual values with those obtained from the above approximations is given in §4, where we find that the relative error is least when the approximation based on $\boldsymbol{A}^\alpha$ is used and largest when that based on $\boldsymbol{I}^\alpha$ is used. Now, comparing (97) and (60) with the exact expression for $\dot{I}^\alpha$ (cf. (80)) we obtain yet another approximation, which is an improvement over that given by (97):

$$\dot{e}^\alpha = \frac{4\pi\rho}{a^3} \Sigma(-1)^n \boldsymbol{A}_{(n)}^\alpha \boldsymbol{A}_{(k)}^\gamma (\boldsymbol{A}^\alpha + \boldsymbol{A}^\gamma)\,(\,\cdot\,)^{n+k+1} \boldsymbol{\nabla}^{(n+k+1)} S_1^r(\boldsymbol{x}^\alpha - \boldsymbol{x}^\gamma). \tag{98}$$

The summation in (98) is to be carried over n, k, and γ. Now the potential contribution to the energy flux can be determined following essentially the same procedure as for $\boldsymbol{\tau}^p$ and, in fact, the final expression can be written down by multiplying (82) with $(\boldsymbol{A}^\alpha + \boldsymbol{A}^\gamma)/a^3$:

$$q_1^p = \frac{3\rho v_b}{Na^6}\left[\Sigma(-1)^n (A_{nm}^\alpha \Delta_m + \tilde{A}_{nm}^\alpha \tilde{\Delta}_m)(A_{kj}^\gamma \Delta_j + \tilde{A}_{kj}^\gamma \tilde{\Delta}_j)(A_{10}^\alpha + A_{10}^\gamma)\,\partial_1^p \right.$$

$$\left. \times \{2\partial_{11}^2 S_2^r - (p+1)S_1^r\} + q_1^{ren}\right], \tag{99}$$

where the summation is over n, m, k, j, γ, and α; $p = n - m + k - j$; S_1^r and S_2^r are evaluated at $\boldsymbol{R} = \boldsymbol{x}^\alpha - \boldsymbol{x}^\gamma$. Here, we have assumed that the mean relative motion is in the x_1-direction. The microstructure is expected to be isotropic in the x_1 plane and hence $q_2^p = q_3^p = 0$. Finally,

$$q_1^{ren} = -\sum_{\alpha=1}^{N}\sum_{\gamma=1}^{N} \frac{16\pi}{\mathscr{V}} A_{10}^\alpha A_{10}^\gamma (A_{10}^\alpha + A_{10}^\gamma). \tag{100}$$

We note that it may be possible to obtain a more accurate expression for q_1^p by adding a virial-type expression to the above, i.e. by adding

$$\frac{1}{N}\sum_{\alpha=1}^{N} \Delta \dot{e}^\alpha x_1^\alpha \tag{101}$$

to (99). Here $\Delta \dot{e}^\alpha$ is the difference between the actual value of $\dot{e}^\alpha$, as evaluated from numerically differentiating the values of e^α obtained in simulations, and its approximate value as given by (98). In the present study, however, we have used only the approximate relation (99).

Finally, the collisional contribution to the dispersed-phase energy flux was also evaluated by an approximate relation. During the collision, impulses of only the colliding bubbles change, whereas, since the velocities of all the bubbles change, $\dot{e}^\alpha$ is non-zero for all the bubbles. Once again, it is difficult to obtain a pair decomposition of the potential energies during the collision, and hence we used an approximate relation in which the changes in e^α of non-colliding bubbles were neglected. Thus, we used

$$q_1^c = \frac{1}{2NT'} \Sigma (\Delta e^1 - \Delta e^2)(x_1^1 - x_1^2), \tag{102}$$

where 1–2 is the colliding pair of bubbles, Δ denotes the total change over a collision, and the summation is carried over all the collisions during a time interval of length T'.

4. Results and discussion

The averaged equations and the expressions for various averaged quantities, including the dispersed-phase stress and energy flux derived in previous sections, should be combined with dynamic simulations for various macroscopic flows to obtain their macroscale description. We shall study specific flows in more detail in our future work. Here, we briefly consider three special cases.

