Kinetic theory of multicomponent dense mixtures of slightly inelastic spherical particles

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(Received 24 February 1995; revised manuscript received 30 May 1995)

A kinetic-type theoretical approach is developed for the transport processes involved in flows of dense mixtures of solid particles with distribution in particle size. The particles are treated as being smooth, nearly elastic, and spherical. Starting from the reduced Liouville equation, a generalized Boltzmann equation that includes the effects of inelastic collisions is stated, assuming the condition of particle chaos. The nonequilibrium velocity distribution function is derived for particles of each size using a generalized Grad moment method. The theory is applied to study the rheology of multicomponent mixtures of granular materials undergoing steady shearing flows.

PACS number(s): 05.20.Dd

I. INTRODUCTION

During the past decade, the problems of granular material flows have received increasing attention due to their importance in various engineering and geological systems, including grain traveling down an inclined chute [1], mixing of grains inside a mixer [2], rockfalls [3], sediment transport [4], and pack-ice flows [5]. By definition, gas-solid flows are considered "granular flows" if solid-body collisions govern the bulk behavior. Experiments by Bagnold [6], Savage and McKeown [7], and Savage and Sayed [8] on shear flows of concentrated suspensions of spherical particles have revealed that, at high shear rate (high Bagnold number), collisional interactions between the particles and between the particles and solid surfaces play an important role in momentum and energy transport and the interstitial fluid plays a negligible role in the flow mechanics. In recent years, attention has been directed toward developing theories [9–11] employing the kinetic theory of dense gases to obtain continuum equations for the mass, momentum, and energy of granular material flows in the grain-inertia regime, where the grains move more or less randomly as a result of collisions, much like the gas molecules in a dense gas. However, there are differences between kinetic theories for dense gases and granular materials. Unlike the kinetic energy of gas molecules, the kinetic energy of the particles is not necessarily conserved in collisions due to the inelasticity of the grains. Therefore, the kinetic theory formulations obtained assuming the reversibility of collisions must be modified to account for the energy dissipated in solid-body collisions [11,12].

Using methods from the standard Enskog theory of dense hard-sphere systems, one requires knowledge of the local equilibrium value of the radial distribution function evaluated at the point of contact between two colliding spheres to account for the increased molecular collision frequency due to the reduction of the volume in which any one of the molecules can be located [13]. In contrast to a normal gas in its equilibrium state, where there is a random thermal velocity of the molecules characterized by the gas temperature, the natural equilibrium state of a granular material is a static configuration due to the inelastic nature of particle collisions. The granular temperature, which is proportional to the mean square of the random component of solid particle velocity, can arise only from gradients in the mean flow in a flowing granular material that is subjected to external sources of energy such as shear forces. Hence, in a granular material, a condition of thermal equilibrium may be maintained by continuous shearing, in which the energy supplied to the particles is balanced by energy dissipation due to inelastic collisions. In this case, it is possible for the distribution of particles to be isotropic, but it is reasonable to expect an anisotropic distribution of collisions. In order to describe the transport properties of rapid granular flows using the Enskog theory of dense gases, Savage [14] assumed an isotropic distribution of collision angles between the colliding grains, which were taken to be smooth, slightly inelastic, spherical particles of uniform size. He suggested that the expression for the radial distribution function at contact proposed by Ogawa, Umemura, and Oshima [15] may be appropriate for a small finite system of monodisperse granular materials undergoing a shear flow. This expression predicts a drastic increase in the value of the radial distribution function as the volume fraction approaches the maximum concentration at the random closest packing. It is worth noting, however, that Ma and Ahmadi [16] indicated that the radial distribution function at contact for a system of identical spheres must diverge with a fractional critical exponent. They proposed an empirical hard-sphere equation of state by making use of a fractional power in their fitting of numerical simulations, from which one can obtain the radial distribution function at contact for a system of identical spheres, which appears to cover the entire range of concentrations as long as the transition to ordered state does not occur. Although the smooth, slightly inelastic, spherical particle model for a grain may not be entirely appropriate and though assumptions such as particle chaos and isotropic distribution of collisions throughout the flow are also simplified representations of the actual flow physics, the generalized Enskog transport theories of granular fluids as developed by Lun *et al.* [11]

1063-651X/95/52(5)/4877(15)/$06.00

have significantly advanced the rheological description of rapid granular flows [17].

Most previous theoretical studies have been limited to the case of monodisperse dense gas-solid suspensions. In reality, the solid phase normally consists of a mixture of different size particles where the particle size distribution can play a significant role in the flow mechanics. With the assumption that the singlet velocity distribution function is a dense Maxwellian, Farrell, Lun, and Savage [18] have calculated the stresses and rate of energy dissipation in a binary mixture of smooth, inelastic, spherical particles undergoing simple shear flow. They proposed an expression for the radial distribution function at contact for a binary mixture of spherical particles for the case of shearing motion in a finite-size system. In their modified form of the standard Enskog theory, the contact value of the radial distribution function was evaluated as a function of the local densities of the components at the midpoint of the line joining the centers of a colliding pair. Using an estimated value for the maximum solids fraction at closest random packing for a binary mixture, Jenkins and Mancini [19] showed that predictions of the theoretical model of [18] were in poor agreement with the computer simulations obtained by Ladd and Walton [20]. However, this disagreement can be lessened if one uses a more precise value for the maximum solids fraction at closest random packing [21]. Jenkins and Mancini [19] modified the revised Enskog theory for dense multicomponent fluid mixtures of de Haro, Cohen, and Kincaid [22] and developed a kinetic theory to predict the transport properties of binary mixtures of smooth, slightly inelastic spheres. In the revised Enskog theory the radial distribution function at contact, which is evaluated as nonlocal functionals of the mixture density fields, takes into account the spatial correlations between two spheres in a nonuniform local equilibrium state. This theory leads to diffusion forces and Onsager relations that are consistent with the laws of irreversible thermodynamics [23], while the standard Enskog theory does not [24]. Jenkins and Mancini [19] predicted the results of the numerical simulation of [20] very well using the Carnahan-Starling approximation [25] for the radial distribution function at contact, which assumes an isotropic distribution of collision angles between the colliding particles. However, in their model Eqs. (30) and (31) seem to be incorrect (see [26]). In the light of the above, more rigorous theoretical studies are clearly required of the transport properties of a multicomponent mixture consisting of an arbitrary number of smooth, slightly inelastic spheres and its polydisperse limit based on the Enskog theory. Xu and Stell [27] discussed the importance of studying polydisperse hard-sphere systems, showed the usefulness of such a theory in modeling the softness and orientational dependence of pair potentials in a monodisperse system, and calculated viscosities of a polydisperse elastic hard-sphere fluid.

In the present effort, a mathematical model for the flow of a dense mixture with distribution in size of smooth, nearly elastic spherical particles is developed. Starting from the reduced Liouville equation, a generalized Boltzmann equation that includes the effects of inelastic collisions is stated, assuming the condition of particle chaos. By using an averaging method, the continuum equations of the solid mixture in terms of the mass weighted mean values are derived, which permit quantitatively reliable simulation of the multicomponent flow of particles of different mass. The nonequilibrium velocity distribution functions are calculated using a generalized Grad moment methods [28]. It is assumed that the multicomponent dense gas-solid flow can be adequately described by consideration of the 13-moment approximation. The method presented in this study differs from that of Grad in that the diffusion velocities, the pressure deviators, and the transport pseudothermal energy flux vectors are determined by solving the approximate form of the balance laws of linear momentum, the deviant part of the mean of the second moment of velocity fluctuation, and the contracted version of the mean of third moment of velocity fluctuation, respectively. This theory is applied to study the rheology of multicomponent mixtures of granular materials undergoing steady shearing flow. The results for the shear stress of binary mixtures undergoing shear flow between parallel boundaries are found to show good agreement with computer simulations of Ladd and Walton [20]. Then, the necessary basis for predicting the separation of two different size particles by shear is discussed. Furthermore, as another example of applications of the theory, the special case of isothermal diffusion in a monosized binary mixture of differently colored particles is investigated and an analytical relation for the particle diffusivity is presented.

II. MATHEMATICAL MODEL

In this section, the flow mechanics of a dense gas-solid stream consisting of hard, smooth, nearly elastic spherical particles is considered. The solid phase includes particles of different sizes, which are assumed to behave macroscopically like interpenetrating fluids dynamically coupled through the particle-to-particle collisions. The interaction between the particles in the stream is assumed to be number-conserving contact collisions. In addition, the gaseous phase is assumed to play a negligible role in the flow mechanics. The idea is that the solid components are treated as a granular material in which momentum is primarily transferred through particle-particle collisions.

A mixture of s different-size solid particles made up of N particles, which are assumed to be hard, smooth, but nearly elastic, in a volume V may be represented by an ensemble described by the particle velocity distribution functions $f^{(N)}(x_j^N, c_j^N, t)$ in γ space. Here $f^{(N)}$ is the Nth-order distribution function. For each component of the mixture there is a distribution function that defines the expected number density of particles of kind n $(n=1,\ldots,s)$ having m^n mass in a fixed volume element dx_j^1 centered at a point x_j^1 with velocities in the range c_j^1 and $c_j^1+dc_j^1$, where dc_j^1 is a velocity element, at time t. The kinetic equation describing how this distribution function varies with time may be given as [29]

$$\frac{\partial f^{n^{(1)}}}{\partial t}+c_j^n\frac{\partial f^{n^{(1)}}}{\partial x_j^n}+F_j^n\frac{\partial f^{n^{(1)}}}{\partial c_j^n}=-\frac{1}{(N-1)!}\int\int X_j^{N,n}\frac{\partial f^{(N)}}{\partial c_j^n}dx_{\mathrm{j}}^{N-1}dp_{\mathrm{j}}^{N-1} , \quad (1)$$

where dp_j is a momentum element, F_j^n is the force per unit mass on the nth size particle due to an external field, and $X_j^{N,n}$ is the force on the nth size particle due to all other particles. The subscript j is a free index; for it the so-called summation convention is applied. However, dp_{j}^{N-1} having a roman subscript is represented as $dp_1^{N-1}dp_2^{N-1}dp_3^{N-1}$, hence the summation convention should be suspended over roman subscripts.

