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**ON THE VISCOSITY OF A MIXTURE OF PARTICULATE
SOLIDS AND A FLUID CONTINUUM**

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On the Viscosity of a Mixture of
Particulate Solids and a Fluid Continuum

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Summary

Developing models capable of describing various multiphase flow regimes has attracted considerable attention due to their relevance to applications in many chemical and transport processes. These existing theories may be roughly classified into two categories. In one class, the basic conservation laws are postulated and the ideas in continuum mechanics are used to arrive at appropriate constitutive relations. In the other class, averaging techniques are used to derive the fundamental balance laws. Whichever approach is used, constitutive relations are needed. These would supply connections among kinematic, mechanical, and thermal fields that are compatible with the balance laws and, in conjunction with the balance laws, provide a theory.

Works related to fluid-solids flows customarily assume that the form of the stress tensor for the solid phase is analogous to that of a linearly viscous (Newtonian) fluid. The shortcomings of this approach have been discussed elsewhere [Massoudi and Boyle (1988)]. In our approach, we consider the mixture to be composed of a granular material, whose constitutive equation has been properly formulated, and a Newtonian fluid. Both the traditional approach and our approach model the solid particles as a flowing continuum, thus a viscosity coefficient appears in the constitutive equation for the stress tensor for the solid. Physically, as in the case of a fluid, this viscosity is related to the resistance of the material to flow. Although this term is present in many of the two-phase formulations, neither its significance nor its meaning has been explored, especially whether it can be measured and quantified.

The problem of the theoretical determination of the viscosity coefficient of an infinitely dilute suspension consisting of a Newtonian incompressible fluid and rigid spherical particles was studied by Einstein (1956). He assumed that the volume fraction is very small compared with unity, indicating that the particles are far apart from each other and are not interacting with each other. Furthermore, he also assumed that the flow around each particle is described by the Navier-Stokes equation with negligible inertia (the Stokes equation). Therefore, as the solids concentration increases, i.e., as we approach a dense-phase flow, the above equation no longer holds. This has been experimentally verified. This regime is of significance in both the pneumatic transport and hydraulic transport of coal particles.

Given the fact that, in this dense-phase regime, the particles are assumed to behave like a continuum with coefficient of viscosity μ_s , it is not very clear how the effective viscosity of the mixture, i.e., μ^* , is related to μ_s and μ_f . Experiments with conventional viscometers are generally designed to measure the effective viscosity μ^* .

In this work, the theory of interacting continua is used to formulate the governing equations of motion for each component and the mixture as a whole. The fluid is assumed to be an incompressible viscous fluid and solid particles are treated as granular materials. Appropriate constitutive relations for stresses are proposed. Based on these, the stress tensor for the mixture is obtained, and in some ideal experiment where the velocities of

the two phases are equal, a relationship among the viscosities of the two components and the viscosity of mixture exists. Based on these observations, simple experiments can be devised to obtain the viscosity of the solid particles. In Section 1, the importance of multicomponent flows is discussed. Section 2 provides a survey of literature on the stress tensors in two-phase flows. In Section 3, the basics of the theory of interacting continua are discussed and equations of motion for the mixture are derived. In the final section, a few experiments are suggested that would help in measuring the material properties of the mixture.

1.0 Introduction

Multiphase flows have increasingly become the subject of considerable attention because of their importance in many industrial applications, such as fluidized beds, pneumatic transport of solids, coal combustion, etc. For example, the flow behavior of gas-particle mixtures in transport lines has been of interest in chemical processes for many years. In general, empirical relations that predict the flow and pressure drop in such processes have been developed for specific ranges of solids and gas properties as well as for various geometries. Also, a great deal of research has been devoted to the use of coal-based slurries as retrofit fuels. Historically, coal-oil mixtures have received the most attention; their combustion characteristics and the flow or handling properties of these fuels have formed the basis of many research programs. Another area of research emphasis is that of fluidization, generally applicable to any particulate material in the size range between 10 μm (diameter) and 3 mm, which includes two classes, powders and granular solids. A powder is composed of particles up to 100 μm , with further subdivision into ultra-fine (0.1 to 1.0 μm), superfine (1 to 10 μm), or granular (10 to 100 μm) particles. A granular solid consists of material ranging from about 100 to 3,000 μm (Botterill 1975).