4.1. *Periodic arrays*

The first is the case of bubbles arranged on the lattice of a periodic array rising under gravity. In this special case, it is possible to obtain analytical expressions for the dispersed-phase stress and energy flux and this is quite useful in testing the accuracy of the numerical calculations, particularly in view of the rather complicated nature of the expressions for these quantities. Moreover, the results for periodic arrays serve as benchmarks.

For dilute cubic arrays with the velocity of all the bubbles equal to V relative to the mixture and aligned along the x_1-axis, A_{nm} are non-zero only for odd n. It is easy to show that, to leading orders,

$$A_{10} = \frac{Va^3}{2(1-\beta)}, \quad A_{30} = -\frac{a^7}{48} A_{10} \partial_{1111}^4 S_1^r(0), \tag{103}$$

which upon substitution into (82), (83), and (107), yield

$$I_t = \frac{mV_i}{2}\left(\frac{1+2\beta}{1-\beta}\right) + O(\beta^{\frac{10}{3}}), \tag{104}$$

$$\tau_{11}^p = \lambda_1 \beta \frac{mV^2}{(1-\beta)^2} + O(\beta^{\frac{10}{3}}), \tag{105}$$

$$\tau_{ii}^p = -\lambda_2 \beta^{\frac{10}{3}} mV^2 + O(\beta^{\frac{13}{3}}), \tag{106}$$

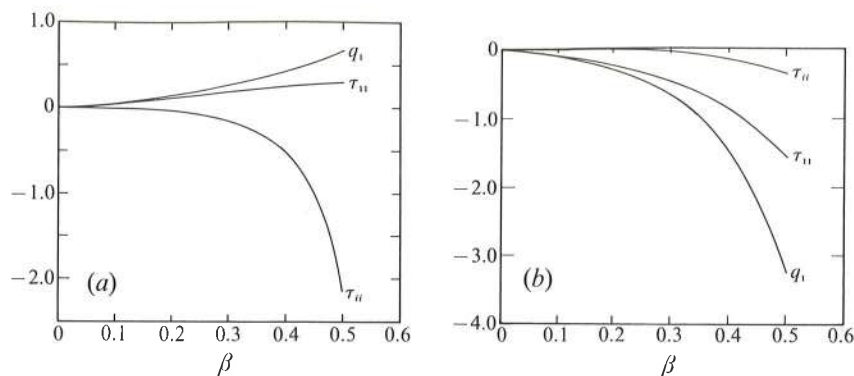$$q_1^p = \lambda_1 \beta \frac{mV^3}{(1-\beta)^3} + O(\beta^{\frac{10}{3}}), \tag{107}$$

FIGURE 1. The non-dimensional q_1, τ_{11}, and τ_{ii} for (a) the simple cubic lattice, and (b) the body-centred cubic lattice.

where $m = \rho v_{\mathrm{b}}$ and the constants λ_1 and λ_2 are given by

$$\lambda_1 = -\frac{9\mathscr{V}}{16\pi}\partial^4_{1111}S^{\mathrm{r}}_2(0), \quad \lambda_2 = \frac{5}{64}\left(\frac{3\mathscr{V}}{4\pi}\right)^{\frac{10}{3}}\partial^4_{1111}S^{\mathrm{r}}_1(0). \tag{108}$$

The constants λ_1 and λ_2 can be evaluated from Hasimoto (1959) and Sangani & Acrivos (1982) to be 0.386 and 3.66 for a simple cubic array, -1.26 and 0.364 for the body-centred array, and -1.17 and 0.212 for the face-centred cubic array. The kinetic and collisional contributions to the stress and energy flux are, of course, zero.

The above results apply when the velocity of all the bubbles is taken to be the same. This corresponds to one value of e, namely $e = mV^2(1+2\beta)/(4(1-\beta))$. In a stability analysis, one would also need to evaluate how each of these quantities varies with e near this state of equal velocities. This can be done, for instance, by calculating various quantities when the velocity of each bubble is given a small random perturbation. Since these calculations are somewhat involved, we shall not pursue them here.

