Lateral diffusive migration of massive particles in high-velocity vertical pipe flow of moderately dense gas-solid suspensions

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Transport processes involved in a gas-particle flow, comprised of spherical particles with a narrow size distribution suspended in a turbulent gas, are investigated theoretically on the basis of the recently developed Enskog theory for multicomponent dense mixtures of slightly smooth inelastic spherical particles [P. Zamankhan, Phys. Rev. E **52**, 4877 (1995)]. The generalized Boltzmann equation of the previous work is modified to incorporate the relevant forces exerted upon individual particles including the drag force by the relative gas motion. Extending the method of moments of Grad [Commun. Pure Appl. Math. **2**, 331 (1949)], the modified Boltzmann equation is solved to obtain the nonequilibrium velocity distribution function for particles of each size. By taking the monodisperse limit, a basic equation is derived for the treatment of the problem of lateral diffusive migration of solids in an assembly composed of separate equisized spherical particles traveling in a fully developed, turbulent upward flow of a gas within a vertical pipe. At moderately high solid concentrations, where the random component of the particle velocity is generated mainly by particle-particle collisions, the particle diffusivity and the thermal diffusion coefficient are found to increase with the square root of the granular temperature, a term that measures the energy of the random motion of the particles.
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I. INTRODUCTION

During the past few years, the hydrodynamics of confined gaseous suspensions has received considerable attention due to its importance in several applications, including fluid catalytic cracking [1] and combustion of low-grade coal in power generation plants [2]. In observing the transport of moderately dense gas solid particles in vertical pipes, where the solid volume fraction is much larger than 0.001, it has been noted [3] that solid particles were distributed nonuniformly over the cross section and that the recirculation of particles occurred against the direction of their net flow. These phenomena clearly influence the particle residence time distribution in the risers, which is important in predicting the behavior of systems in which the particles catalyze reactions between species in the gas or in which they react with the gas.

Several approaches towards developing two-fluid models, in which the gas and the particle phases are treated as a mixture of continua, have been used in an effort to predict the aforementioned observations. The results of the earliest approach, used by Berker and Tulig [4], demonstrated that for the pipe flow regimes relevant to high-velocity gas fluidized beds (6–9 m/s), the particle-eddy interactions tend to move particles laterally due to the presence of a turbulence gradient. As a result of this motion, an uneven distribution of particles over the cross section may appear. The essential approximation in their approach is that the particle collision time τ_c, which characterizes the mean time between successive collisions of a particle, is much larger than the particle viscous relaxation time τ_d, which describes the response of a particle to the drag force created by turbulent fluctuating velocity of the surrounding gas.

Under the operating conditions used for gas fluidized beds, there are many parameters that affect the motion of suspended solid particles relative to a turbulent carrier fluid, including particle inertia and solids loading. For moderately concentrated particle loadings, recent tests [5] indicated that in high-gas-velocity flows, large-scale solid structures (clusters) [3,6], which may be observed at lower velocities with the same rate of transport of solid, give way to a population composed of separate particles whose free and independent motions resemble that of gas molecules in a dense gas in thermal equilibrium. Thus, for this case there is some justification for assuming that the particle collision time τ_c is much smaller than the particle viscous relaxation time τ_d. Under such circumstances the flow regime is not fluid dominated; instead, the frequency of a single-particle displacements is controlled by the rate of collision with the neighboring solid particles [7]. Hence the study of the lateral diffusive migration of solids in a turbulent upward flow of a gas within a vertical pipe at moderately high solid concentrations, which seems to be a possible cause for the tendency of particles to concentrate in the wall region, requires an essentially different approach from those proposed for particle-laden turbulent flows [8].

Developing theories based on the kinetic theory of dense gases [9] to obtain continuum equations for the mass, momentum, and energy of the solid phase, therefore, could

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present an important step toward a better understanding of the fundamentals involved in gas-particle flows in vertical risers. The idea is that the particle phase in vertical pipe flow of gas-solid suspensions is treated as a granular material, where momentum is primarily transferred during instantaneous particle-particle collisions. The difficulty with applying kinetic theory analysis to particle phase of suspension flows is that the theory depends on assumptions that are appropriate for gas molecules. Unlike the kinetic energy of gas molecules, the solid-particle kinetic energy is not necessarily conserved in collisions due to the inelasticity of the particles. Hence the formulations in the kinetic theory obtained assuming the reversibility of collisions should be modified to account for the energy dissipated in solid-body collisions.

Recently, such a modified kinetic theory [10] has been used by Sinclair and Jackson [7] in describing the fully developed flow of gas and monosized particles in a vertical pipe. More recently, Louge, Mastorakos, and Jenkins [11] have developed an improved model by considering the effect of gas turbulence, making the approximation that the particles react sluggishly to turbulent velocity fluctuations of the surrounding gas and therefore velocity fluctuations at the level of individual particles are induced by interparticle collisions. After the study described in this paper was underway, Dasgupta, Jackson, and Sunderasan [12] developed a different approach to studying the role of the fluctuations associated with the organized motion of collections of particles on the occurrence of segregation in turbulent gas-particle flows in vertical risers. Considering the dispersion flux of small particles, Dasgupta, Jackson, and Sunderasan applied a semiempirical scheme [13], which has been widely used to model the radial dispersion velocity for a gas suspension of small solid particles when the particle-fluid interaction is the dominating mechanism that controls particle diffusivity. The idea was that the response of individual particles to the fluid velocity fluctuations leads to the occurrence of a time-averaged diffusive flux of solids in the direction of decreasing turbulence intensity. More general studies of the particle dispersion flux, when the frequency of the particles' displacement is controlled by their rate of collisions with the neighboring solid particles, appear to be lacking, however.

As suggested by Batchelor [14], a detailed theoretical study on transport properties such as the particle diffusion coefficient, which characterizes a tendency of particles to migrate from high- to low-solid-concentration regions, allows for the development of more accurate criteria to assess the instability of gas-particle flows. Recently, Koch [15] stated that the relative diffusion of particles and fluid and the associated forces in a monodisperse gas-solid suspension play a similar role to the effective pressure [12] in the particle phase. However, the distinctions have not been explored between drift of grains from regions of high to low shear in a confined flow, which generates concentration inhomogeneities that induce ordinary diffusion, and those due to the presence of gradients of granular temperature, which results in an extra diffusive flux along these gradients. Hence more general studies on the particle diffusivity and the particle thermal diffusion coefficient in moderately dense gas-solid suspensions based on rigorous kinetic theory of granular fluids are needed both to test existing approximation methods and to pave the way towards new approaches.