For a mixture having only two-body forces between the particles, Eq. (1) reduces to the Bogoliubov-Born-Green-Kirkwood-Yvon equation

$$\frac{\partial f^{n^{(1)}}}{\partial t}+c_j^n\frac{\partial f^{n^{(1)}}}{\partial x_j^n}+F_j^n\frac{\partial f^{n^{(1)}}}{\partial c_j^n}=-\int\int X_j^{n,p}\frac{\partial f^{np^{(2)}}}{\partial c_j^n}dx_{\mathrm{j}}^p dp_{\mathrm{j}}^p , \quad (2)$$

where the pair distribution function $f^{np^{(2)}}$ $(n,p=1,\ldots,s)$ defines the number density of pairs of particles such that a particle of kind n is located in the volume element dx_j^1 placed at x_j^1 with its velocity in the range dc_j^1 and c_j^1 while a particle of kind p is located in dx_j^2 at x_j^2 with its velocity in the range c_j^2 and $c_j^2+dc_j^2$.

Since the $f^{n^{(1)}}$ equation involves a higher-order distribution function it does not in itself define the behavior of $f^{n^{(1)}}$. Therefore, the higher-order distribution function must be defined in terms of distribution functions of lower order. In order to obtain an expression for the pair distribution function $f^{np^{(2)}}$ in terms of $f^{n^{(1)}}$ and $f^{p^{(1)}}$, one may apply the particle chaos approximation, which means there are no correlations between the velocities of any two particles at $t=0$ and that for the time interval between 0 and t, where $t\gg t_{\mathrm{coll}}$, recollisions of any pair of interacting particles can be neglected. Here t_{coll} is the duration of a collision. On the other hand, to incorporate the effect of the reduction of available volume in a dense mixture due to the finite size of the particles, the frequency of collisions should be increased by a factor g^{np} $(n,p=1,\ldots,s)$, which is the radial distribution function at contact for two particles, one of component n and the other of component p. In this case, the mixture radial distribution functions at contact must be taken as nonlocal functionals of the density fields of the various components in the mixture [23]. Following Andrews [30], for a nonequilibrium dense mixture of solid particles in a state of small gradient of inhomogeneity over distances of several times the separation between neighboring particles, assuming that the distribution functions do not change appreciably during the time of a collision and only binary encounters of particles possessing no internal degrees of freedom are considered, then Eq. (2) reduces to the generalized Boltzmann equation

$$\begin{aligned}\frac{df^{n^{(1)}}(x_j^n,C_j^n,t)}{dt}=&-\left[C_j^n\frac{\partial}{\partial x_j}+C_j^n\frac{\partial u_j}{\partial x_k}\frac{\partial}{\partial C_k}+\left[\frac{du_j}{dt}\frac{\partial}{\partial C_j}-F_j^n\frac{\partial}{\partial C_j}\right]\right]f^{n^{(1)}}(x_j^n,C_j^n,t)\\&+\sum_{p=1}^{s}\int\int[g^{np}(x_j,x_j+\sigma^{np}k_j|\{n_s\})f^{n^{(1)}}(x_j,c_j^{n'},t)f^{p^{(1)}}(x_j+\sigma^{np}k_j,c_j^{p'},t)\\&-g^{np}(x_j,x_j-\sigma^{np}k_j|\{n_s\})f^{n^{(1)}}(x_j,c_j^n,t)f^{p^{(1)}}(x_j-\sigma^{np}k_j,c_j^p,t)]\\&\times c_j^{np}k_j\sigma^{np^2}H(c_j^{np}k_j)dk_{\mathrm{j}}dc_{\mathrm{j}}^p . \end{aligned} \quad (3)$$

Here $d/dt=\partial/\partial t+u_j\partial/\partial x_j$ is the substantial time derivative, $\{n_s\}$ is solid component densities, $C_j^n=c_j^n-u_j$ is the peculiar velocity, $f^{n^{(1)}}(x_j^n,C_j^n,t)=f^{n^{(1)}}(x_j^n,c_j^n,t)$ is the single-particle velocity distribution function of kind n, u_j is the mass-average velocity of the solid mixture, $c_j^{np}=c_j^n-c_j^p$ is the relative velocity of the particles, H is the Heaviside step function, and k_j is the unit vector directed from the center of the first particle having m^n mass at x_j to the center of the second particle of kind p at $x_j+\sigma^{np}k_j$ at the instant of the collision, when the distance of their centers is $\sigma^{np}=(\sigma^n+\sigma^p)/2$. Note that F_j^n is a function of x_j and t but not of velocity.

$c_j^{n'}$ and $c_j^{p'}$ denote the velocities of the m^n and m^p particles after collision, respectively, which are related to those before collision c_j^n and c_j^p according to

$$\begin{aligned}c_j^{n'}&=c_j^n-\frac{m^p}{m^p+m^n}(1+e^{np})(k_kc_k^{np})k_j ,\\c_j^{p'}&=c_j^p+\frac{m^n}{m^p+m^n}(1+e^{np})(k_kc_k^{np})k_j ,\end{aligned} \quad (4)$$

where e^{np} is the coefficient of restitution for a collision between two particles, one of component n and the other of component p, which is assumed to be a material constant independent of the particle impact velocity. Although the coefficient of restitution may be a function of impact velocity, a reasonable agreement can be obtained between theoretical predictions assuming a constant, velocity-independent coefficient of restitution and experimental results [31].

In Eq. (3) $f^{n^{(1)}}$ is given as a function of C_j^n rather than

c_j^n [13] and as such the meanings of $\partial/\partial t$ and $\partial/\partial x_j$ are changed, since C_j^n is now regarded as independent of x_j. The term on the left-hand side of Eq. (3) represents the time derivative of the velocity distribution function $f^{n^{(1)}}$ following the mean motion of the solid mixture. The first term on the right-hand side is called drift term, which represents the rate of change in $f^{n^{(1)}}$ due to motion of particles without collisions. The last term on the right-hand side takes into account the particle-to-particle collisions. Equation (3) for particles of the same size and density of kind n is coupled with those of the neighboring solid particles having different sizes and densities through the solid-body collision terms. The collisional operator, which lacks the symmetry, depends on the coefficients of restitution [32]; however, the standard techniques of the kinetic theory that employ symmetries in conservative collisions are used to derive Eq. (3). Due to this assumption the theory may be applied only to nearly elastic particles. Equation (3) forms the basis for the discussion of the transport properties of multicomponent dense mixtures of slightly inelastic spherical particles.

Multiplying Eq. (3) by $m^n\psi^n$, where ψ^n is any property of particles of kind n, integrating it over the instantaneous velocity c_j^n gives the Maxwell's equation of change for the particles of the nth component in terms of the mass-weighted mean values

$$\frac{d}{dt}(\rho_m^n\phi^n\overline{\psi}^n)+\rho_m^n\phi^n\frac{\partial u_j}{\partial x_j}\overline{\psi}^n$$
$$=\rho_m^n\phi^n\overline{\frac{d\psi^n}{dt}}-\frac{\partial}{\partial x_j}(\rho_m^n\phi^n\overline{C_j^n\psi^n})-\rho_m^n\phi^n\frac{du_j}{dt}\overline{\frac{\partial\psi^n}{\partial C_j}}-\rho_m^n\phi^n\frac{\partial u_j}{\partial x_i}\overline{\frac{C_i^n\partial\psi^n}{\partial C_j}}+\rho_m^n\phi^n\overline{\frac{F_j^n\partial\psi^n}{\partial C_j}}+\rho_m^n\phi^n\overline{\frac{C_j^n\partial\psi^n}{\partial x_j}}$$
$$+\sum_{p=1}^{s}\left\langle\chi^{np}(\psi^n)-\frac{\partial\Theta_j^{np}(\psi^n)}{\partial x_j}-\frac{1}{2}\int\int\int m^n k_i\frac{\partial(\psi^{n'}-\psi^n)}{\partial C_l^n}\sigma^{np^3}\frac{\partial u_l}{\partial x_i}(c_k^{np}k_k)H(c_i^{np}k_i)f^{p^{(1)}}(x_j,c_j^p,t)f^{n^{(1)}}(x_j,c_j^n,t)\right.$$
$$\left.\times g_c^{np}\left[1+\frac{\sigma^{np}}{2}k_j\frac{\partial}{\partial x_j}\left[\ln\frac{f^{p^{(1)}}(x_j,c_j^p,t)}{f^{n^{(1)}}(x_j,c_j^n,t)}\right]+\cdots\right]dk_j dc_j^n dc_j^p\right\rangle . \tag{5}$$

Here ρ_m^n and ϕ^n are the material density and volume fraction of the particles of kind n, respectively, and g_c^{np} is the contact value of the equilibrium radial distribution function evaluated at the density n. The expression for the radial distribution function in the revised Enskog theory has been presented by van Beijeren and Ernst [33]. The mass weighted mean value of a property of m^n particles is defined by

$$\overline{\psi}^n(x_j,t)=\frac{\int m^n f^{n^{(1)}}\psi^n dc_j^n}{\int m^n f^{n^{(1)}}dc_j^n} . \tag{6}$$