The numerical results for simple and body-centred cubic arrays, obtained by including a sufficient number of multipoles are shown in figures 1(a) and 1(b), respectively. For higher values of β, up to 2^7 multipoles were needed for convergence. In these figures, τ_{11} and τ_{ii} are non-dimensionalized with mV^2, and q_1 with mV^3. The calculations apply to the case when bubbles' velocities are equal. The numerical results obtained from computer programs were checked against the asymptotic formulae presented above and perfect agreement was obtained in the limit of $\beta \to 0$. For finite β, an agreement within 2% was obtained for τ^{p}_{11} and q^{p}_1 for $\beta \leqslant 0.1$, while the computed value of τ_{ii} was about 20% higher than the asymptotic value at $\beta = 0.1$. The departure from the asymptotic values of τ_{11} and τ_{ii} increased quickly as β was increased beyond 0.1, and in fact as mentioned earlier the multipoles of as high as 2^7 order were needed to reach convergence in the numerical results.

We note that q_1 and τ_{11} are positive for the simple cubic arrangement and negative for the body-centred cubic arrangement. A positive pressure implies a repulsive potential. It is well known from the work of Biesheuvel & van Wijngaarden (1982) that a repulsive potential exists between a pair of bubbles moving vertically when their orientation makes an angle less than about 30° with the vertical direction. Thus, we expect the repulsive potential for the 11-component in a simple cubic arrangement in accordance with our numerical and asymptotic analyses. As
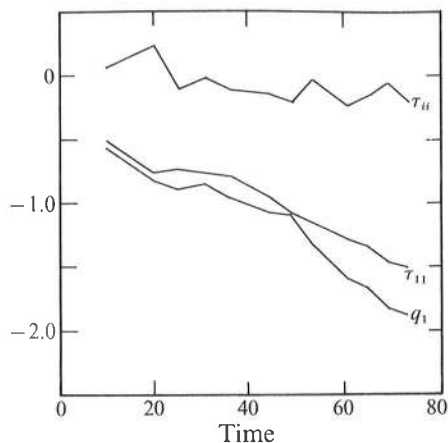
FIGURE 2. The non-dimensional q_1, τ_{11}, and τ_{ii} as a function of non-dimensional time. The initial velocity distribution is uniform, $N = 20$, $\beta = 0.1$, and $Re = 500$.

mentioned in the Introduction, a positive gradient in τ_{11} with β or V will have a stabilizing influence for the planar void fraction waves travelling in the x_1-direction. Thus, we expect the simple cubic arrangement to be stabilized and the body-centred cubic arrangement to be destabilized. On the other hand, since τ_{ii} is negative for both arrangements, these configurations are likely to be unstable to arbitrary disturbances.

4.2. *Random arrays of bubbles*

The second case we consider is that of an initially random arrangement of bubbles rising under gravity. The initial velocity of all the bubbles is V and in the x_1-direction, which is taken to be along the direction opposite to gravity. The simulations were carried out with $G = 0$, which corresponds to a constant average liquid velocity (cf. I).

First, let us discuss the comparison of various approximate formulae for determining $\dot{e}^\alpha$ (cf. (97)). We defined the relative error in estimating $\dot{e}^\alpha$ by

$$ e_r^2 = \frac{\sum (\dot{e}_c^\alpha - \dot{e}^\alpha)^2}{\sum (\dot{e}_c^\alpha)^2}, \tag{109} $$

where $\dot{e}_c^\alpha$ is the exact value as determined from numerical differentiation of the results and $\dot{e}^\alpha$ the approximate estimate obtained using different formulae in (97). For a configuration of 10 bubbles per unit cell with no viscous or gravitational forces we computed the relative errors for the three approximations. The calculations were made by including up to 2^3-multipoles. For $\beta = 0.01$, 0.1, and 0.3, the relative error e_r based on the dipole approximation was, respectively, 0.039, 0.079, and 0.16. The corresponding values for that based on velocity were 0.043, 0.12, and 0.23. Finally, the relative error based on the values of the impulse was the largest, being equal to 0.30 for $\beta = 0.3$. Thus, we conclude that the least error is made when the approximations based on the dipole (the middle equality in (97)) were used.