In light of the above concerns, the current study has two objectives.

First, an approximate theory is constructed for describing the motion of particles in a flow comprised of spherical particles with a narrow size distribution, suspended in a moderately dense suspension of multisized massive particles in a turbulent carrier gas. The theory is based on a generalized Boltzmann equation using the recently developed kinetic theory of the dense mixtures of solid particles with distribution in particle size [16], which is modified to include the resistant forces [17] with those due to pressure gradient and external field in addition. By definition, particles are considered "massive" if their hydrodynamic relaxation times τ_d are much larger than the particle-eddy crossing time τ_i [18], which is assumed to be smaller than the Lagrangian time macroscale of the turbulence τ_l. Extending the method of moments of Grad [19], the generalized Boltzmann equation is solved to obtain the nonequilibrium velocity distribution function for particles of each size. It is assumed that the flow mechanics of particles in a multicomponent gas suspension flow can be adequately described by consideration of the 13-moment approximation. In conjunction with this effort, analytical relations are developed for the lateral particle diffusion coefficients and thermal diffusion ratios when the random component of the particle velocity is generated mainly by solid-body collisions between the particles. For this case, the particle diffusivity and the thermal diffusion coefficient are found to increase with the square root of the granular temperature. The granular temperature is a quantity that measures the energy of the random motion of the particles.

Second, the theoretical particle diffusivity and thermal diffusion coefficient are tentatively used to evaluate the lateral particle diffusion velocity in fully developed, turbulent, vertical pipe flows of moderately dense gas-solid suspensions of the uniform-size spherical particles. The results of the present theory for particle diffusivities as a function of particle size are compared with the values of long-time diffusion coefficients obtained for collisionless conditions [20]. Moreover, the segregation effect arising from the particle thermal diffusion, which results from a coupling between dissipative mass and heat flows, is discussed.

II. ANALYSIS

Massive particles respond sluggishly to gas velocity variations and their trajectories are relatively straight in spite of the gas turbulence [20], in situations where the solids concentration tends towards zero. Consider a massive particle perturbed from the state of local equilibrium slip condition (which would hold if the particle was subjected only to viscous interaction with the gas) by a collision with its neighboring particle. To return to the local state of slip, the particle is acted on by the resistant force tending to accelerate (particle velocity after the collision less than the local gas speed) or decelerate (particle velocity after the collision greater than the local gas speed) it toward the local gas velocity. In the limit $\tau_c \ll \tau_d$ where the particle mean free path is not large compared to its diameter, the random fluctuations that result from one collision have not significantly decayed before the next collision takes place. For this case, as in motions of molecules of a dense gas in a state of molecular chaos, where colliding molecules are distributed at random without correlation between velocity, two arbitrary particles will move in such a way that statistically the distance between them increases with time. In the presence of a particle concentration gradient, this gives rise to an observable effect [21], which is a gradual spread or ''diffusive motion'' of the particles traveling in the gas. For the situation to be considered here, where the particle Stokes [15] number is large, the rapidness of the particle movement depends on the angle of the particle direction after a collision relative to the line of action of the gravitational force. It is the highest if the particle after a collision is directed parallel to the direction of the gravitational field due to the complete conversion of particle potential energy into particle motion. Since the particle path is random, in an average sense, the gravitational force may not affect the particle collision time.

The problem addressed here is the derivation of a basic equation to describe the diffusive motion of massive spherical particles, particularly in the lateral direction, in a fully developed, turbulent, moderately dense gas-particle flow within a vertical pipe, where the particle displacements frequency is controlled by its rate of collision with the neighboring solid particles. Due to the random character of particle-particle collisions, the statistical-mechanics approach makes an important contribution to the understanding of the diffusional processes involved in this system. In the following statistical descriptions, only broad features of the particulate motion are made use of and a connection is sought with the particle diffusivity and the thermal diffusion coefficient. In the first place, this is hoped to assist in the interpretation of experimental results and to point the way towards new approximate theories.

Consider a moderately dense suspension of massive solid particles consisting of a mixture of s different-size particles in a vertical upflow of a turbulent gas. The particles are assumed to be hard, smooth, but nearly elastic. They are sufficiently large that the effects of Brownian motion, virtual mass, and Basset history forces can be neglected. Additionally, the effects of the rotational motion of the particles and electrostatic interparticle forces are not accounted for.

The expected number density of the n particles in the s-component mixture having m_n mass and σ_n diameter in a fixed volume element $d\mathbf{x}_n$ centered at a point $\mathbf{x}_n$ having velocities in the range $\mathbf{c}_n$, $\mathbf{c}_n+d\mathbf{c}_n$, where $d\mathbf{c}_n$ is a velocity element, at time t is represented by

$$f_n=f_n(\mathbf{x}_n,\mathbf{c}_n,t). \tag{1}$$

The evolution of the number density of the n particles may be described by the generalized Boltzmann equation of the previous work [16], which has been modified to incorporate the forces upon the particles resulting from the relative gas motion with those due to pressure gradient in addition. Then one may write

$$\begin{aligned}\frac{\mathrm{d}f_n(\mathbf{x}_n,\mathbf{C}_n,t)}{\mathrm{d}t}=&\left[-\mathbf{C}_n\cdot\frac{\partial f_n(\mathbf{x}_n,\mathbf{C}_n,t)}{\partial\mathbf{x}}+\frac{\partial\mathbf{u}}{\partial\mathbf{x}}:\left(\mathbf{C}_n\frac{\partial f_n(\mathbf{x}_n,\mathbf{C}_n,t)}{\partial\mathbf{C}}\right)+\frac{\mathrm{d}\mathbf{u}}{\mathrm{d}t}\cdot\frac{\partial f_n(\mathbf{x}_n,\mathbf{C}_n,t)}{\partial\mathbf{C}}-\frac{\partial}{\partial\mathbf{C}}\cdot\langle f_n(\mathbf{x}_n,\mathbf{C}_n,t)\mathbf{F}_n\rangle\right]\\&+\sum_{j=1}^{s}\int\int\sigma_{nj}^2[g_{nj}(\mathbf{x}_n,\mathbf{x}_n+\sigma_{nj}\mathbf{k}|\{n_n\})f_n(\mathbf{x}_n,\mathbf{C}_n,t)f_j(\mathbf{x}_n+\sigma_{nj}\mathbf{k},\mathbf{C}_j',t)-g_{nj}(\mathbf{x}_n,\mathbf{x}_n\\&-\sigma_{nj}\mathbf{k}|\{n_n\})f_n(\mathbf{x}_n,\mathbf{C}_n,t)f_j(\mathbf{x}_n-\sigma_{nj}\mathbf{k},\mathbf{C}_j,t)](\mathbf{c}_{nj}\cdot\mathbf{k})H[\mathbf{c}_{nj}\cdot\mathbf{k}]\mathrm{d}\mathbf{k}\,\mathrm{d}\mathbf{c}_j.\end{aligned} \tag{2}$$