Following Jenkins and Richman [12], the particle-particle collisional flux Θ_j^{np} and the source-like χ^{np}, appearing in Eq. (5), can be written as

$$\chi^{np}(\psi)=\frac{1}{2}\int\int\int(m^n\psi^{n'}+m^p\psi^{p'}-m^n\psi^n-m^p\psi^p)\sigma^{np^2}(c_l^{np}k_l)H(c_i^{np}k_i)g_c^{np}f^{p^{(1)}}(x_j,c_j^p,t)$$
$$\times f^{n^{(1)}}(x_j,c_j^n,t)\left[1+\frac{\sigma^{np}}{2}k_s\frac{\partial}{\partial x_s}\left[\ln\frac{f^{p^{(1)}}(x_j,c_j^p,t)}{f^{n^{(1)}}(x_j,c_j^n,t)}\right]+\cdots\right]dk_j dc_j^n dc_j^p \quad \text{for } n=p ,$$
$$\chi^{np}(\psi)=\int\int\int m^n(\psi^{n'}-\psi^n)\sigma^{np^2}(c_l^{np}k_l)H(c_i^{np}k_i)g_c^{np}f^{p^{(1)}}(x_j,c_j^p,t)f^{n^{(1)}}(x_j,c_j^n,t)$$
$$\times\left[1+\frac{\sigma^{np}}{2}k_s\frac{\partial}{\partial x_s}\left[\ln\frac{f^{p^{(1)}}(x_j,c_j^p,t)}{f^{n^{(1)}}(x_j,c_j^n,t)}\right]+\cdots\right]dk_j dc_j^n dc_j^p \quad \text{for } n\neq p , \tag{7}$$
$$\Theta_j^{np}(\psi)=-\frac{1}{2}\int\int\int m^n(\psi^{n'}-\psi^n)k_j\sigma^{np^3}(c_l^{np}k_l)H(c_i^{np}k_i)g_c^{np}f^{p^{(1)}}(x_j,c_j^p,t)f^{n^{(1)}}(x_j,c_j^n,t)$$
$$\times\left[1+\frac{\sigma^{np}}{2}k_s\frac{\partial}{\partial x_s}\left[\ln\frac{f^{p^{(1)}}(x_j,c_j^p,t)}{f^{n^{(1)}}(x_j,c_j^n,t)}\right]+\cdots\right]dk_j dc_j^n dc_j^p .$$

Jenkins and Mancini [19,34] reported a different expression for the particle-particle collisional flux. Therefore, a different expression for the flux of the pressure tensor than that given in the subsequent analysis can be derived from their Eq. (31). Since the pressure deviators are symmetric tensors, it can be seen from Eq. (11) in the present study that the flux of the pressure tensor may be assumed to be symmetric only with respect to the first two indices. Flux of the pressure tensor is important if one uses the 13-field theory to derive an expression for the phase speed. It is shown that for high frequencies the 13-field theory provides a finite phase speed while that provided by the five-field theory becomes infinite as the coefficient of restitution approaches unity [35]. The latter is known as the paradox of heat conduction.

The hydrodynamic equations of change—the balance equations of mass, momentum, kinetic energy, the mean of the second moment of velocity fluctuation, and the mean of the third moment (contracted version) of velocity fluctuation, which characterize the 13-moment theory of Grad [28]—may be derived by taking ψ^n, in Eq. (5), to be 1, C_j^n, C^{n^2}, $C_j^n C_q^n$, and $C_j^n C^{n^2}$, respectively. The balance of mass is given as follows:

$$\frac{d\rho^n}{dt}+\rho^n\frac{\partial u_j}{\partial x_j}+\frac{\partial}{\partial x_j}(\rho^n\overline{C_j^n})=0 , \tag{8}$$

where $\rho^n=\rho_m^n\phi^n$ is the apparent density of particles of the nth component; the balance of linear momentum is given by

$$\rho^n\frac{d\overline{C_j^n}}{dt}-\overline{C_j^n}\frac{\partial}{\partial x_l}(\rho^n\overline{C_l^n}) = -\frac{\partial P_{jq}^n}{\partial x_q}+\rho^n\overline{F_j^n}-\rho^n\frac{du_j}{dt}-\rho^n\frac{\partial u_j}{\partial x_l}\overline{C_l^n}+\sum_{p=1}^{s}\chi^{np}(C_j^n) , \tag{9}$$

where $P_{ij}^n=\rho^n\overline{C_i^nC_j^n}+\sum_{p=1}^{s}\theta_i^{np}(C_j^n)$ is the macroscopic pressure tensor of particles of kind n; the balance of kinetic energy (pseudothermal energy) can be written as

$$\frac{3}{2}\left[n^n\frac{dT^n}{dt}-T^n\frac{\partial}{\partial x_j}(n^n\overline{C_j^n})\right] = -\frac{\partial q_j^n}{\partial x_j}-\gamma^n-P_{jq}^n\frac{\partial u_q}{\partial x_j}+\rho^n\overline{F_j^nC_j^n}-\rho^n\overline{C_j^n}\frac{du_j}{dt} , \tag{10}$$

where $T^n=\frac{1}{3}m^n\overline{C^{n^2}}$ is the granular temperature of particles of kind n, which varies from the mixture temperature T by θ^n, $q_i^n=\frac{1}{2}[\rho^n\overline{C^{n^2}C_i^n}+\sum_{p=1}^{s}\theta_i^{np}(C^{n^2})]$ is the energy flux vector of the nth component and $\gamma^n=-\frac{1}{2}\sum_{p=1}^{s}\chi^{np}(C^{n^2})$ is the rate of energy dissipation per unit volume of the mixture due to inelastic nature of the collisions. It is shown in the subsequent analysis that γ^n is proportional to $(1-e^{np})$. Here the attention is restricted to such situations that the differences between component granular temperatures and the mixture temperature are small.

The balance of the mean of the second moment of velocity fluctuation is given as follows:

$$\rho^n\frac{d}{dt}(\overline{C_i^nC_q^n})-\overline{C_i^nC_q^n}\frac{\partial}{\partial x_l}(\rho^n\overline{C_l^n}) = -\frac{\partial q_{iql}^n}{\partial x_l}+\sum_{p=1}^{s}\chi^{np}(C_i^nC_q^n)-P_{lq}^n\frac{\partial u_i}{\partial x_l}-P_{li}^n\frac{\partial u_q}{\partial x_l} +\rho^n\overline{F_i^nC_q^n}+\rho^n\overline{F_q^nC_i^n}-\rho^n\frac{du_q}{dt}\overline{C_i^n}-\rho^n\frac{du_i}{dt}\overline{C_q^n} , \tag{11}$$

where $q_{liq}^n=[\rho^n\overline{C_l^nC_i^nC_q^n}+\sum_{p=1}^{s}\theta_l^{np}(C_i^nC_q^n)]$ is the flux of the pressure tensor. The balance law for the pressure deviators $a_{\langle ij\rangle}^n$ can be derived using the kinetic energy equation (10) to eliminate dT^n/dt from Eq. (11). The balance of the mean of the third moment of velocity fluctuation (contracted version) is given by

$$\rho^n\frac{d}{dt}(\overline{C_j^nC^{n^2}})-\overline{C_j^nC^{n^2}}\frac{\partial}{\partial x_l}(\rho^n\overline{C_l^n}) = -\frac{\partial q_{jl}^n}{\partial x_l}+\sum_{p=1}^{s}\chi^{np}(C_j^nC_q^nC_q^n)-q_{rqq}^n\frac{\partial u_j}{\partial x_r} -2q_{rjq}^n\frac{\partial u_q}{\partial x_r}+\rho^n\overline{F_j^nC^{n^2}}+2\rho^n\overline{F_q^nC_j^nC_q^n} -\rho^n\frac{du_j}{dt}\overline{C^{n^2}}-2\rho^n\frac{du_q}{dt}\overline{C_j^nC_q^n} , \tag{12}$$

where $q_{li}^n=[\rho^n\overline{C_l^nC_i^nC^{n^2}}+\sum_{p=1}^{s}\theta_l^{np}(C_i^nC^{n^2})]$ is the flux of the energy flux.

For dense mixtures of solid particles the transport mechanism for diffusion is kinetic. However, energy and momentum flows are outcomes of two distinct causes: translational motion of particles and the solid-body interactions. The first terms on the right-hand side of the expressions given above for the pressure tensor, energy flux vector, flux of pressure tensor, and flux of the energy flux represent kinetic fluxes, which arise from the translational motion of particles of the nth component, and the second terms are the potential fluxes, which arise from particle-particle collisions.

III. CONSTITUTIVE EQUATIONS

In order to evaluate the constitutive quantities such as pressure tensor P_{jq}^n, energy flux q_{jqq}^n, rate of energy dissipation per unit volume of the mixture γ^n, flux of the pressure tensor q_{ljp}^n, flux of the energy flux q_{lj}^n, and sourcelike terms $\chi^{np}(C_j)$, $\chi^{np}(C_{jq})$, and $\chi^{np}(C_{jqq})$, it is necessary to determine the nonequilibrium distribution function $f^{n^{(1)}}$ for particles of kind n ($n=1,\ldots,s$). If the state of the solid mixture is near equilibrium, the linearized form of the one-particle distribution function $f^{n^{(1)}}$ about the local Maxwellian distribution corresponding to local macroscopic properties of particles of kind n may be developed following the moment method of Grad [28]

$$f^{n^{(1)}}(x_j,C_j,t)=n^n\left[\frac{m^n}{2\pi T}\right]^{3/2}\exp\left[-\frac{m^nC^{n^2}}{2T}\right]\left[1+\frac{m^nv_i^n}{T}C_i^n+\frac{3}{2}\left[-\frac{\theta^n}{T}+\frac{m^n}{3T^2}\theta^nC^{n^2}\right]+\frac{a^n_{\langle ij\rangle}}{2}\left[\frac{m^n}{T}\right]^2C_i^nC_j^n+\frac{a^n_{imm}}{10}\left[\frac{m^n}{T}\right]^2\left[\frac{m^n}{T}C^{n^2}-5\right]C_i^n\right], \tag{13}$$

where $T=\sum_{n=1}^{s}n^nT^n/\sum_{n=1}^{s}n^n$ is the granular temperature of the multicomponent mixture, $v_i^n=\overline{C_i^n}$ are the diffusion velocities, $a^n_{\langle ij\rangle}=\overline{C_i^nC_j^n}-(T/m^n)\delta_{ij}$ are the pressure deviators, and $a^n_{imm}=\overline{C_i^nC^{n^2}}-5(T/m^n)v_i^n$ are the transport pseudothermal energy flux vectors. Assuming equal granular temperatures for all components represents the simplest case, and it would seem necessary to understand this case first before attempting more complex energy transfer problems involving mixtures of granular materials in which the component temperatures are different from the mixture temperature [35]. Hence, in this study the effects of the particle granular temperature perturbations θ^n are not considered.