Figure 2 shows the results for τ_{11}, τ_{ii}, and q_1. The non-dimensionalization is the same as in the periodic array case. The Reynolds number based on the terminal speed of isolated bubbles was taken to be 500 in these calculations, and $N = 30$. We note that τ_{11} is negative, and thus the dispersed-phase pressure has a destabilizing influence. The relative contributions of the kinetic, potential, and collisional stresses
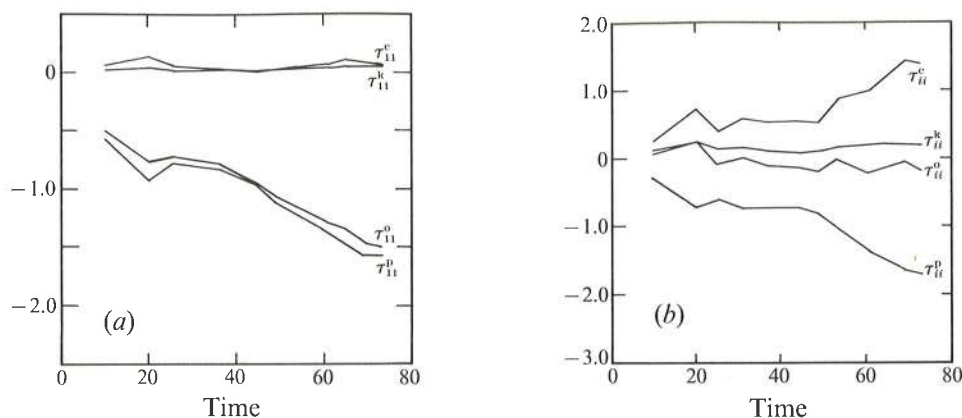
FIGURE 3. The collisional (c), kinetic (k), and potential (p) contributions to the overall (o) τ_{11} in (a) and τ_{ii} in (b) for simulation conditions described in figure 2.

to τ_{11} and τ_{ii} are shown in figures 3(a) and 3(b) respectively. We see that the magnitude of the potential contribution to τ_{11} is far greater than the translational and collisional contributions. The latter two are positive, but the overall stress component is negative. van Wijngaarden & Kapteyn (1990) ignored the collisional and potential stresses in their analysis of dilute bubbly liquids. We see that accounting for the potential contribution changes the role of dispersed-phase stress from being a stabilizing influence to a destabilizing one. Indeed, this is what we found in I: the initial uniform structure changed to one in which the bubbles formed planar aggregates. The trace of the stress tensor τ_{ii} shown in figure 3(b) is also negative but its magnitude is much smaller than that of τ_{11} indicating that τ_{22} and τ_{33} are positive. The collisional contribution to these components is generally much larger than for τ_{11} owing to the fact that most collisions among bubbles occur in the plane transverse to the mean motion. This suggests that τ_{22} and τ_{33} are positive since the collisional contributions to them are larger than the potential interaction contribution.

As seen in figure 2, τ_{11} continues to decrease with time as bubbles arrange themselves to form planar aggregates. This result at first may appear counter-intuitive since it is the negative value of the stress tensor that destabilizes the uniform spatial distribution of the bubbles, and hence the bubbles should form a structure which will reduce this tendency. Actually, it is the gradient, and not the magnitude, of τ_{11} that plays an important role in the stability analysis of the void fraction waves. The magnitude of the stress increases because bubbles get more closely packed as time progresses. Since the difference in the structures among dispersions with different values of β become small when planar aggregates form, the magnitude of the gradient of τ_{11} with β will decrease with the formation of planar aggregates.

The results shown in figures 2 and 3 were obtained by including only the dipoles in our solution. The velocity and spatial distributions of the bubbles are reasonably well approximated even with this highly truncated solution. We found, however, that the accurate evaluation of the potential stress would require higher-order multipoles in the solution. The computational time required for carrying out dynamic simulations increases rapidly with the order of multipoles included in the solution mainly due to the time required in evaluating the potential stress. Fortunately, the magnitude of the potential stress for the given spatial and velocity distribution increased with the number of multipoles retained in the solution, and

thus the conclusion reached above, i.e. the overall stress being negative, remains valid. If more accurate evaluation of the stress is required, it can be accomplished by storing the spatial and velocity configurations of the bubbles at every few hundred time steps and then evaluating the potential contribution for those configurations by including higher-order multipoles. Most of the dynamic simulation can be carried out to an adequate degree of accuracy by truncating the solution to the dipoles and quadrupoles. As mentioned in I, CPU time for a configuration of 40 bubbles per one configuration is less than 2 s on the IBM 3090 at Cornell Theory Center.