Since the particle diffusion processes involved in a gas-particle flow are of interest in the present study, the new variable $\mathbf{C}_n=\mathbf{c}_n-\mathbf{u}$, which is the random velocities of the n particles with reference to the mass mean velocity $\mathbf{u}$ of the solid mixture, is now regarded as independent of $\mathbf{x}_n$ rather than $\mathbf{c}_n$. The generalized Boltzmann equation for the n particles is coupled with that of carrier gas through the external force per unit mass $\mathbf{F}_n$ of the drift term, which is the first term on the left-hand side of Eq. (2), which represents the rate of change of the distribution function due to motion of particles without collisions, and with those of the neighboring solid particles having different masses m_j and diameters σ_j ($j=1,2,...,s$) through the collision terms, which are incorporated in the second term on the right-hand side of Eq. (2), which represents the effect of collisions between the particles. Here d/dt is the substantial time derivative, $\mathbf{c}_{nj}$ is the relative velocity of two particles with velocities $\mathbf{c}_n$ and $\mathbf{c}_j$, $\{n_n\}$ is the component densities, $\mathbf{k}$ is the apse vector for collisions, g_{nj} is the radial distribution function of two particles, one of component n and the other of component j, at contact when the distance of their centers is $\sigma_{nj}=(\sigma_n+\sigma_j)/2$, σ_{nj} represents the collision diameter, and the Heaviside function $H[\mathbf{c}_{nj}\cdot\mathbf{k}]$ selects those particles that have had a collision and are leaving the collision cylinder [22].

To gain an understanding of the inelasticity effects of particles, which leads to energy flows unidirectionally from the translational degrees of freedom into internal modes of the particles during particle-particle collisions, the velocities of the restituting collision $\mathbf{c}_n'$ and $\mathbf{c}_j'$, which are related to those of the direct collision $\mathbf{c}_n$ and $\mathbf{c}_j$, are defined using the concept of the coefficient of restitution e:

$$\begin{aligned}\mathbf{c}_n'&=\mathbf{c}_n-M_{jn}(1+e_{nj})(\mathbf{c}_{nj}\cdot\mathbf{k})\mathbf{k},\\\mathbf{c}_j'&=\mathbf{c}_j+M_{nj}(1+e_{jn})(\mathbf{c}_{nj}\cdot\mathbf{k})\mathbf{k},\end{aligned} \tag{3}$$

where $M_{jn}=m_j/(m_j+m_n)$ and e_{nj} is the coefficient of restitution for a collision between the nj pair of particles. Although the coefficient of restitution e_{nj} depends on the particle impact velocity, the concept of e as a velocity-independent material constant is still widely used [23]. Here the coefficients of restitution for the particles are assumed to be constant and to have values near unity.

In the present attempt to generalize the Boltzmann equation to higher densities, the collision integral is expressed in the form used in the revised Enskog theory of Van Beijeren and Ernst [24]. The *stosszahlansatz* is modified by introducing a factor g_{nj} to account for the correlation between the positions of two colliding particles and the resulting increase in the frequency of binary collisions. The factor g_{nj}, which is an important factor in determining the thermodynamic properties of the particle assembly, is chosen to be the nonuniform pair distribution function, which takes into account the spatial nonuniformities in the local equilibrium state. Consider an assembly of particles in a flowing gas-particle suspension with a set of states α, each of which has an energy $\mathcal{H}_\alpha$, which is the total energy of the assembly in that state. If the assembly is at a granular temperature $T>0$, its state α will vary with time and quantities such as $\mathcal{H}_\alpha$ that depend on the state will fluctuate. After a change of parameters, the fluctuations will have on average a definite direction, say, the direction of decreasing energy $\mathcal{H}_\alpha$. After a while any such trend vanishes and the assembly just fluctuates around a condition that is called thermal equilibrium. If the effect of the interstitial gas is negligible, the equilibrium state of the particle assembly is a static configuration with a zero granular temperature, due to the dissipation of particle fluctuating energy via inelastic collisions. However, in gas-particle flows in vertical risers energy will be supplied to the particles by the suspending gas. Thus, in this instance there is some justification assuming that the particle assembly behaves like the dense gas molecules in nonuniform equilibrium and therefore using the pair distribution function defined by

$$g_{nj}(\mathbf{x}_n,\mathbf{x}_n\pm\sigma_{nj}\mathbf{k}|\{n_n\})=g^c_{nj}(\sigma_{nj}|\{n_n\})+\sum_{l=1}^{s}\int \mathrm{d}\mathbf{x}_l H_{njl}(\mathbf{x}_n,\mathbf{x}_n\pm\sigma_{nj}\mathbf{k},\mathbf{x}_l|\{n_n\})(\mathbf{x}_l-\mathbf{x}_n)\cdot\frac{\partial n_l}{\partial \mathbf{x}}+O(\nabla^2), \quad (4)$$

where g^c_{nj} is the equilibrium value of the radial distribution function for nj pair of particles at contact

$$g^c_{nj}(\sigma_{nj}|\{n_n\})=1+\sum_{l=1}^{s} n_l(\mathbf{x}_n)\int V_{njl}(\mathbf{x}_n,\mathbf{x}_n\pm\sigma_{nj}\mathbf{k}|\mathbf{x}_l)\mathrm{d}\mathbf{x}_l+\cdots . \quad (5)$$

Here

$$H_{njl}(\mathbf{x}_n,\mathbf{x}_n\pm\sigma_{nj}\mathbf{k},\mathbf{x}_l|\{n_n\})=V_{njl}(\mathbf{x}_n,\mathbf{x}_n\pm\sigma_{nj}\mathbf{k}|\mathbf{x}_l)+\sum_{l'=1}^{s} n_{l'}(\mathbf{x}_n)\int V_{njll'}(\mathbf{x}_n,\mathbf{x}_n\pm\sigma_{nj}\mathbf{k}|\mathbf{x}_l\mathbf{x}_{l'})\mathrm{d}\mathbf{x}_{l'}+\cdots ,$$