The constitutive quantities mentioned above can be calculated in terms of 13-field quantities ρ^n, v_i^n, T, $a^n_{\langle ij\rangle}$, and a^n_{imm} by substituting the one-particle distribution function (13) into expressions for pressure tensor, energy flux, rate of energy dissipation per unit volume of the mixture, flux of the pressure tensor, flux of the energy flux given in Sec. II as well as appropriately truncated expressions for collisional source and flux. The series for collisional source and flux (7) are truncated such that they are valid when the spatial gradients of the field quantities are small. Neglecting products of diffusion velocities, pressure deviators, and energy flux vectors with those terms involving the spatial gradient, integrations for the collisional source and flux terms appearing in expressions for pressure tensor, energy flux, rate of energy dissipation per unit volume of mixture, flux of the pressure tensor, and flux of the energy flux can be carried out. To shorten the calculations, the collision operators were obtained using only the terms of the two lowest orders in Eq. (7). The results of integrations are given in the Appendix.

By substituting the calculated values of the quantities P_{ij}^n, q_{iqq}^n, γ^n, q_{liq}^n, $\chi^{np}(C_iC_q)$, $\chi^{np}(C_i)$, and $\chi^{np}(C_iC_qC_q)$ into balance equations (8)–(12), a system of field equations for scalar fields of ρ^n, v_i^n, T, $a^n_{\langle ij\rangle}$, and a^n_{imm} can be obtained. Here the work is directed toward finding approximate expressions for $a^n_{\langle ij\rangle}$, a^n_{iqq}, and v_i^n as functions of gross condition of the mixture of particles using scaling argument. To this end a term-by-term order of magnitude analysis of the conservation equation was carried out to find the dominant terms. Taking L, t_0, and $T_0^{1/2}$, respectively, as a characteristic length, time, and particle fluctuating velocity, five dimensionless quantities $r_1^n=\sigma^n/L$, $r_2^n=\sigma^n/t_0T_0^{1/2}$, $r_3^n=v_i^n/T_0^{1/2}$, $r_4^n=a^n_{\langle iq\rangle}/T_0$, and $r_5^n=a^n_{iqq}/T_0^{3/2}$ that are useful for the scaling can be defined.

When the spatial gradients of the mean fields are small, in the balance law for the deviant part of the mean of the second moment of velocity fluctuation, all quantities r_i^n ($n=1,2,\ldots,s$; $i=1,2,\ldots,5$) are supposed of the same order of magnitude and small, then, in this approximation pressure deviators may be taken to be independent of diffusion velocities and transport pseudothermal energy flux vectors satisfying

$$\sum_{p=1}^{s}\sigma^{np^2}(1+e^{np})g_c^{np}n^p\rho^n\left[2\pi M^{pn}\frac{T}{m^n}\right]^{1/2}\times\langle\{\tfrac{4}{3}M^{np}+\tfrac{2}{5}M^{pn}[2+(1-e^{np})]\}a^n_{\langle iq\rangle}+\{\tfrac{2}{5}[2+(1-e^{np})]-\tfrac{4}{3}\}M^{pn}a^p_{\langle iq\rangle}\rangle$$
$$=-\rho^n\frac{T}{m^n}\left\langle 1+\sum_{p=1}^{s}\pi\sigma^{np^3}(1+e^{np})g_c^{np}n^p\{\tfrac{1}{3}(M^{pn}-M^{np})-\tfrac{2}{5}M^{pn}[1+(1-e^{np})]+\tfrac{1}{3}\}\right\rangle\frac{\overline{\overline{\overset{\circ}{\partial u_i}}}}{\partial x_q},\quad n=1,2,\ldots,s, \tag{14}$$

where $\overline{\overline{\overset{\circ}{\partial u_i}}}/\partial x_q$ is nondivergent symmetric part of the velocity-gradient tensor. It is worth noting that Eq. (14) reduces for a monodisperse solid phase to the expression obtained by Lun *et al.* [11] for the deviant part of the transport pressure tensor. This agreement is a valuable check for the present calculations.

Because of the coupling between the conservation of linear momentum (9) and the balance of the mean of the third moment of velocity fluctuation (12) through the sourcelike terms, the evaluation of the diffusion velocities and pseudothermal energy flux vectors is more involved. By retaining only the dominant terms in Eq. (9) one can get

$$\sum_{p=1}^{s}\sigma^{np^2}(1+e^{np})g_c^{np}\rho^n\frac{n^p}{n}\times\left[\frac{4}{3}\left[2\pi M^{pn}\frac{T}{m^n}\right]^{1/2}(v_i^n-v_i^p)-\frac{2}{15}\left[2\pi M^{pn^3}\frac{m^n}{T}\right]^{1/2}(a^p_{iqq}-a^n_{iqq})\right]=-d_i^n, \tag{15}$$

where n is the solid mixture number density and d_i^n is the standard diffusion force defined by

$$d_i^n=\frac{1}{n}\frac{T}{m^n}\left[\rho^n\left[1+\sum_{p=1}^{s}\frac{2\pi}{3}M^{np}(1+e^{np})n^p\sigma^{nj^3}g_c^{np}\right]\frac{\partial \ln T/m^n}{\partial x_i}+\sum_{p=1}^{s}\frac{\rho^n}{T}\left[\frac{\partial\mu^n}{\partial n^p}\right]_{T,n^h\neq n^p}\frac{\partial n^p}{\partial x_i}\right.$$
$$\left.-\frac{\rho^n}{\rho}\frac{m^n}{T}\sum_{p=1}^{s}\rho^p(\overline{F_i^n}-\overline{F_i^p})-\frac{\rho^n}{\rho}\frac{m^n}{T}\frac{\partial P_{iq}}{\partial x_q}\right]. \quad (16)$$

Here $P_{iq}=\sum_{p=1}^{s}P_{iq}^p$ is the total solid pressure, $\rho=\sum_{p=1}^{s}\rho^n$ is the mixture density, and μ^n is the chemical potential for particles of kind n.

When the step above is carried out on Eq. (12) the following expression is obtained:

$$\sum_{p=1}^{s}\frac{1}{5}\sigma^{np^2}g_c^{np}\frac{n^p}{n}\rho^n(1+e^{np})\Bigg\langle-\frac{20}{3}(1-e^{np})\left[2\pi M^{pn}\left[\frac{T}{m^n}\right]^3\right]^{1/2}(M^{pn}v_i^p+M^{np}v_i^n)$$
$$-\left[2\pi M^{pn}\left[\frac{T}{m^n}\right]^3\right]^{1/2}(v_i^n-v_i^p)[8-\tfrac{4}{3}M^{np}+8(1-e^{np})M^{pn}-\tfrac{16}{3}(1-e^{np^2})M^{pn}]$$
$$+\left[2\pi M^{pn}\frac{T}{m^n}\right]^{1/2}\{\tfrac{2}{3}M^{pn}M^{np}(a_{iqq}^p-a_{iqq}^n)+4(M^{pn^2}a_{iqq}^p-M^{np^2}a_{iqq}^n)+\tfrac{12}{5}M^{pn^2}(a_{iqq}^p-a_{iqq}^n)$$
$$-\tfrac{82}{15}(1-e^{np})M^{np}(M^{pn}a_{iqq}^p+M^{np}a_{iqq}^n)-\tfrac{16}{15}(1+e^{np})M^{pn}(a_{iqq}^pM^{pn}+a_{iqq}^nM^{np})+\tfrac{4}{5}(1-e^{np})(1-2e^{np})M^{pn^2}(a_{iqq}^p-a_{iqq}^n)\Bigg\rangle$$
$$=\left[\frac{T}{m^n}\right]d_i^n+\left[\frac{T}{m^n}\right]^2\Bigg\langle\frac{\rho^n}{n}\left[1+\sum_{p=1}^{s}\pi\sigma^{np^3}(1+e^{np})g_c^{np}n^pM^{pn}M^{np}[\tfrac{4}{5}-\tfrac{2}{5}(1-e^{np})-\tfrac{4}{5}(1-e^{np^2})]\right]$$
$$\times\frac{\partial \ln T/m^n}{\partial x_i}+\sum_{p=1}^{s}\frac{3}{5}M^{pn}\frac{m^n}{n}e^{np}(e^{np}-1)\left[\frac{n^n}{T}\left[\frac{\partial\mu^n}{\partial n^p}\right]_{T,n^h\neq n^p}-\delta_{np}\right]\frac{\partial n^p}{\partial x_i}\Bigg\rangle . \quad (17)$$

Here the external force per unit mass F_i^n is assumed to be a function of x_i only but not of C_i^n. Equations (15) and (17), along with Eqs. (16), form a set of equations for particle diffusion velocities and energy flux vectors. It should be noted that the diffusion forces d_i^n ($n=1,2,\ldots,s$) are not all independent, but satisfy the relation

$$\sum_{n=1}^{s}d_i^n=0 . \quad (18)$$

It may be worthwhile to point out that when $\sigma^n u_0/LT_0^{1/2}\ll 1$ ($n=1,2,\ldots,s$), the granular flow is such that the properties of a small element of solid mixture do not change appreciably in a mean time between collisions. In such a circumstance, the deviant part of the transport pressure tensor, transport pseudothermal energy flux vectors, and diffusion velocities approach values given by Eqs. (14), (15), and (17) within a few collision times.

Finally, the nonequilibrium distribution function for particles of kind n of a dense mixture with distribution in size of smooth, slightly inelastic spherical particles in terms of the mixture granular temperature, density, mixture mean mass velocity, and their derivatives with respect to place can be found by placing the expressions for diffusion velocities, pressure deviators, and energy flux vectors into Eq. (13).