Because the fluidized particles are separated from one another by the fluid, intimate contact between the particles and the fluid is achieved. Hence, a bed of fluidized particles is an excellent medium for the transfer of heat or mass between solids and fluids. Perhaps the first major successful application of the gas fluidized-bed technique was the catalytic cracking process. Many metallic ores are roasted in fluidized beds for the removal of sulfur. Fluidized-bed drying has been applied, not only to granular materials, but also to pastes, suspensions, and molten materials. For instance, in manufacturing certain chemicals, the usual operations of evaporation, crystallization, filtration, drying, and pulverization are replaced by a single process of drying in a fluidized-bed. The fluidization phenomenon has also been used for the transportation of powdered solids. The mixing of particles in a normally fluidized bed is rapid and thorough, so that a uniform temperature is quickly achieved and easily maintained.

Another area of interest involving flow of particles is the structural design of bins and silos. In processing minerals, in many processes such as the transport and combustion of coal, and in preventing or protecting against natural phenomena, such as avalanches or debris flows, it is essential to understand the factors governing the packing and flow of powders and particulate matters. Flowing granular materials can represent a limiting case of two-phase flow at high solids concentration and high solid-to-fluid density ratios. Many situations, such as discharge through bin outlets, flow through hoppers and chutes, pneumatic transport of coal, and fluidized beds require information on material properties of the granular media, flow patterns, concentration profile, etc. These and many other applications of the multicomponent flows have motivated many researchers of the last two decades to study these phenomena theoretically to better understand its basis. Consequently, the analytical modeling of the multiphase flows has received much attention (Massoudi 1986).

Much work has been reported in the literature about the mathematical modeling of multicomponent systems. The two foremost approaches use either volume-averaging technique or mixture theory. In the first approach, used by many authors (Anderson and Jackson 1967; Drew and Segel 1971a,b; and Ishii, 1975), point-wise equations of motion, valid for a single fluid or a single particle, are modified to account for the presence of the other components and the interactions between components. These equations are then averaged over time or some suitable volume, which is large compared with some characteristic dimension (for example, particle spacing or the diameter of solid particles) but small compared to the dimensions of the whole system. The averaging process yields several terms, which represent the interactions between the constituents. Constitutive relations to represent these terms, as well as for the stress tensors for each constituent, are then required.

Anderson and Jackson (1967) were among the first workers who applied this method. They obtained a set of equations describing the fluid mechanical aspects of fluidized beds. Simple constitutive relations, necessary to close the system of equations, were proposed. They assumed that the form of

the stress tensor for the solid constituent is analogous to that of a Newtonian fluid, with the coefficients of viscosity depending on the local mean voidage, ϵ . Drew and Segel (1971a,b) developed a different averaging technique, and obtained equations of motion for a broad class of two-phase flow conditions. They also considered the specific application of these equations to fluidized beds. The constitutive equations they assumed for the stress relations were much the same as those used by Anderson and Jackson (1967).

The second approach used in the mathematical modelling of multi-component systems is mixture theory. This theory traces its root to the work of Fick (1855) and was first given a rigorous mathematical structure by Truesdell (1957). It is a means for studying the interaction between several constituents by generalizing the equations and principles of the mechanics of a single continuum. The fundamental assumption in this theory is that at any instant of time, every point in space is occupied by one particle from each constituent. That is, each constituent is assumed to be homogenized to occupy the volume of the mixture. The historical development and the details of this theory are given in the review articles by Bowen (1976), Atkin and Craine (1976), Bedford and Drumheller (1983), and Passman, et al. (1983). Several articles that have relevance to mixture theory appear as appendices to the recent edition of Rational Thermodynamics by Truesdell (1984).