$V_{njl}(\mathbf{x}_n,\mathbf{x}_n\pm\sigma_{nj}\mathbf{k}|\mathbf{x}_l)$ and $V_{njll'}(\mathbf{x}_n,\mathbf{x}_n\pm\sigma_{nj}\mathbf{k}|\mathbf{x}_l\mathbf{x}_{l'})$ represent Husimi functions [25]. There is, however, very little justification for assuming the isotropic equilibrium value of the radial distribution function (5) for the particle assembly in a gas-particle flow in vertical risers, where, for instance, the particle diffusion coefficients associated with the streamwise direction are somewhat larger than those associated with the lateral directions. While the significance of an ellipsoidal distribution function, whose principal axes is parallel to the streamwise direction, for a nonequilibrium particle assembly in gas-particle flows in vertical risers cannot be neglected, it is not obvious how to treat this problem rigorously. An attempt to model the linear anisotropy of the pair distribution function in phase space at the point of collision using a functional of local solid volume fraction was suggested by Mello, Diamond, and Levine [26], but the proposed expression for the pair distribution function was in fact a crude approximation.

The solution of Eq. (2), based on a generalized Grad moment method [19], may be approximated by

$$f_n(\mathbf{x}_n,\mathbf{C}_n,t)=n_n\left[\frac{m_n}{2\pi T}\right]^{3/2}\exp\left[-\frac{m_n\mathbf{C}_n\cdot\mathbf{C}_n}{2T}\right]\left[1+\frac{m_n\mathbf{v}_n\cdot\mathbf{C}_n}{T}+\frac{3}{2}\left(-\frac{\theta_n}{T}+\frac{m_n}{3T^2}\,\theta_n\mathbf{C}_n\cdot\mathbf{C}_n\right)+\frac{1}{2}\left(\frac{m_n}{T}\right)^2\mathbf{A}_n:\mathbf{C}_n\mathbf{C}_n+\frac{1}{10}\left(\frac{m_n}{T}\right)^2\times\left(\frac{m_n}{T}\,\mathbf{C}_n\cdot\mathbf{C}_n-5\right)\mathbf{a}_n\cdot\mathbf{C}_n\right], \quad (6)$$

where $\mathbf{A}_n$ and $\mathbf{a}_n$ represent the pressure deviator tensor, which is a second rank tensor, and the transport pseudothermal energy flux vector of the n particles, respectively. Here $\mathbf{v}_n$ represents the diffusion velocities of the n particles relative to the local mass mean velocity of the solid mixture. Thus the mean velocity of the n particles in a space-fixed coordinate system can be written as $\mathbf{u}_n=\mathbf{v}_n+\mathbf{u}$. It is further assumed that the particle relaxation time is much greater than the particle-eddy crossing time, ensuring the particles-turbulence interaction may be limited to the influence of the gas on the mean velocity of the solid mixture $\mathbf{u}$. The particles, which are moving randomly due to solid-body collisions, transfer energy from the mean flow of the suspension to the particle fluctuating energy that is dissipated mainly by inelastic collisions or in the long run by viscous forces. Note that the granular temperature of the mixture $T=1/n\Sigma_{j=1}^{s}n_jT_j$ differs from T_j $(j=1,2,...,s)$, which is the granular temperature of the j particles, by a quantity θ_j. Here $n=\Sigma_{j=1}^{s}n_j$ represents the solid mixture number density.

In the spirit of linearization, quantities such as $\mathbf{v}_j$, $\mathbf{A}_j$, and $\mathbf{a}_j$ for the s components $(j=1,2,...,s)$, which describe deviations from equilibrium, are all regarded as small. Moreover, by considering the recent tests by Ippolito *et al.* [27], the present theory may be valid in the limit in which the particulate phase is comprised of spherical particles with a narrow-size distribution where all the s-particle granular temperature perturbations θ_j $(j=1,2,...,s)$ can be considered small.

At a hydrodynamic stage, in which scales of length and time are considerably larger than those characteristic of the particulate level, an approximate equation can be constructed for the lateral diffusive motion of the n particles in a flow of moderately dense gas-solid suspension of multi-sized particles in a vertical riser using scaling arguments. To this end the approximate solution (6) is substituted into the generalized Boltzmann equation (2). After both sides have been multiplied by the quantity $\mathbf{C}_n$ the mass weighted average [16] is taken to derive a balance of momentum. Then a term-by-term order of magnitude analysis of the balance of momentum is carried out to find the dominant terms. In order to cast the lateral component of momentum balance in dimensionless form, dimensionless ratios for the lateral diffusion velocity v_{nr}, the lateral pseudothermal energy flux a_{nr}, and the particle granular temperature perturbation θ_n of the n-particles are made by dividing them by $T_0^{1/2}$, $T_0^{3/2}$, and T_0, respectively. Here $T_0^{1/2}$ represents the characteristic value of the solid mixture fluctuating velocity. The dimensionless ratios $v_{nr}^*=v_{nr}/T_0^{1/2}$, $\theta_n^*=\theta_n/T_0$, and $a_{nr}^*=a_{nr}/T_0^{3/2}$ are all assumed to be small and of the same order of magnitude. Three more dimensionless group can be defined that are useful for the scaling in this problem. They are as follows.

(i) $\tau_n^{(1)}=\tau_{nd}/\tau_{ni}$ represents the ratio of the n-particle relaxation time to the relevant fluid time scale, which is the n-particle eddy crossing time due to gravity defined by $\tau_{ni}=l_e/\Delta W_n^T$. Here l_e represents the eddies characteristic size and ΔW_n^T is the free fall velocity of a single n particle. A large value of $\tau_n^{(1)}$ means that the n particles react sluggishly to the gas velocity variations.

(ii) $\tau_{nj}^{(2)}=\tau_{nj}^c/\tau_{ni}$ represents the ratio of the mean time between successive collisions of an n particle with particles of species j $(j=1,2,...,s)$ to the n-particle eddy crossing time due to gravity. For small values of $\tau_{nj}^{(2)}$ the n particles, crossing through an eddy, will encounter several collisions with the j particles. The consequence is that the frequency of the particle displacements are controlled by its rate of collision with the neighboring solid particles.