All the results represented thus far are for multicomponent systems. For binary mixtures these formulas may be considerably simplified. In the subsequent section, the special case of an isothermal dense binary mixture of spherical particles that is undergoing a simple shear flow will be considered as an application of this theory and the necessary basis for predicting the separation of two different size particles by shear is discussed. Moreover, an analytical relation for the particle diffusivity is presented when there are no external forces acting on the mixture of monosized differently colored particles and mechanical equilibrium has been attained.

IV. BINARY MIXTURES OF FRICTIONLESS SPHERES

A simple solution of the balance of energy for a binary mixture is obtained when the granular material is undergoing a steady, simple shear flow with a constant granular temperature. Consider a binary mixture consists of two solid components with masses m^1 and m^2, diameters σ^1 and σ^2, and number densities n^1 and n^2, respectively, undergoing a shearing motion at a constant and high rate of strain $\dot{\epsilon}$. It is assumed that the particles are nearly elastic and all with the same coefficient of restitution **e** for normal collisions. In the absence of external forces and gradient of number densities, by use of Eq. (10) the energy balance for the mixture at thermal equilibrium, where the granular temperature of both components are equal, can be written as

$$0=\sum_{n=1}^{2}\left[\rho^n a^n_{\langle xy\rangle}+\sum_{p=1}^{2}\frac{2\pi}{15}(1+e^{np})\rho^n M^{pn}\times n^p \sigma^{np^3} g_c^{np}(a^n_{\langle xy\rangle}+a^p_{\langle xy\rangle})\right]\dot{\epsilon}+\sum_{n=1}^{2}\sum_{p=1}^{2}2\rho^n\sigma^{np^2}(1-e^{np^2})g_c^{np}n^p\frac{T}{m^n}\times\left[2\pi M^{pn}\left[\frac{T}{m^n}\right]\right]^{1/2}, \tag{19}$$

where $\rho^n a^n_{\langle xy\rangle}$ is the component of the force acting on particles of kind n in the direction of flow on a unit area of surface perpendicular to the direction of shear gradient. This may be interpreted as a statement that pseudothermal energy generated due to work done by the pressure in a region of the mixture moving with the mean mixture velocity should be locally dissipated by way of inelastic particle-particle collisions so as to maintain a constant granular temperature.

By placing the expressions for the pressure deviators $a^1_{\langle xy\rangle}$ and $a^2_{\langle xy\rangle}$ resulting from Eq. (14) into Eq. (19) the mixture granular temperature can be expressed in terms of the number densities, strain rate, and the particle properties. Before the shear stress for a binary mixture of frictionless spheres can be evaluated, an expression for the mixture shear stress is needed. The non-dimensional shear stress may be given by

$$P^*_{xy}=\sum_{n=1}^{2}\left[\frac{\rho^n}{\rho^*}\frac{a^n_{\langle xy\rangle}}{\sigma^{1^2}\dot{\epsilon}^2}+\sum_{p=1}^{2}\frac{2\pi}{15}(1+e^{np})\frac{\rho^n}{\rho^*}M^{pn}n^p\frac{\sigma^{np^3}}{\sigma^{1^2}}g_c^{np}\left[\frac{a^n_{\langle xy\rangle}}{\dot{\epsilon}^2}+\frac{a^p_{\langle xy\rangle}}{\dot{\epsilon}^2}\right]\right], \tag{20}$$

where ρ^* is the particle density given by Walton and Braun [36]. Equation (20) has been written in a form that allows easy comparison with the results of computer simulations by Ladd and Walton [20]. The first term on the right-hand side can be interpreted as the nondimensional kinetic flux of momentum and the second term represents an extra contribution due to particle-particle collisions. Now by substituting the results for the mixture granular temperature as described above into Eq. (20) the nondimensional mixture shear stress can be evaluated, although the expression is complicated. The nondimensional shear stresses $-P^*_{xy}$ are calculated using Eq. (20) for two different volume fraction ratios $\phi_1/\phi_2=0.5$ and 2 and two different coefficient of restitution $e=0.8$ and 0.95 with size ratios from 1 to 3.5. The total solid volume fraction is taken to be 0.5. Since there is no exact, explicit equation for g_c^{np} in terms of the set of particle diameters $\{\sigma_s\}$ and particle number densities $\{n_s\}$ available, the Carnahan-Starling expression is used, which is accurate in the range of volume fraction used in Ladd-Walton computer simulations and is based on the assumption of an isotropic distribution of collision angles between the colliding particles as proposed by Mansoori *et al.* [25] for mixtures of hard-sphere fluids. However, their expression gives values of the radial distribution function that are too low at high mixture volume fractions, especially as the system approaches the random close packing density. The Carnahan-Starling approximation is given by

$$g^{np}=\sum_{i^*=1}^{3}\frac{a_{i^*}}{1-\xi_3}\left[\frac{\xi_2\sigma^n\sigma^p}{(1-\xi_3)\sigma^{np}}\right]^{i^*-1},\qquad \xi_l=\frac{\pi}{6}\sum_{k=1}^{s}n^k\sigma^{k^l},\quad a_1=1,\quad a_2=\tfrac{3}{2},\quad a_3=\tfrac{1}{2}, \tag{21}$$

where n^k is the m^k particle number density.

Figure 1 shows the results of the calculations for a binary mixture of smooth spheres as plots of $-P^*_{xy}$ calculated using this theory and compares these results with the computer simulations results of [20]. The agreement is good except for the large size ratios at $e=0.95$ and $\phi_1/\phi_2=2$. The computer simulations predict too rapid a decrease with increasing size ratio for the large size ratios. This may be interpreted as a tendency to dissipate the state of high pseudothermal energy of a mixture at large size ratio by reducing the work done by the pressure. The agreement with the results for the smaller volume fraction ratios is good. Of considerable significance is the observation that the predicted values of the nondimensional shear stress from the computer simulations have a strong dependence on sample size for larger size ratio for $e=0.8$ and $\phi_1/\phi_2=2$ [37]. The

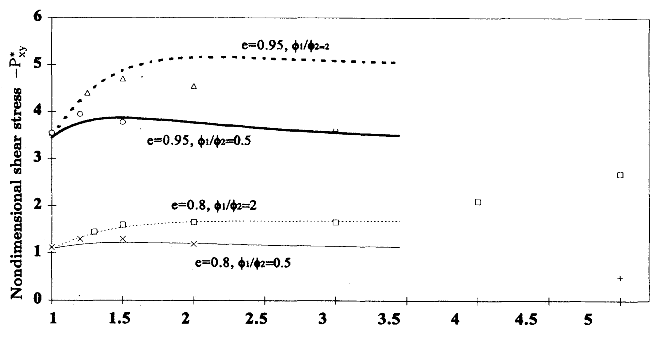

FIG. 1. Comparison of the present model calculations for the nondimensional shear stress $-P^*_{xy}$, for binary mixtures of frictionless in elastic spheres undergoing steady, simple shear flow with the computer simulations of Ladd and Walton [20]. The results of Ladd and Walton are plotted as nondimensional shear stress versus size ratio. Symbols and conditions are as follows. (1) □, large sample size; +, small sample size for $e=0.8$, $\phi_1/\phi_2=2$. (2) ×, $e=0.8$, $\phi_1/\phi_2=0.5$. (3) ○, $e=0.95$, $\phi_1/\phi_2=2$. (4) △, $e=0.95$, $\phi_1/\phi_2=0.5$. Curves are the present kinetic model predictions for constant granular temperature conditions.

kinetic theory predictions seem to be in agreement with the results calculated using a larger sample size, suggesting that the shear stresses in binary mixtures appeared to be approaching a limit at very high size ratios, which is shown in Fig. 2. However, this prediction shows somehow disagreement with the results of computer simulations, using large sample size, for the larger volume fraction at very large size ratio. If the computer simulations results are considered to be reliable, then one needs a new approach for studying this unusual behavior. The idea of particulate clusters seems to be a worthwhile idea in the study of the rheology of multicomponent dense mixtures of solid spherical particles at very large size ratio, where every kind of cluster makes a distinctive contribution to the mixture rheology. It is worth noting that the present theory, which is based on the assumptions of no three-body effects, collision time much less than the local averaging time, and small change in local properties of the mixture over distance of order of the separation between neighboring particles, is restricted to a relatively small range of size ratio.

One of the most interesting questions pertaining to binary mixture of solid particles is the question of possible separation of two species. The separation of particles with different size can occur when a nonhomogeneous binary mixture of solid particles undergoes a linear shear flow. To visualize the nature of the processes involved consider a collection of particles in a layer of thickness δy, where y is the direction normal to the plane of shear. Since the mixture is present with inhomogeneities in the concentration, the mixture viscosity is not constant in the direction normal to the plane of shear. As a consequence, shear-induced particle migrations arise from gradients in shear stress. Here the attention is focused on finding a necessary basis to investigate separation by shear in a nonhomogeneous binary mixture using the present kinetic theory. By taking Eq. (9) into account,

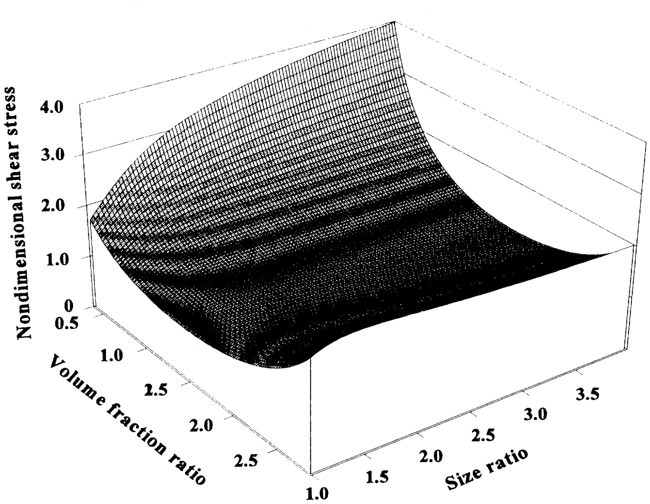

FIG. 2. A $(-P^*_{xy})-(\phi_1/\phi_2)-(\sigma_1/\sigma_2)$ surface for a binary mixture of slightly inelastic spherical particles undergoing a shearing motion at constant and high rates of strain with coefficient of restitution $e=0.8$.