El-Kaissy (1975) adopted the mixture theory approach and derived constitutive relations that were properly frame-invariant. An important part of his work was the emphasis laid upon the principle of material frame-indifference and the derivation of some objective combinations of the independent variables to be used in the constitutive equations. Perhaps, a shortcoming of this work, as well as the previous works by Anderson and Jackson (1967), and Drew and Segel (1971), was the assumption that the stress tensor for the solid phase had the same basic structure as that of a linearly viscous Newtonian fluid. The validity and appropriateness of this assumption will be discussed in the next section.

2.0 Survey of Literature on the Stress Tensors in Two-Phase Flows

In the two-phase flow and fluidized-bed literature (Anderson and Jackson, 1967, Drew and Segel 1971, and Homsy, et al. 1980), the customary assumption is that the stress tensors for the two constituents have the same form as the stress tensor of a Newtonian fluid, where the coefficients of viscosities are allowed to depend on the voidage ϵ ($= 1 - v$).

Murray (1965) in his paper on the mathematics of fluidization assumed the following forms for the stress tensors:

$$^f T_{ij} = -p^f \delta_{ij} + (\zeta_f - \frac{2}{3} \mu_f) \delta_{ij} \frac{\partial v_k}{\partial x_k} + \mu_f \left(\frac{\partial v_i}{\partial x_j} + \frac{\partial v_j}{\partial x_i} \right), \quad (2.1)$$

$$^s T_{ij} = -p^s \delta_{ij} + (\zeta_s - \frac{2}{3} \mu_s) \delta_{ij} \frac{\partial u_k}{\partial x_k} + \mu_s \left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right), \quad (2.2)$$

where p^f is the fluid pressure, p^s the solid pressure (a form of collision pressure), μ_f and μ_s are the shear viscosities and ζ_f , ζ_s the bulk viscosities of the fluid and solids, respectively. Subscripts (or superscripts) f and s refer to the fluid and solid phases, respectively. Murray (1965) assumed the solid pressure, p^s , is in general negligible, since in fluidized beds practically no noise is noticeable (which indicates that the collision of particles is rare). This point has been questioned considerably [see for example Pai (1973)]. In the Appendix of his paper, Murray (1965) showed that reasonable forms for μ_s and ζ_s are

$$\mu_s = \mu_f A \phi_s, \quad (2.3)$$

$$\zeta_s = \mu_f B \phi_s^3, \quad (2.4)$$

where A and B are constants of $O(1)$ and

$$\phi_s = \frac{v}{v_s - v}, \quad (2.5)$$

where v_s is the saturation value of v (when the particles are not fluidized).

Anderson and Jackson (1967) and Jackson (1971) proposed the following forms for the stress tensors

$$T_{ik}^f = -p^f \delta_{ik} + \lambda_f(\epsilon) \frac{\partial v_m}{\partial x_m} \delta_{ik} + \mu_f(\epsilon) \left[\frac{\partial v_i}{\partial x_k} + \frac{\partial v_k}{\partial x_i} - \frac{2}{3} \delta_{ik} \frac{\partial v_m}{\partial x_m} \right], \quad (2.6)$$

$$T_{ik}^s = -p^s \delta_{ik} + \lambda_s(\epsilon) \frac{\partial u_m}{\partial x_m} \delta_{ik} + \mu_s(\epsilon) \left[\frac{\partial u_i}{\partial x_k} + \frac{\partial u_k}{\partial x_i} - \frac{2}{3} \delta_{ik} \frac{\partial u_m}{\partial x_m} \right], \quad (2.7)$$

where p^f and p^s are local mean pressures for the fluid and particle phases, λ_f and λ_s are effective bulk viscosities, μ_f and μ_s are effective shear viscosities, and $\underline{v}$ and $\underline{u}$ are the velocities of the fluid and particle phases, respectively. While Anderson and Jackson (1967) and Jackson (1971) mentioned that they made "the simplest possible assumption" in proposing constitutive equation of type (2.1), they admitted that it would be very unlikely that the rheological properties of the two phases were accurately represented by Equations (2.1) and (2.2) at very low rates of shear. They justified their assumption by referring to experiments that indicated that the behavior of fluidized suspensions in Couette viscometers was approximately Newtonian at higher rates of shear. Many attempts have been made to calculate the viscosity of fluidized beds experimentally and theoretically. The experimental measurements have given values of viscosity in the range of 1 to 20 poise. It should be mentioned that both the experimental and theoretical values obtained for the viscosity of the bed agree within an order of magnitude and are comparable to the viscosity of highly viscous fluids (the viscosity of glycerine being about 10 poise at ambient conditions) [Davidson et al. (1977)].