(iii) $\eta_n=[T_0^{1/2}/T_{\rm cl}^{1/2}][T_{\rm cl}^{1/2}/\overline{V_g^2}^{1/2}][\overline{V_g^2}^{1/2}/\Delta W_n^T]$ represents the ratio of the characteristic value of the solid mixture fluctuating velocity to the free fall velocity of a single n particle. Here $T_{\rm cl}^{1/2}$ is the collisionless particle fluctuating velocity and $\overline{V_g^2}^{1/2}$ is the rms fluctuating gas velocity. The second bracketed term $[T_{\rm cl}^{1/2}/\overline{V_s^2}^{1/2}]$ decreases [18] with increasing $\tau_n^{(1)}$. For a large particle the third term $[\overline{V_g^2}^{1/2}/\Delta W_n^T]$ is small, indicating that large particles, in crossing a gas, experiences the gas turbulence whose direction is rapidly changing.

The dimensionless lengths, velocities, pressure, time, diameter, and granular temperature of the n particles are defined by $x_r^*=x_r/l_e$, $x_z^*=x_z/l_e$, $u_r^*=u_r/T_0^{1/2}$, $u_z^*=u_z/T_0^{1/2}$, $P^*=P/\rho_0 T_0$, $t^*=t/\tau_j$, $\sigma_n^*=\sigma_n/l_e$, and $T_n^*=T_n/m_n T_0$, respectively. Here ρ_0 represents the average material density of the solid mixture. Thus the momentum balance for the n particles in the lateral direction for a fully developed, axisymmetric gas-solid flow of multisized spherical particles, with the approximate expression [28] for the external force per unit mass of the n particles

$$\mathbf{F}_n=-\mathbf{g}-\frac{1}{\rho_n^m}\frac{\partial P_g}{\partial \mathbf{x}}+\frac{\rho_g}{\rho_n^m}[\beta_1 K_1(\phi_s)+\beta_2 K_2(\phi_s)|\mathbf{U}_n^R|]\mathbf{U}_n^R$$

can be cast in the form

$$\eta_n\left[u_r^*\frac{\partial v_{nr}^*}{\partial x_r^*}+v_{nr}^*\frac{\partial u_r^*}{\partial x_r^*}-\frac{1}{\rho_n}\frac{v_{nr}^*}{x_r^*}\frac{\partial(\rho_n x_r^* v_{nr}^*)}{\partial x_r^*}\right]=\sum_{j=1}^{s}\frac{(1+e_{nj})}{\pi\tau_{nj}^{(2)}}\left\{\frac{\pi}{3}\sigma_{nj}^*\left[(M_{jn}-M_{nj})\frac{\partial T_n^*}{\partial x_r^*}-\left(\frac{n_n}{n_j}\right)T_n^*\frac{\partial}{\partial x_r^*}\left(\frac{n_j}{n_n}\right)\right]\right.$$
$$\left.-\frac{4}{3}(2\pi M_{jn}T_n^*)^{1/2}(v_{nr}^*-v_{jr}^*)+\frac{2}{15}\left(2\pi\frac{M_{jn}^3}{T_n^*}\right)^{1/2}(a_{jr}^*-a_{nr}^*)\right\}+\eta_n$$
$$\times\left\{\sum_{j=1}^{s}\left[\delta_{nj}+\frac{\pi}{3}(1+e_{nj})n_j\sigma_{nj}^3 g_{nj}^c\right]\frac{\partial T_n^*}{\partial x_r^*}\right.$$
$$\left.+\frac{T_n^*}{\rho_n}\sum_{j=1}^{s}\frac{n_j}{T}\left(\frac{\partial\mu_j}{\partial n_n}\right)_{T,n_k\neq n_j}\frac{\partial\rho_n}{\partial x_r}\right\}+\eta_n\frac{\rho_0}{\rho}\left\{\frac{\partial P_s^*}{\partial x_r^*}\right.$$
$$\left.+\sum_{j=1}^{s}\left[\phi_j-\left(\frac{\rho_j}{\rho_n}\right)\phi_n\right]\frac{\partial P_g^*}{\partial x_r^*}\right\}+\frac{\rho_n}{\rho}\sum_{j=1}^{s}\rho_j\left(\frac{\overline{U_r^{Rn*}}}{\tau_n^{(1)}}-\frac{\overline{U_r^{Rj*}}}{\tau_j^{(1)}}\right), \qquad (7)$$

where ϕ_j represents the volume fraction of the j particles in the s-component solid mixture $(j=1,2,...,s)$, $P_s^*=P_s/\rho_0 T_0$, and $P_g^*=P_g/\rho_0 T_0$ are the dimensionless solid and gas pressures, respectively, δ_{nj} is the Kronecker delta, $\rho_j=m_j n_j$ is the density of the j particles $(j=1,2,...,n,\dots,s)$, ρ is the mixture density, ρ_n^m is the material density of the n particles, ρ_g is the gas density, the overbar indicates the mass-weighted averaged values, and $\mathbf{U}_n^R$ is the relative velocity between the gas and the n particles whose norm is denoted by $|\mathbf{U}_n^R|$. The explicit representation for the coefficients β_1, β_2, $K_1(\phi_s)$, and $K_2(\phi_s)$ in the above-mentioned expression for external force per unit mass acting on the n particles in a gas suspension is given by Buyevich and Kapbasov [28].

The second term on the right-hand side of Eq. (7) represents the radial component of dimensionless pressure gradient of the particle assembly of kind n, which depends on the chemical potential approximated as [29]

$$\mu_n = T \ln n_n - \tfrac{2}{3}\pi T \frac{\partial}{\partial n_n} \times \left(\sum_{p=1}^{s} \sum_{q=1}^{s} n_p n_q \sigma_{pq}^3 \int_{\infty}^{V} \frac{1}{V'^2} g_{pq}^c dV' \right). \tag{8}$$

The first term on the right-hand side of Eq. (8) represents the chemical potential of the particle assembly of type n when approaching ideal-gas behavior and the remaining terms are the residual chemical potential. Equation (8) is a good approximation if the particle assembly of type n in a gas-particle flow is treated as made up of hard spheres. Hence an approximate expression for the chemical potential of the particle assembly of type n can be obtained by substituting for g_{pq}^c from Eq. (5) in Eq. (8), although the expression is complicated.