the balances of momentum in the direction of shear plane for particles of the nth component undergoing a shearing motion between two parallel horizontal boundaries at a constant and high rate of strain $\dot{\epsilon}$, can be given by

$$0=\frac{\partial}{\partial y}\left[\rho^n a^n_{\langle xy\rangle}+\sum_{p=1}^{2}\frac{2\pi}{15}(1+e^{np})\rho^n n^p \sigma^{np^3} g_c^{np} M^{pn}\times(a^n_{\langle xy\rangle}+a^p_{\langle xy\rangle})\right]-\rho^n\dot{\epsilon}v_y^n\ , \quad (22)$$

where v_y^n represents the shear-induced diffusion velocity of particles of kind n normal to the plane of shear. Here it is assumed that the diffusion velocity due to gradients in viscosity is controlling the particle migrations. By placing the expressions for the pressure deviators $a^1_{\langle xy\rangle}$ and $a^2_{\langle xy\rangle}$ given above in the balance of momentum of the first and the second component, expressions for shear-induced diffusion velocities in terms of particle number densities may be derived. This is a good approximation when the granular temperature is a relatively weak function of concentration. Then, by substituting those expressions in the balance of mass of the first and the second component and using Eq. (18) one can provide the necessary basis to predict the separation of two kinds of particles from one another by shear. According to Eq. (20), for monosized suspensions $a_{\langle xy\rangle}$ is proportional to $\dot{\epsilon}^2\sigma^2$; hence the shear-induced diffusion velocities of particles are proportional to $\dot{\epsilon}\sigma^2$. This is in agreement with the shear-induced self-diffusion velocity predicted by Leighton and Acrivos [38].

At this stage it is of interest to take up the question of possible separation in a homogeneous mixture undergoing a simple shear flow. Consider a mixture of solid particles of two species 1 and 2 with masses of m^1 and m^2, diameter σ^1 and σ^2, and constant densities ρ_0^1 and ρ_0^2, respectively, undergoing linear shear flow at a constant and high rate of strain $\dot{\epsilon}_0$ between two parallel planes. In this case the mixture viscosity does not depend on y, where y is the direction normal to the plane of shear. To understand the process of particle separation one needs to study the stability of that state of the mixture. A linear stability analysis yields the spectrum of small fluctuations $\rho^1(y,t)$, $\rho^2(y,t)$, and $\dot{\epsilon}(y,t)\sim\exp(i\beta y-i\omega t)$, where ω is the frequency and β is the wave number. At moderate concentrations, the result suggests that fluctuations are unstable when the shear rate $\dot{\epsilon}_0$ exceeds a threshold

$$\dot{\epsilon}^{*2}=\frac{4\rho_0^1\rho_0^2\Upsilon^1\Upsilon^2}{(\rho_0^1\Pi^1+\Upsilon^1)(\rho_0^2\Pi^2+\Upsilon^2)}\ . \quad (23)$$

The functions in Eq. (23) are defined as

$$\Upsilon^1=\frac{A_1}{B},\quad \Upsilon^2=\frac{A_2}{B},\quad \Pi^1=\frac{(C_1B-C_2A_1)}{B^3},$$

$$\Pi^2=\frac{(C_3B-C_2A_2)}{B^3}\ ,$$

$$A_1 = -\left[\frac{m^1}{m^2}\right]^{1/2} \kappa_{21}^{12} n_0^2 (1 + K_2^{21} n_0^1 + K_2^{22} n_0^2) + (1 + K_2^{11} n_0^1 + K_2^{12} n_0^2)(\kappa_{11}^{21} n_0^1 + \kappa_{11}^{22} n_0^2 + \kappa_{21}^{22} n_0^2) ,$$

$$A_2 = \left[\frac{m^1}{m^2}\right]^{1/2} (\kappa_{11}^{11} n_0^1 + \kappa_{21}^{11} n_0^1 + \kappa_{11}^{12} n_0^2) \times (1 + K_2^{21} n_0^1 + K_2^{22} n_0^2) - \kappa_{21}^{21} n_0^1 (1 + K_2^{11} n_0^1 + K_2^{12} n_0^2) ,$$

$$B = \kappa_{21}^{21} \kappa_{21}^{12} n_0^1 n_0^2 - (\kappa_{11}^{11} n_0^1 + \kappa_{21}^{11} n_0^1 + \kappa_{11}^{12} n_0^2) \times (\kappa_{11}^{21} n_0^1 + \kappa_{11}^{22} n_0^2 + \kappa_{21}^{22} n_0^2) ,$$

$$C_1 = -\left[\frac{m^1}{m^2}\right]^{1/2} \kappa_{21}^{12} (1 + K_2^{21} n_0^1 + 2K_2^{22} n_0^2 + K_2^{21} n_0^2) + (K_2^{11} + K_2^{12})(\kappa_{11}^{21} n_0^1 + \kappa_{11}^{22} n_0^2 + \kappa_{21}^{22} n_0^2) + (1 + K_2^{11} n_0^1 + K_2^{12} n_0^2)(\kappa_{11}^{21} + \kappa_{11}^{22} + \kappa_{21}^{22}) , \tag{24}$$

$$C_2 = \kappa_{21}^{21} \kappa_{21}^{12} n_0^2 + \kappa_{21}^{21} \kappa_{21}^{12} n_0^1 - (\kappa_{11}^{11} + \kappa_{21}^{11} + \kappa_{11}^{12})(\kappa_{11}^{21} n_0^1 + \kappa_{11}^{22} n_0^2 + \kappa_{21}^{22} n_0^2) - (\kappa_{11}^{11} n_0^1 + \kappa_{21}^{11} n_0^1 + \kappa_{11}^{12} n_0^2)(\kappa_{11}^{21} + \kappa_{11}^{22} + \kappa_{21}^{22}) ,$$

$$C_3 = \left[\frac{m^1}{m^2}\right]^{1/2} [(\kappa_{11}^{11} + \kappa_{21}^{11} + \kappa_{11}^{12})(1 + K_2^{21} n_0^1 + K_2^{22} n_0^2) + (\kappa_{11}^{11} n_0^1 + \kappa_{21}^{11} n_0^1 + \kappa_{11}^{12} n_0^2)(K_2^{21} + K_2^{22})] - \kappa_{21}^{21} (1 + 2K_2^{11} n_0^1 + K_2^{12} n_0^2 + K_2^{12} n_0^1) ,$$

$$K_1^{ij} = \sigma^{ij^2} (1 + e^{ij}) g_c^{ij} (2\pi M^{ij})^{1/2} ,$$

$$K_1^{ij} = \pi \sigma^{ij^3} (1 + e^{ij}) g_c^{ij} \{ \tfrac{1}{3}(M^{ji} - M^{ij}) - \tfrac{2}{5} M^{ji} [1 + (1 - e^{ij})] + \tfrac{1}{3} \} ,$$

$$X_1^{ij} = \tfrac{4}{3} M^{ij} + \tfrac{2}{5} M^{ij} [2 + (1 - e^{ij})] ,$$

$$X_2^{ij} = \{ \tfrac{2}{5} [2 + (1 - e^{ij})] - \tfrac{4}{3} \} M^{ij} ,$$

$$\kappa_{pq}^{ij} = X_p^{ij} K_q^{ij} ,$$

where n_0^1 and n_0^2 are number densities of particles of kind 1 and 2, respectively. Here the necessary basis for predicting the separation of two kinds of particles from one another by shear is discussed. An investigation on the stability of a sheared binary mixture of slightly inelastic spherical particles is of interest because it can be applied to the study of stress fluctuations observed in shear cells [35].

As another example of applications of the theory, the isothermal diffusion in a monosized binary mixture of differently colored particles is considered. The focus of this section is in examining the mixing that occurs during a flow of relatively dense monosized solid particles. Consider a binary mixture consists of two differently colored solid components with the same diameter σ and mass m, but different number densities n^1 and n^2 undergoing an isothermal motion between two parallel walls with negligible shearing. According to Eq. (10), in order to maintain a constant granular temperature, energy must be added to the mixture at a rate γ. Assuming that there are no external forces acting on the mixture and that mechanical equilibrium has been initiated, $\bar{F}_i^n = 0$ and $\partial P_{iq}/\partial x_q = 0$. Using Eq. (16) for d_i^n in Eq. (15), the balance of momentum for particles of kind n $(n = 1,2)$ in the direction perpendicular to the direction of main flow may be given as

$$\sum_{p=1}^{2} \sigma^{np^2} (1 + e^{np}) g_c^{np} \rho^n \frac{n^p}{n} \left[\frac{4}{3} \left[2\pi M^{pn} \frac{T}{m^n} \right]^{1/2} (v_y^n - v_y^p) - \frac{2}{15} \left[2\pi M^{pn^3} \frac{m^n}{T} \right]^{1/2} (a_{yqq}^p - a_{yqq}^n) \right] = -\frac{1}{n} \frac{T}{m^n} \left[\sum_{p=1}^{2} \frac{\rho^n}{T} \left[\frac{\partial \mu^n}{\partial n^p} \right]_{T, n^h \neq n^p} \frac{\partial n^p}{\partial y} \right] . \tag{25}$$