4.3. *No mean relative motion*

The third case we examined is one with zero relative motion. The gravitational and viscous forces are absent, and the initial velocity distribution is non-uniform. As in the previous case, the simulations were carried out with $G = 0$, which corresponds to a constant pressure gradient across the dispersion. As shown in I, the sum of impulses and e^{α} remain invariant for this situation. To avoid any bias due to the finite number of bubbles used in simulation toward making the dispersion anisotropic, we required that the sum of impulses be exactly zero throughout the simulation. Thus, instead of specifying the initial velocity distribution of the bubbles, we specify the impulse distribution. The three components of the impulses for the individual bubbles were chosen to be uniform random variables between -0.5 and 0.5. The initial spatial and velocity distribution determines the kinetic energy of the liquid. For this special situation, the liquid average velocity remains zero throughout the simulation, and both the average velocity of the gas phase and the average velocity of the mixture fluctuate around zero. The fluctuations for the gas-phase average velocity were less than 5%. The trace of the Reynolds stress Σ_{ii}^{R} is related to the total kinetic energy of the liquid and hence remains constant during the simulation. Therefore, the dispersed-phase stress was non-dimensionalized with Σ_{ii}^{R}. Similarly, the viscous drag coefficient to be used in the energy equation was obtained by using $V^2 = \Sigma_{ii}^{R}/m$. (For simulations, however, we chose $Re = \infty$.)

The situation described above is relevant to the flow of bubbly liquids through a pipe. In the absence of gravitational forces acting on the bubbles, the gas-phase velocity is roughly the same as that of the liquid phase. If the gradient in the average liquid velocity is small but non-zero, the mean motion it produces would introduce a non-zero variance in the velocity of the bubbles. In the limit of large Reynolds number, one would expect the root-mean-squared velocity to be much greater than the mean relative motion which produces this variance. The question of modelling the flow inside pipes is, of course, much more complex, and therefore it will be addressed in future work. Here, we shall simply give the preliminary results for the idealized situation described in the previous paragraph.

Figure 4 shows the results of dynamic simulations for three different values of β. N was chosen to equal 20 for $\beta = 0.1$ and 0.3, and 16 for $\beta = 0.5$. The results for $\beta = 0.1$ were obtained using dipoles and those for $\beta = 0.3$ and 0.5 with dipoles and quadrupoles. The kinetic component of τ_{ii} is unity for all β. This is simply the consequence of choosing the characteristic velocity based on the Reynolds stress, and this may be thought of as analogous to the ideal gas law. Interestingly, we found the viscous drag coefficient to be relatively insensitive to β, changing from about 2.0 to 2.4 as β varied from 0.1 to 0.5. The isolated bubble approximation corresponds to a drag coefficient of 2. We also find that the magnitude of the potential contribution to the stress is much smaller than the kinetic and collisional contributions, with the latter increasing rapidly with β. Because of the isotropy, we expect q_j to be zero and
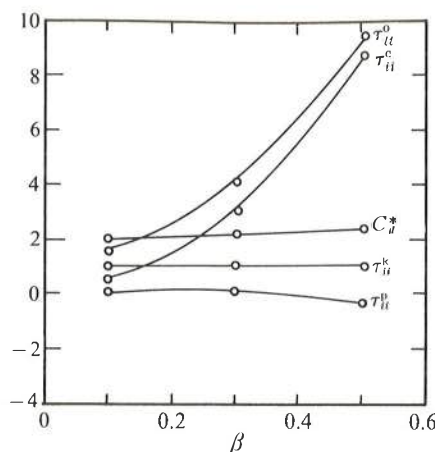
FIGURE 4. The viscous drag coefficient based on energy dissipation, and the kinetic (k), collisional (c) and potential (p) contributions to the overall (o) τ_{ii} as a function of β. The mean relative motion is zero.

hence we did not evaluate it. To save computational time, we also did not evaluate the potential contribution to τ_{11}. However, since the magnitude of the potential contribution to τ_{ii} is small, we found that the sum $\tau_1^c + \tau_{11}^k$ was reasonably close to $\frac{1}{3}\tau_{ii}$, as required by the isotropy of the dispersion.