Pai (1971) in his study of a mixture of a gas and small solid particles assumed that the solid particles behave like a pseudo-fluid such that

$$T_{ij}^s = -p_s \delta_{ij} + \mu_s \left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right) + \lambda_s \left(\frac{\partial u_k}{\partial x_k} \right) \delta_{ij}, \quad (2.8)$$

where p_s is the partial pressure of the particles in the mixture, μ_s is the coefficient of shear viscosity and λ_s is the second or bulk coefficient of viscosity of the particles. The restrictions on μ_s and λ_s were given as $\mu_s \geq 0$ and $2\mu_s + 3\lambda_s \geq 0$. Pai (1971) used a similar constitutive equation for the gas. However, he did not assume that the partial pressures are determined from the solution to the problem; instead, for the gas phase, he assumed the perfect gas law

$$p_g = R \bar{\rho}_g T_g = R (1-v) \rho_g T_g = (1-v) p, \quad (2.9)$$

where p_g is the partial pressure of the gas in the mixture, T_g is the partial temperature of the gas, and R is the gas constant. Furthermore, he assumed that the total pressure of the mixture p is also given by the perfect gas law:

$$p = \rho_g R T_g \quad (2.10)$$

(It is not clear why the density and temperature of the mixture are not used in the above equation.) He defined the total pressure of the mixture as the sum of the partial pressures p_g and p_s of the gas and solid phases, respectively,

$$p = p_g + p_s. \quad (2.11)$$

Comparing Equations (2.9) and (2.11), indicates that

$$p_s = v p. \quad (2.12)$$

This indicates that the partial pressure of the solid phase is proportional to the solids volume fraction. If v is much smaller than one, then p_s can be neglected. On the basis of the above assumptions Pai (1971) disagrees with Murray's (1965) assumption that p_s is negligible for finite values of v . However, it is not evident that the assumptions made by Pai (1971) are indeed the right ones to make.

For the equation of state of solid particles, Pai (1971), assumed

$$\rho_s = \text{Const} \quad (2.13)$$

where the density of the mixture is given by

$$\rho = v \rho_s + (1-v) \rho_g \quad (2.14)$$

Pai (1971) also considered the conservation of energy for each phase. In his theory, the eleven governing equations are the conservation of mass for each phase (2), the conservation of linear momentum for each phase (6), conservation of energy for each phase (2), and equation of state for the mixture pressure. Since he considered the density of solid particles ρ_s as a constant, the unknowns are the velocities of both phases u, v (6), temperatures of the two phase T_g, T_p (2), density of the gas ρ_g (1), the solids volume fraction, v (1), and the pressure p (1). Later, Pai (1973) derived, basically, the same set of equations and conclusions from simple arguments based on kinetic theory.

Drew and Segel (1971a, 1971b) proposed stress relating for the two phases very much the same as those of Jackson (1971). They adopted the "hypothesis of phase separation,"* which indicates that the internal state

*This so-called "hypotheses" or "principle" which is often attributed to Drew and Segel (1971) was actually suggested earlier by Adkins (1963):

"It is assumed that within the mixture, the properties of any given component are defined by means of constitutive equations relating the partial stress tensor, the density (or concentration) and the kinematic quantities, such as velocity gradients, acceleration gradients and their successive time derivatives for that component."