It can easily be verified from Eq. (7) that in the limit $\tau_{nj}^{(2)}/\tau_n^{(1)} \ll 1$, the first term on the right-hand side, which represents the source of momentum due to particle-particle collisions, is dominant. Thus, in this instance the explicit expression for particle ordinary diffusion, which is caused by the relative motion of the components of the mixture due to the presence of a density gradient, can be obtained from Eq. (7) assuming that the motion of the assembly of particles in a moderately dense gas-particle flow resembles that of simple fluid molecules for which the heat flux vector is proportional to the gradient of temperature. In an opposite limiting case when $\tau_{nj}^{(2)}/\tau_n^{(1)} \gg 1$, the last term on the right-hand side, which describes the viscous forces exerted on particles by the relative gas motion, plays an important role. In what follows, this limiting case will not be discussed.

Assuming that the wall of the pipe serves as a sink of pseudothermal energy, a pseudothermal particle diffusion, which is caused by the relative motion of the particles due to the presence of a granular temperature gradient, can be observed from regions of high to low granular temperature across the pipe. As a result of this diffusive motion, nonuniform spatial distributions of particle concentration develop in the radial direction, which produce ordinary diffusion, which tends to eliminate these nonuniformities. The separating effect brought about by thermal diffusion may also cause partial separation of particle components in multisized particle-gas flows in vertical risers in which the larger particles usually in the lower granular temperature region and the smaller particles in the higher granular temperature region [30]. This discussion may shed light on the physical origin of the lateral particle mixing observed [5] in gas-particle flows in vertical risers.

The case where the heat flux vector depends on the particle number density gradients, which means that there is interference of diffusion and heat flow, is somewhat more complicated. The phenomenological expression for the mass flux of the n particles in the radial direction relative to the local center of mass velocity, under the condition of no external force and mechanical equilibrium, is

$$\rho_n v_{nr} = -\sum_{j=1}^{s} (1-\delta_{nj}) D_{nj} m_j \left[\frac{\partial n_j}{\partial x_r} + n_j k_{T_j} \frac{\partial \ln T}{\partial x_r} \right], \tag{9}$$

where D_{nj} is the mutual diffusion coefficient and k_{T_j} represents thermal diffusion ratio of the j particles $(j=1,2,...,s)$. In order to derive a constitutive equation for the particle diffusion velocity in the radial direction the balance of the mean of the third moment of velocity of the particle assembly of type n is needed, which is coupled with the balance of momentum through the first term on the right-hand side of Eq. (7). In the limit $\tau_{nj}^{(2)}/\tau_n^{(1)} \ll 1$, the leading-order dimensionless balance of the third moment of velocity in the horizontal plane for a fully developed axisymmetric gas-solid flow of multisized particles may be given as [16]

$$\begin{aligned}
&\sum_{j=1}^{s} \frac{1+e_{nj}}{5\pi\tau_{nj}^{(2)}} \{ -\tfrac{20}{3}(1-e_{nj})(2\pi M_{jn} T^{*3})^{1/2}(M_{jn} v_{jr}^* + M_{nj} v_{nr}^*) - (2\pi M_{jn} T^{*3})^{1/2}(v_{nr}^* - v_{jr}^*)[8 - \tfrac{4}{3}M_{nj} + 8(1-e_{nj})M_{jn} \\
&\quad - \tfrac{16}{3}(1-e_{nj}^2)M_{jn}] + (2\pi M_{jn} T^*)^{1/2}[\tfrac{2}{3}M_{jn}M_{nj}(a_{jr}^* - a_{nr}^*) + 4(M_{jn}^2 a_{jr}^* - M_{nj}^2 a_{nr}^*) + \tfrac{12}{5}M_{jn}^2(a_{jr}^* - a_{nr}^*) \\
&\quad - \tfrac{82}{15}(1-e_{nj})M_{nj}(M_{jn}a_{jr}^* + M_{nj}a_{nr}^*) - \tfrac{16}{15}(1+e_{nj})M_{jn}(M_{jn}a_{jr}^* + M_{nj}a_{nr}^*) + \tfrac{4}{5}(1-e_{nj})(1-2e_{nj})M_{jn}^2(a_{jr}^* - a_{nr}^*)]\} \\
&= \eta_n T^* \left\langle d_{nr}^* + T^* \left\{ \sum_{j=1}^{s} \left[\delta_{nj} + 6\left(\frac{\sigma_{nj}}{\sigma_j}\right)^3 (1+e_{nj}) g_{nj}^c \phi_j M_{jn} M_{nj} [\tfrac{4}{5} - \tfrac{2}{5}(1-e_{nj}) - \tfrac{4}{5}(1-e_{nj}^2)] \right] \frac{\partial \ln T^*}{\partial x_r^*} \right.\right. \\
&\quad \left.\left. + \sum_{j=1}^{s} \frac{3}{5} M_{jn} e_{nj}(e_{nj}-1) \left[\frac{n_n}{\rho_n T} \left(\frac{\partial \mu_n}{\partial n_j}\right)_{T, n_k \neq n_j} - \delta_{nj} \right] \frac{\partial \rho_j}{\partial x_r^*} \right\} \right\rangle,
\end{aligned} \tag{10}$$

where

$$d_{nr}^* = T^* \left\{ \frac{\rho_n}{\rho} \sum_{j=1}^{s} \left[\delta_{nj} + \frac{2\pi}{3} M_{nj}(1+e_{nj}) \left(\frac{\sigma_{nj}}{\sigma_j}\right)^3 n_j \sigma_j^3 g_c^{nj} \right] \frac{\partial \ln T^*}{\partial x_r^*} + \sum_{j=1}^{s} \frac{\rho_n}{\rho T} \left(\frac{\partial \mu_n}{\partial n_j}\right)_{T, n_k \neq n_j} \frac{\partial n_j}{\partial x_r^*} \right\}$$

is the dimensionless diffusion force in the radial direction under the condition of no external forces and mechanical equilibrium.