As in the case of random arrays with mean relative motion, we found that in order to evaluate the potential contribution to the stress with greater accuracy, we need to include higher-order multipoles. Interestingly, the magnitude of the potential contribution decreased with higher-order multipoles. For example, for an initial configuration of 10 bubbles with a random velocity distribution, we found that the potential contribution changed by 50% as the highest-order multipoles retained in the solution were increased from dipoles to octupoles. However, in contrast to the case of random arrays with mean relative motion, we found that the magnitude of the potential contribution decreased with higher-order multipoles. Thus, the actual potential contribution to the total dispersed-phase stress is even smaller than is indicated in figure 4.

5. Summary

We have derived a set of equations to describe the behaviour of gas–liquid dispersions for the special case of large Reynolds number and slowly varying fields. The emphasis was on deriving expressions for various average quantities in terms of dynamic variables accessible through numerical simulations such as those described in I. For easy reference, these are listed below.

Equations for the overall medium:

$$\frac{\partial U_i}{\partial x_i} = 0, \tag{110}$$

$$\rho\left[\frac{\partial}{\partial t}(1-\beta)\,U_i^{\mathrm{L}} + \frac{\partial}{\partial x_j}(1-\beta)\,U_i^{\mathrm{L}}\,U_j^{\mathrm{L}}\right] = -\frac{\partial P}{\partial x_i} + \rho(1-\beta)\,g_i - \frac{\partial}{\partial x_j}\beta\Sigma_{ij}. \tag{111}$$

Equations for the dispersed phase:

$$\frac{\partial \beta}{\partial t} + \frac{\partial}{\partial x_j}(\beta U_j^G) = 0, \tag{112}$$

$$\frac{\partial I_i}{\partial t} + U_j^G \frac{\partial I_i}{\partial x_j} = -\frac{1}{\beta}\frac{\partial}{\partial x_j}(\beta \tau_{ij}) - 12\pi\mu a C_{d1} V_i - mg_i, \tag{113}$$

$$\frac{\partial e}{\partial t} + U_j^G \frac{\partial e}{\partial x_j} = -\frac{1}{\beta}\frac{\partial q_j}{\partial x_j} - 12\pi\mu a C_{d2} V^2 - mg_i V_i, \tag{114}$$

where U_i is the average velocity of the mixture, $V_i = U_i^G - U_i$, $(1-\beta) U_i^L = U_i - \beta U^G$, C_{d1} and C_{d2} are the viscous drag coefficients based, respectively, on the average drag force and viscous dissipation per unit volume of the mixture. The impulse is related to U_i and V_i by the added mass coefficient:

$$I_i = m[\tfrac{1}{2}C_a V_i - U_i]. \tag{115}$$

The added mass and viscous drag coefficients are functions of β and e or (e/mV^2). Similarly, the dispersed-phase tensor is defined via

$$\tau_{ij} = m[(V_i V_j - \tfrac{1}{3}V^2\delta_{ij})\,T_{11} + \tfrac{1}{3}V^2 T_{kk}\,\delta_{ij}], \tag{116}$$

where T_{11} and T_{ii} are functions of β and e.

Finally, it should be emphasized that the above expressions are correct only to the first-order spatial derivatives in β and V_i, except for those terms multiplied by the viscosity of the liquid. Since we are dealing with large Reynolds number flows, the first-order spatial derivatives in those terms have been neglected. When gradients are significant, we shall also need to include them in the constitutive relations for τ_{ij} etc. and this will introduce new quantities such as the viscosity of the dispersed phase.

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Note added in proof

When the gradients in average velocity $\mathbf{U}$ are non-zero, there will be an additional contribution to the rate of change of impulse associated with each bubble. To first order in small gradients, the contribution due to this (referred to as the lift force) is similar to the viscous and buoyancy forces, and can be expressed in terms of the rate of strain tensor and vorticity of the mean flow. Thus, $-m[(1+1/2C_E)E_{ij}V_j + 1/4C_\Omega \epsilon_{ijk}\Omega_j V_k]$ must be added to the right-hand side of (113). Here, $E_{ij} = (\partial_i U_j + \partial_j U_i)/2$, $\Omega_i = \epsilon_{ijk}\partial_j U_k$, and C_E and C_Ω are coefficients accounting for the effect of finite β. It can be shown that $C_E = C_a$ for periodic arrays. Whether this simple relation between C_a and C_E applies even to random arrays will be investigated in a future work. Note that a similar correction will apply to the dispersed phase energy equation (114) as well.