Adkins (1963) also laid the foundation for a more general approach:

"It would be possible, for example, to postulate that the partial stress tensor for a given fluid should be dependent not only upon the kinematical quantities defined for the fluid itself, but also upon the concentration, velocity gradients, acceleration gradients, and time derivatives of these quantities defined, at the point under consideration, for the other fluids in the mixture. This would give an explicit coupling of mechanical properties."

of one constituent is not related to the internal state of the other constituent. According to this assumption, for example, the stress tensor for the solid particles would only depend on variables such as ρ_s , λ_s , μ_s , $\text{grad } \underline{u}_s$, etc. However, they proposed that surface variables (for example, I , the interactive force) may depend on quantities from both phases. They mentioned that the pressure term p^s in Equation (2.2) is due to a reaction force to deformation, and they claimed that the term $(p^s - p^f)$ represents a pressure due to collisions.

A more general and rational way of postulating the stress tensors was presented by El-Kaissy (1975). He used the principles of equipresence and frame-indifference in proposing the constitutive relations (Truesdell and Noll 1965); however, his theory retained a shortcoming namely that the structure of the stress tensor for the solid is the same as that of a Newtonian fluid. The stress relations for the two phases were modified and presented in Homsy, El-Kaissy, and Didwania (1980) as

$$^sT_{ij} = -p^s (1-\epsilon) \delta_{ij} + \sum_{\gamma=1}^2 [\beta^{\gamma 1} D_{nn}^{\gamma} \delta_{ij} + 2\eta^{\gamma 1} (D_{ij}^{\gamma} - \frac{D_{kk}^{\gamma}}{3} \delta_{ij})] , \quad (2.15)$$

and

$$^fT_{ij} = -p^f \epsilon \delta_{ij} + \sum_{\gamma=1}^2 [\beta^{\gamma 2} D_{nn}^{\gamma} \delta_{ij} + 2\eta^{\gamma 2} (D_{ij}^{\gamma} - \frac{D_{kk}^{\gamma}}{3} \delta_{ij})] , \quad (2.16)$$

where

$$^1D_{ij} = \frac{1}{2} \left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right) , \quad (2.17)$$

and

$$^2D_{ij} = \frac{1}{2} \left(\frac{\partial v_i}{\partial x_j} + \frac{\partial v_j}{\partial x_i} \right) . \quad (2.18)$$

That is, they assumed that the individual phases behave like viscous fluids, with $^aD_{ij}$ and an isotropic term being the only variables in which the stresses of each phase would depend. The reason for choosing the above form of the isotropic pressure was given as: "... the isotropic term gives the

pressure measured over an element of volume of the mixture and not of the single phase ...". The material functions $\beta^{\gamma a}$ and $\eta^{\gamma a}$, in general, depend on the scalar invariants of the objective quantities appropriate for the formulation of this theory. This dependence would inevitably lead to nonlinear material response. However, since Homsy et al. (1980) were primarily interested in a linear instability analysis of a homogenous fluidized bed, they treated these material functions as dependent on p_s , p_f , and ϵ .

In their stability analysis of a fluidized bed, Garg and Pritchett (1975) assumed that the gas behaves like an inviscid fluid

$$\underline{T}^f = -p \underline{1} , \quad (2.19)$$

where p denotes the thermodynamic pressure and depends on ϵ , ρ_f , and gas temperature T_f . They also assumed that the solid particles are incompressible ($\rho_s = \text{constant}$) and that the particle assemblage behaves as a Newtonian fluid (Equation 2.1). They indicated that the assumption of Newtonian behavior is to be adopted as a first approximation and its validity is very much in question in situations (e.g., boundary layers) where the rotation of solid particles plays an important role. They also introduced interstitial gas pressure p^e and effective solid pressure p^{se} such that

$$p = \epsilon p^e , \quad (2.20)$$

and

$$p^s = (1 - \epsilon) p^{se} . \quad (2.21)$$

Garg and Pritchett (1975) related these quantities through an empirically determined function $f(\epsilon)$, such that

$$p^{se} = p^e + f(\epsilon) \quad (2.22)$$

where, " $f(\epsilon)$ physically corresponds to the normal component of particle-particle interactions." It was also assumed that for incompressible particles $f(\epsilon)$ decreases monotonically with ϵ and has the following limits:

$$\begin{aligned}\epsilon \rightarrow 0 & : f(\epsilon) \rightarrow \infty, \\ \epsilon \rightarrow 1 & : f(\epsilon) \rightarrow 0.\end{aligned}\tag{2.23}$$