The chemical potential for particles of kind n can be obtained from Eq. (21) for the radial distribution function [39]

$$\frac{\mu^n}{T} = \ln n^n - \ln(1 - \xi_3) + \sigma^{n^3} \left[\frac{\xi_0}{1 - \xi_3} + \frac{3\xi_1 \xi_2}{(1 - \xi_3)^2} + \frac{3\xi_2^3}{(1 - \xi_3)^3} - \frac{\xi_3 \xi_2^3}{(1 - \xi_3)^3} \right] + \frac{3\xi_2 \sigma^n}{1 - \xi_3} + \frac{3\xi_1 \sigma^{n^2}}{1 - \xi_3} + \frac{9\xi_2^2 \sigma^{n^2}}{2(1 - \xi_3)^2} + 3 \left[\frac{\xi_2 \sigma^n}{\xi_3} \right]^2 \left[\ln(1 - \xi_3) + \frac{\xi_3}{1 - \xi_3} - \frac{\xi_3^2}{2(1 - \xi_3)^2} \right] - \left[\frac{\xi_2 \sigma^n}{\xi_3} \right]^3 \left[2 \ln(1 - \xi_3) + \frac{\xi_3 (2 - \xi_3)}{1 - \xi_3} \right] . \tag{26}$$

The first term on the right-hand side of Eq. (26) represents the free energy of the particles of kind n when approaching ideal-gas behavior. The remaining terms are the residual chemical potential. As an initial speculation, the contribution of an extra flux due to the difference between the transport pseudothermal energy flux vectors of the first and second component is neglected. Hence, Eq. (25) for particles of kind 1 and 2, with a flow without a net mass flow in the y direction, reduce to an expression for the particle self-diffusion coefficient given by Savage [40]

$$D = \frac{\sigma \pi^{1/2}}{8(1 + e)\phi g_c} \left[\frac{T}{m} \right]^{1/2} , \tag{27}$$

where D is the particle self-diffusion coefficient, g_c is the contact value of the equilibrium radial distribution function, and ϕ is solid volume fraction.

Hsiau and Hunt [41] compared their measurements for a dense rapid granular flow with the estimates for the mixing layer thickness using Eq. (27) and found that the model predictions overestimated the experimental observations by 60–300%. It is worth noting that particle diffusion in the direction normal to the shear plane in a dense granular material undergoing shear flow between rough boundaries is caused by several thermodynamic forces. However, the theoretical study of Hsiau and Hunt [41] seems to have considered only the ordinary diffusion. Hence a complete description is required of the diffusion processes involved in a sheared dense granular material based on generalized Enskog theory of granular fluids. Zamankhan and Polashenski [42] present a complete description of the problem as well as the comparison between the experimental measurements [41] and the estimates for the mixing layer thickness based on the present theory.

V. CONCLUDING REMARKS

The analysis presented above gives a theoretical foundation for the Enskog theory for dense multicomponent mixtures of slightly inelastic spherical particles. It is assumed that the level of information provided by the single-particle velocity distribution functions is adequate to describe the behavior of dense multicomponent mixtures, where the effect of more than two-body collisions may be neglected. The nonequilibrium functions for particles of each size are derived, using a generalized Grad moment method. The method presented in this study differs from Grad's in that the diffusion velocities, the pressure deviators, and the pseudothermal energy flux vectors are derived by scaling arguments. In the present theory, the diffusion forces at uniform temperature are presented as gradients of the chemical potentials.

The theory is applied to calculate the shear stresses for a binary mixture of frictionless spheres with a constant granular temperature far away from the walls using Carnahan-Starling approximation for the contract value of the equilibrium radial distribution function (21). The results show good agreement with the computer simulation results of Ladd and Walton [20], where the total solid volume fraction was $\phi_s = 0.5$, except for the large size ratios at the coefficient of restitution $e = 0.95$. The kinetic theory predictions suggest that the shear stresses in binary mixtures appeared to be approaching a limit at very high size ratio.

As another example of applications of the theory, the special case of isothermal diffusion in a monosized binary mixture of differently colored particles is investigated and an analytical relation for the flow-induced particle diffusivity is presented. Moreover, the necessary basis for predicting the separation of two kinds of particles from one another by shear is discussed.

ACKNOWLEDGMENTS

The author wishes to express his appreciation to Dr. O. Walton for sending him computer simulation results. He also thanks Merja Salminen for her support.

APPENDIX: CONSTITUTIVE QUANTITIES

Here the expressions for quantities P^n_{iq}, q^n_i, q^n_{liq}, q^n_{li}, and γ^n and sourcelike terms $\chi^{np}(C_i)$, $\chi^{np}(C_iC_q)$, and $\chi^{np}(C_iC_qC_q)$ in terms of 13-field quantities are given. The algebraic manipulations were carried out by hand. A symbolic manipulation program in MATHEMATICA [43] was developed to check the calculations. The constitutive quantities can be written, to first order in the gradients, as follows: the pressure tensor

$$P^n_{iq} = \rho^n \left[\frac{T}{m^n} + \frac{\theta^n}{m^n} \right] \delta_{iq} + \rho^n a^n_{\langle iq \rangle}$$
$$+ \sum_{p=1}^{s} (1+e^{np}) \rho^n n^p \sigma^{np^3} g_c^{np} \left\{ \frac{\pi}{3} \left[\frac{T}{m^n} + M^{pn} \left[\frac{\theta^n}{m^n} + \frac{\theta^p}{m^p} \right] \right] \delta_{iq} + \frac{2\pi}{15} M^{pn} (a^n_{\langle iq \rangle} + a^p_{\langle iq \rangle}) \right.$$
$$\left. - \frac{4}{15} \sigma^{np} \left[2\pi M^{pn} \frac{T}{m^n} \right]^{1/2} \left[\overline{\overline{\frac{\partial u_i}{\partial x_q}}}^{\circ} + \frac{5}{6} \frac{\partial u_r}{\partial x_r} \delta_{iq} \right] \right\}, \tag{A1}$$

where $M^{jn} = m^j/(m^j + m^n)$ and δ_{iq} is the Kronecker delta; the energy flux

$$q_i^n=\frac{5}{2}\rho^n\left[\frac{T}{m^n}v_i^n+\frac{1}{5}a_{iqq}^n\right]$$
$$+\sum_{p=1}^{s}\sigma^{np^3}g_c^{np}(1+e^{np})\Bigg\langle-\sigma^{np}\left[2\pi M^{pn}\left[\frac{T}{m^n}\right]\right]^{1/2}\left[\frac{1}{6}m^n(1-e^{np})\frac{T}{m^n}\left[n^p\frac{\partial n^n}{\partial x_i}-n^n\frac{\partial n^p}{\partial x_i}\right]+\frac{2}{3}n^p\rho^n M^{np}\frac{\partial T/m^n}{\partial x_i}\right.$$
$$\left.+\frac{1}{4}(1-e^{np})n^p\rho^n(M^{pn}-M^{np})\frac{\partial T/m^n}{\partial x_i}\right]$$
$$+\pi n^p\rho^n\frac{T}{m^n}\left[\frac{1}{3}(M^{pn}v_i^p+M^{np}v_i^n)+\frac{1}{2}(1-e^{np})M^{pn}(v_i^n-v_i^p)\right]$$
$$+\frac{\pi}{10}M^{pn}n^p\rho^n[2(M^{pn}a_{imm}^p+M^{np}a_{imm}^n)+M^{pn}(1-e^{np})(a_{imm}^n-a_{imm}^p)]\Bigg\rangle\;; \tag{A2}$$

the rate of energy dissipation per unit volume

$$\gamma^n=\sum_{p=1}^{s}2\sigma^{np^2}(1+e^{np})g_c^{np}n^p\rho^n\Bigg\langle\frac{T}{m^n}\left\{(1-e^{np})\left[2\pi M^{pn}\left[\frac{T}{m^n}\right]\right]^{1/2}\right.$$
$$\left.+\frac{\pi}{6}\sigma^{np}(M^{pn}-M^{np})\frac{\partial u_p}{\partial x_p}-\frac{\pi}{2}\sigma^{np}M^{pn}(1-e^{np})\frac{\partial u_p}{\partial x_p}\right\}$$
$$-\left[2\pi M^{pn}\left[\frac{T}{m^n}\right]\right]^{1/2}\left[2\left[\frac{\theta^p}{m^p}M^{pn}-\frac{\theta^n}{m^n}M^{np}\right]-\frac{3}{2}M^{pn}(1-e^{np})\left[\frac{\theta^p}{m^p}+\frac{\theta^n}{m^n}\right]\right]\Bigg\rangle\;; \tag{A3}$$

the flux of the pressure tensor

$$q_{lip}^n=3\rho^n\left[\frac{T}{m^n}v_{(s}^n\delta_{li}\delta_{ps)}+\frac{1}{5}a_{(sqq}^n\delta_{li}\delta_{ps)}\right]$$
$$+\sum_{p=1}^{s}\sigma^{np^3}(1+e^{np})g_c^{np}\Bigg\langle-\frac{1}{5}\left[2\pi M^{pn}\left[\frac{T}{m^n}\right]^3\right]^{1/2}\sigma^{np}(1-e^{np})m^n\left[n^p\frac{\partial n^n}{\partial x_{(s}}\delta_{li}\delta_{ps)}-n^n\frac{\partial n^p}{\partial x_{(s}}\delta_{li}\delta_{ps)}\right]$$
$$-\frac{1}{10}\sigma^{np}n^p\rho^n\left[2\pi M^{pn}\frac{T}{m^n}\right]^{1/2}[8M^{np}+3(1-e^{np})(M^{pn}-M^{np})]\frac{\partial T/m^n}{x_{(s}}\delta_{li}\delta_{ps)}$$
$$+n^p\rho^n\frac{T}{m^n}\left[\frac{1}{5}\pi M^{pn}[3(1-e^{np})-2][v_{(s}^n\delta_{li}\delta_{ps)}-v_{(s}^p\delta_{li}\delta_{ps)}]+\frac{\pi}{3}(v_l^n\delta_{ip}+v_i^n\delta_{lp})\right]$$
$$+\frac{\pi}{5}\rho^n n^p\frac{m^p}{m^n}(M^{pn}-M^{np})\left[\left[\frac{3}{5}(1-e^{np})-\frac{2}{5}\right]\left[M^{pn}a_{(sqq}^p\delta_{li}\delta_{ps)}+M^{np}a_{(sqq}^n\delta_{li}\delta_{ps)}\right]\right.$$
$$\left.+\frac{1}{3}\left[M^{pn}(a_{lqq}^p\delta_{ip}+a_{iqq}^p\delta_{lp})+M^{np}(a_{iqq}^n\delta_{lp}+a_{lqq}^n\delta_{ip})\right]\right]$$
$$+\frac{2}{25}\pi\frac{m^p}{m^n}n^p\rho^n\left[\frac{1}{2}[3(1-e^{np})-2][M^{np^2}a_{(sqq}^n\delta_{li}\delta_{ps)}-M^{pn^2}a_{(sqq}^p\delta_{li}\delta_{ps)}]\right.$$
$$\left.+\frac{5}{6}[M^{np^2}(a_{lqq}^n\delta_{pi}+a_{iqq}^n\delta_{lp})-M^{pn^2}(a_{lqq}^p\delta_{pi}+a_{iqq}^p\delta_{lp})]\right]$$
$$+\frac{\pi}{5}M^{pn}n^p\rho^n\left[\frac{4}{5}[M^{pn}a_{(sqq}^p\delta_{li}\delta_{ps)}+M^{np}a_{(sqq}^n\delta_{li}\delta_{ps)}]\right.$$
$$\left.+\frac{1}{3}[M^{pn}(a_{lqq}^p\delta_{pi}+a_{iqq}^p\delta_{lp})+M^{np}(a_{lqq}^n\delta_{pi}+a_{iqq}^n\delta_{lp})]\right]\Bigg\rangle\,, \tag{A4}$$