Of special interest is the case of a bidisperse particle-gas suspension turbulent flow consisting of normal and tagged particles, with the same mass m, diameter σ, coefficient of restitution e, and different number densities n_n and n_t, in a vertical riser. An approximate expression for the diffusion velocity of the normal particles in the radial direction, in the limit $\tau_{nj}^{(2)}/\tau_n^{(1)} \ll 1$, from Eqs. (7) and (10) can be obtained, correct to first order, as

$$v_{nr} = -\frac{\pi^{1/2}\sigma}{8\rho_{nr}(1+e)\phi g_c}\left(\frac{T}{m}\right)^{1/2}\left\langle \frac{\rho_t\rho_n}{n}\,\frac{\frac{n}{T}\frac{\partial\mu_n}{\partial n_n}-\frac{n}{T}\frac{\partial\mu_t}{\partial n_n}}{\frac{\rho_t}{T}\frac{\partial\mu_t}{\partial n_n}+\frac{\rho_n}{T}\frac{\partial\mu_n}{\partial n_n}} \times\left[1+\frac{\pi}{3}n\sigma^3(1+e)g_c\right]\frac{\partial \ln T}{\partial x_r}+\frac{\partial\rho_n}{\partial x_r}\right\rangle . \tag{11}$$

The first term in the angular brackets on the right-hand side accounts for the diffusion due to the radially nonuniform granular temperature distribution. This effect results in the migration of particles from the high-granular-temperature regions toward regions of lower granular temperature. The second term in the angular brackets describes the ordinary particle diffusive motion due to nonuniform spatial distributions of particle concentration. It can be concluded from Eq. (11) that the particle diffusivity

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

as well as the particle thermal diffusion coefficient

$$D_T = \left(\frac{\rho_{\rm tag}}{T}\frac{\partial\mu_{\rm nor}}{\partial n_{\rm nor}}-\frac{\rho_{\rm tag}}{T}\frac{\partial\mu_{\rm tag}}{\partial n_{\rm nor}}\right)\Bigg/\left(\frac{\rho_{\rm tag}}{T}\frac{\partial\mu_{\rm tag}}{\partial n_{\rm nor}}+\frac{\rho_{\rm nor}}{T}\frac{\partial\mu_{\rm nor}}{\partial n_{\rm nor}}\right)\times\left[1+\frac{\pi}{3}n\sigma^3(1+e)g_c\right]D,$$

increases with the square root of the granular temperature.

In order to evaluate the lateral particle diffusion velocity relative to the local mass mean velocity of the solid from Eq. (11), it is necessary to know the granular temperature of the normal particles. Using the modified form of the energy balance [16], which accounts for the forces exerted on the particles by the gas, for the normal particle assembly in a particle-gas suspension consisting of a binary mixture of normal and tagged particles, for which $n_t \ll n_n \approx n$, one can set up the balance of pseudothermal energy in the form

TABLE I. Parameters used in computing the particle diffusion coefficient.

Particle size (μm)	Solid volume fraction (%)	Granular temperature[a] (cm/s)^2	$T/T_{\rm cl}$
200	0.16	531	15
200	0.43	378	10
500	0.18	708	110
500	0.50	554	89
Solid density	1000 kg/m^3		
Pipe diameter	30.5 mm		

[a]Reference [11].

$$\frac{3}{2}\frac{\rm d}{{\rm d}t}\frac{T}{m} = \left\langle \frac{1}{\rho}\frac{T}{m}\frac{\partial}{\partial \mathbf{x}}\cdot(\rho\mathbf{v}_n)-\frac{1}{\rho}\frac{\partial}{\partial\mathbf{x}}\cdot\mathbf{q}_n+\frac{1}{\rho}\mathbf{v}_n\cdot\left(\frac{\partial}{\partial\mathbf{x}}\cdot\mathbf{P}\right)\right\rangle - \frac{1}{\rho}\gamma_n + \frac{1}{\rho}\left\langle -\mathbf{P}_n : \frac{\partial\mathbf{u}}{\partial\mathbf{x}} - \phi_n\rho_g[\beta_1 K_1(\phi_s) + \beta_2 K_2(\phi_s)|\mathbf{U}_n^R|]\overline{\mathbf{U}_n^{R'}\mathbf{C}_n}\right\rangle , \tag{12}$$

where q_n is the energy flux vector of the normal particles and γ_n is the rate of energy dissipation per unit volume of mixture due to inelastic collisions [16]. Note that, in evaluating these quatities, terms of $O(n_t/n_n)$ or higher can be neglected with respect to those of $O(n_n/n)$. Here, prime indicates the functuating velocity.

The term on the left-hand side of Eq. (12) presents the rate of change of the pseudothermal energy for an observer moving with the mean solid mixture velocity. The terms in the first set of angular brackets on the right-hand side represent the transport of the particle pseudothermal energy and the second term is the rate of energy dissipation due to inelastic collisions. The terms in the second set of angular brackets represent the rate of production of the pseudothermal energy at which particle pressure performs work on mean flow and the contribution from particle-turbulence interaction [11], respectively. A numerical solution of Eq. (12) has been presented for gas suspension flows of moderately dense solid spherical particles in vertical tubes elsewhere [31].

III. RESULTS AND DISCUSSION

In the preceding section, the particle diffusivity was found to increase with the square root of the particle granular temperature when the random component of the particle velocity is generated mainly by particle-particle collisions. In what follows, the proposed particle diffusivity is applied to predict lateral diffusion in fully developed dilute and relatively dense gas-particle flows in vertical pipes using the values of the local granular temperature predicted by Louge, Mastorakos and Jenkins [11]. Then the results using the present model are compared with the predictions of Govan, Hewitt, and Ngan [20] for dilute suspensions as the solids concentration tends toward zero. Table I represents the parameters used in computing the particle diffusion coefficient. Here $T/T_{\rm cl}$ represents the ratio of the particle granular temperature to the collisionless particle fluctuating velocity at the same

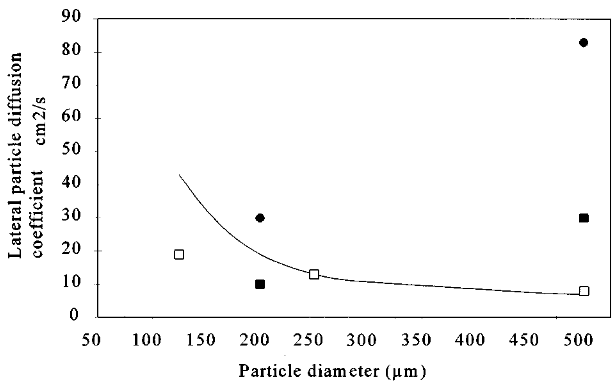

FIG. 1. Comparison of lateral particle diffusion coefficients predicted by Eq. (11) for the parameters given in Table I, with the model of Govan [18], which is shown by the solid curve. Symbols and conditions are as follows: □, the long-time particle diffusion coefficient for collisionless condition [18]; ●, dilute suspension in which the collisional contributions are dominant ($\phi = 0.18\%$); and ■, relatively dense suspension ($\phi = 0.5\%$).