where the parentheses around a set of N indices including M free indices represent the sum of all permutations of the free indices only divided by $M!$: it is worth noticing that the flux of the pressure tensor is symmetric only with respect to the two first indices; the flux of the energy flux

$$q_{li}^{n}=5\rho^{n}\frac{T}{m^{n}}\left[\left[\frac{T}{m^{n}}+2\frac{\theta^{n}}{m_{n}}\right]\delta_{li}+\frac{7}{5}a_{\langle li\rangle}^{n}\right]$$

$$+\sum_{p=1}^{s}\sigma^{np^{3}}(1+e^{np})g_{c}^{np}\rho^{n}n^{p}\left\langle\pi\left[\frac{T}{m^{n}}\right]\left[\frac{T}{m^{n}}\left[\frac{5}{3}+(1-e^{np})M^{pn}-(1-e^{np^{2}})M^{pn}\right]\right.\right.$$

$$-2(1-e^{np^{2}})M^{pn^{2}}\left[\frac{\theta^{n}}{m^{n}}+\frac{\theta^{p}}{m^{p}}\right]-\left[(1-e^{np})M^{pn}-2M^{pn}-\frac{5}{3}\right]\left[M^{pn}\frac{\theta^{p}}{m^{p}}-M^{np}\frac{\theta^{n}}{m^{n}}\right]$$

$$+2(1-e^{pn})M^{pn}\frac{\theta^{n}}{m^{n}}+\frac{10}{3}\frac{\theta^{n}}{m^{n}}\Bigg]\delta_{li}-\frac{\sigma^{np}}{3}(1-e^{np})\left[2\pi M^{pn}\left[\frac{T}{m^{n}}\right]^{3}\right]^{1/2}\frac{\partial u_{l}}{\partial x_{i}}+\sigma^{np}\left[2\pi M^{pn}\left[\frac{T}{m^{n}}\right]^{3}\right]^{1/2}$$

$$\times\left[-\frac{4}{5}(1-e^{np})M^{pn}+\frac{16}{15}(1-e^{np^{2}})M^{pn}+\frac{4}{15}(1-e^{np})(M^{pn}-M^{np})+\frac{4}{15}M^{np}-\frac{8}{5}\right]\left[\frac{5}{6}\frac{\partial u_{p}}{\partial x_{p}}\delta_{li}+\overset{\circ}{\overline{\overline{\frac{\partial u_{l}}{\partial x_{i}}}}}\right]$$

$$+\frac{T}{m^{n}}\left[\frac{2\pi}{3}(M^{np^{2}}a_{\langle li\rangle}^{n}+M^{pn^{2}}a_{\langle li\rangle}^{p})+\left[\frac{2\pi}{3}M^{np}M^{pn}+\frac{14\pi}{15}M^{pn^{2}}+\frac{7\pi}{5}(1-e^{np})M^{pn^{2}}\right.\right.$$

$$\left.\left.\left.-\frac{4\pi}{5}(1-e^{np^{2}})M^{pn^{2}}\right](a_{\langle li\rangle}^{n}+a_{\langle li\rangle}^{p})+M^{pn}\left[\frac{9\pi}{5}(1-e^{np})+\frac{2\pi}{15}\right](M^{np}a_{\langle li\rangle}^{n}-M^{pn}a_{\langle li\rangle}^{p})\right]\right\rangle ; \tag{A5}$$

and the sourcelike terms

$$\chi^{np}(C_{i})=\sigma^{np^{2}}(1+e^{np})g_{c}^{np}\rho^{n}n^{p}\left\{\frac{\pi}{3}\sigma^{np}\left[(M^{pn}-M^{np})\frac{\partial T/m^{n}}{\partial x_{i}}-\frac{T}{m^{n}}\frac{n^{n}}{n^{p}}\frac{\partial}{\partial x_{i}}\left[\frac{n^{p}}{n^{n}}\right]\right]\right.$$
$$\left.-\frac{4}{3}\left[2\pi M^{pn}\frac{T}{m^{n}}\right]^{1/2}(v_{i}^{n}-v_{i}^{p})+\frac{2}{15}\left[2\pi M^{pn^{3}}\frac{m^{n}}{T}\right]^{1/2}(a_{iqq}^{p}-a_{iqq}^{n})\right\} , \tag{A6}$$

$$\chi^{np}(C_{i}C_{q})=2\sigma^{np^{2}}(1+e^{np})g_{c}^{np}n^{p}\rho^{n}\left\langle\left\{-\frac{2}{3}(1-e^{np})\left[\frac{T}{m^{n}}\right]+\left[\frac{4}{3}\left[M^{pn}\frac{\theta^{p}}{m^{p}}-M^{np}\frac{\theta^{n}}{m^{n}}\right]-(1-e^{np})M^{pn}\left[\frac{\theta^{p}}{m^{p}}+\frac{\theta^{n}}{m^{n}}\right]\right]\right\}\right.$$
$$\times\left[2\pi M^{pn}\frac{T}{m^{n}}\right]^{1/2}\delta_{iq}-\frac{4}{3}\left[2\pi M^{pn}\frac{T}{m^{n}}\right]^{1/2}(M^{np}a_{\langle iq\rangle}^{n}-M^{pn}a_{\langle iq\rangle}^{p})$$
$$-\frac{2}{5}\left[2\pi M^{pn^{3}}\frac{T}{m^{n}}\right]^{1/2}[2+(1-e^{np})](a_{\langle iq\rangle}^{n}+a_{\langle iq\rangle}^{p})$$
$$-\pi\sigma^{np}\frac{T}{m^{n}}\left\{\frac{1}{3}(M^{pn}-M^{np})D_{iq}\right. \tag{A7}$$
$$\left.\left.-\frac{1}{5}M^{pn}\left[2\overset{\circ}{\overline{\overline{\frac{\partial u_{i}}{\partial x_{q}}}}}+(1-e^{np})\left[2D_{iq}+\frac{\partial u_{p}}{\partial x_{p}}\delta_{iq}\right]\right]\right\}\right\rangle ,$$

where D_{iq} is the rate of deformation tensor, and

$$\chi^{np}(C_i C_q C_q) = \sigma^{np^2} g_c^{np} n^p \rho^n (1+e^{np})$$
$$\times \Bigg\langle \pi\sigma^{np} \left[\left[\frac{T}{m^n} \right]^2 \left[(1-e^{np^2})M^{pn} - (1-e^{np})M^{pn} - \frac{5}{3} \right] \frac{n^n}{n^p} \frac{\partial}{\partial x_i} \left[\frac{n^p}{n^n} \right] \right.$$
$$+ M^{pn} \frac{T}{m^n} \left[-\frac{2}{3} + 4M^{pn} + 6(1-e^{np})M^{np} + 2(1-e^{np})(M^{pn}-M^{np}) \right.$$
$$\left. \left. - 2(1-e^{np^2})(M^{pn}-M^{np}) \right] \frac{\partial T/m^n}{\partial x_i} \right]$$
$$- \frac{20}{3}(1-e^{np}) \left[2\pi M^{pn} \left[\frac{T}{m^n} \right]^3 \right]^{1/2} (M^{pn} v_i^p + M^{np} v_i^n)$$
$$- \left[2\pi M^{pn} \left[\frac{T}{m^n} \right]^3 \right]^{1/2} \left[8 - \frac{4}{3} M^{np} + 8(1-e^{np})M^{pn} - \frac{16}{3}(1-e^{np^2})M^{pn} \right] (v_i^n - v_i^p)$$
$$+ \left[2\pi M^{pn} \frac{T}{m^n} \right]^{1/2} \left[\frac{4}{5}(1-e^{np})(1-2e^{np})M^{pn^2}(a_{iqq}^p - a_{iqq}^n) \right.$$
$$- \frac{16}{15}(1+e^{np})M^{pn}(M^{pn} a_{iqq}^p + M^{np} a_{iqq}^n)$$
$$- \frac{82}{15}(1-e^{np})M^{pn}(M^{pn} a_{iqq}^p + M^{np} a_{iqq}^n) + \frac{2}{3} M^{pn} M^{np} (a_{iqq}^p - a_{iqq}^n)$$
$$\left. + 4(M^{pn^2} a_{iqq}^p - M^{np^2} a_{iqq}^n) + \frac{12}{5} M^{pn^2}(a_{iqq}^p - a_{iqq}^n) \right] \Bigg\rangle . \quad \text{(A8)}$$

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