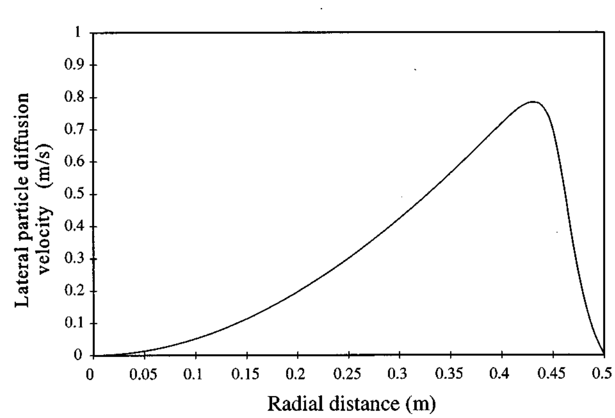

FIG. 2. Predicted lateral particle diffusion velocity due to the granular temperature gradient versus radial distance using the radial variation of particle volume fraction and the granular temperature for the fully developed gas solid flow in the vertical tube given by Pita and Sundaresan [34].

eddies' characteristic size l_e. The results of the calculation are shown in Fig. 1 as plots of the particle diffusion coefficient versus the particle size. For the conditions mentioned in Table I the hydrodynamic relaxation time is much larger than the relevant turbulent time scale. In this case, particles trajectories are relatively straight in spite of the gas turbulence. In their model Louge, Mastorakos, and Jenkins [11] assumed that the frequency of the particle displacements are controlled by particle-particle collisions, although the particle collision time is not much smaller than the hydrodynamic relaxation time. The particle collision time τ_c, particle viscous relaxation time τ_d, and the particle eddy interaction time τ_i are defined as

$$\tau_c = \frac{1}{\pi n \sigma^2 g_c T^{1/2}}, \quad \tau_i = \frac{l_e}{\Delta w^T}, \quad \tau_d = \frac{\rho_n^m \sigma}{\rho_g \beta_2 K_2(\phi_s) \Delta w^T}. \tag{13}$$

The predicted particle diffusivity using Eq. (11) versus the particle diameter for the parameters given in Table I is illustrated in Fig. 1. It is worth pointing out that for the cases mentioned in Table I, the contact value of the equilibrium radial distribution function g_c approaches unity. Moreover, the Govan predictions for collisionless conditions are illustrated by solid curve in Fig. 1. The predicted particle diffusion based on the present theory shows the opposite trend compared to the collisionless particle diffusion coefficient of Govan, supporting the idea that inertia tends to increase particle diffusivity. However, this qualitative variation is in slight disagreement with that indicated by Meek and Jones [32]. In this connection, it is relevant to refer to the observation by Halder and Basu [33] that the smaller glass beads result in higher rates of mass transfer from a large naphthalene particle to the fast bed of fines. The predicted particle diffusivity is also found to decrease with the solid volume fraction. For the lack of experimental data, one cannot claim an accuracy of the above-estimated diffusion coefficient.

In order to estimate the particle diffusion velocity caused by the radially nonuniform granular temperature, the turbulent, fully developed flow of a moderately dense suspension in a vertical pipe that has been simulated by Pita and Sundaresan [34] is considered. Particles with density $\rho_s = 1500\ \mathrm{kg/m^3}$ and diameter $\sigma = 70\ \mu\mathrm{m}$ are transported in air with density $\rho_g = 1.2\ \mathrm{kg/m^3}$ and kinematic viscosity $\nu_g = 15 \times 10^{-6}\ \mathrm{m^2/s}$. The turbulent length scale of gas in the fully developed pipe flow may be approximated by $l_e/R \approx 0.2$ [35]. Here R is the diameter of the pipe. For these conditions the ratio of the particle relaxation time to the turbulent time scale of gas τ_l is of order 10^{-1}, indicating that the particles follow the fluid motion in the dilute suspensions as the solid concentration tends toward zero. In a moderately dense suspension such as that considered by Pita and Sundaresan [34], the ratio of the particle collision time to the particle eddy interaction is very small, suggesting that the frequency of the particle displacements are controlled by their rate of collision with neighboring solid particles. Therefore, the analysis that is presented in Sec. II can be used here to estimate the particle diffusion velocity, although it contains assumptions that are strictly valid only for large particles. Pita and Sundaresan presented three different possible solutions for the steady-state, fully developed flow when the values of the solid flux and gas velocity are selected as $25\ \mathrm{kg/m^2 s}$ and 8 m/s, respectively. Here the solution that was presented in Fig. 4 of their paper is considered. Figure 2 in this paper presents the radial variation of the particle diffusive flux due to the temperature gradient. As a tentative attempt at the contact value of the equilibrium radial distribution function g_c, use is made of the *ad hoc* model by Carnahan and Starling [36], which is in almost exact agreement with the numerical molecular-dynamics calculations for values of solid volume fraction up to about 0.5. The expression for the chemical potential consistent with Carnahan and Starling's approximation can be obtained from Eq. (8).

As can be seen from Fig. 2, the diffusion velocity increases slowly with radial distance until $R = 0.43$ m. At this point, the velocity begins to decrease sharply due to the sharp decrease in the granular temperature and approaches zero at the wall. These results represent the effect of radially nonuniform granular temperature distributions, namely, that particles migrate from the high-granular-temperature regions toward regions of lower granular temperature, say, the wall region. The magnitude of the radial diffusion velocity exceeds 0.8 m/s at $R=0.43$ m, which indicates that some key physics must be missing in their model.

Although the effect of the lateral particle diffusive flux in gas-solid suspensions, in which solid clusters are present, is a question of fundamental interest, Dasgupta, Jackson, and Sundaresan [12] have demonstrated that it can be neglected without any significance consequences. The present study, however, provides a context in which to investigate the problem of mass transfer in a mixtures of polydisperse particles entrained by the gas under conditions in which fractal-like solid structures are present [37].

IV. CONCLUSION

A kinetic-type theoretical approach is developed for the transport processes involved in turbulent, fully developed, confined vertical flow of a moderately dense mixture of gas and particles with a narrow particle size distribution, in the limit $\tau_c/\tau_d \ll 1$. Here τ_c is the particle mean free time and τ_d is the particle hydrodynamic relaxation time. Analytical relations for flow-induced particle diffusivity are developed. There are two mechanisms for particle diffusion mass flux; the particle ordinary diffusion and the particle thermal diffusion due to the radially nonuniform granular-temperature distributions. The latter effect tends to move particles from the high-granular-temperature regions toward regions of lower granular temperature at the wall. The particle diffusivity coefficients are found to increase with the square root of the particle granular temperature when the collisional contributions are dominant.

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