Drew (1976) studied some basic two-phase flows where he considered the effects of lift and Brownian motion in addition to the usual effects such as drag, virtual mass, etc., which are considered in the interaction between the two phases. He introduced two principles*: the principle of local dependence on dispersed (solid) phase variables, and the principle of low concentration limits.** The former states that non-local effects (for example, gradients) of dispersed phase variables should be omitted from constitutive equations. The latter principle states that when the dispersed phase is sufficiently dilute, the mixture should behave as if it were made up of only the continuous (fluid) phase. According to this principle, as $\epsilon \rightarrow 1$ (very low concentration of solid particles), the equations of motion for the continuous phase should reduce to the appropriate equations for an incompressible viscous fluid. This principle is often used in two-phase flows, although it is not always stated explicitly. It should be remembered that this is only an assumption which was suggested much earlier by Green and Adkins (1964).

One of the severe consequences of the "principle of local dependence" is that the particle stress tensor $\underline{S}_I$ cannot depend on the gradient of the solid velocity. Drew (1976), furthermore, assumed that

*It would be more appropriate to call these "principles" assumptions.

**Interestingly, a similar idea was put forward earlier by Green and Adkins (1964). In their formulation for a mixture of non-Newtonian fluids, they state: "Also, in the absence of one fluid the constitutive equation for the other must reduce to the usual one for that fluid alone."

$$s_I = -p \underline{1} , \quad (2.24)$$

where p is the averaged fluid pressure.

In their study on the formation of bubbles in gas particulate fluidized beds, Fanucci, et al. (1979) assumed very simple constitutive equations for s_I and f_I . They proposed the following:

$$f_I = -p^f \underline{1} , \quad (2.25)$$

$$s_I = -p^s \underline{1} , \quad (2.26)$$

where p^f and p^s related through

$$p^s = p^f + f(\epsilon) . \quad (2.27)$$

Later, Fanucci, et al. (1981) generalized their previous study by including the effects of shear in their constitutive equations. They assumed that

$$T_{ik}^f = -p^f \delta_{ik} + \mu_f \left[\frac{\partial v_i}{\partial x_k} + \frac{\partial v_k}{\partial x_i} - \frac{2}{3} \frac{\partial v_m}{\partial x_m} \delta_{ik} \right] , \quad (2.28)$$

$$T_{ik}^s = -p^s \delta_{ik} + \mu_s(\epsilon) \left[\frac{\partial u_k}{\partial x_k} + \frac{\partial u_k}{\partial x_i} - \frac{2}{3} \frac{\partial u_m}{\partial x_m} \delta_{ik} \right] , \quad (2.29)$$

where the pressures are once again related through Equation (2.27), μ_f is the constant fluid viscosity, and μ_s is assumed to be a function of ϵ . Following a suggestion by Mutsers and Rietema (1977), Fanucci, et al. (1981) assumed that the particle viscosity decreases exponentially with an increase in fluid voidage. Accordingly, they assumed

$$\mu_s(\epsilon) = \mu_{s0} e^{\left[\frac{1}{1-\epsilon_0} - \frac{1}{1-\epsilon} \right]} , \quad (2.30)$$

where the subscript zero refers to the state of minimum fluidization. When Equation (2.25) and (2.26) or (2.28) and (2.29) were substituted in the

equations of motion, they were multiplied by ϵ and $(1 - \epsilon)$, respectively, prior to being operated by the divergence operator.

Ahmadi (1980) studied the mechanics of saturated granular materials and derived constitutive relations for the stress tensors $\underline{T}^S$ and $\underline{T}^f$. For the case of a mixture of an incompressible fluid and incompressible grains, he showed that

$$T_{kl}^f = (-\pi^f + \lambda^f v_{i,i}) \delta_{kl} + \mu^f (v_{k,l} + v_{l,k}) , \quad (2.31)$$

and

$$\begin{aligned} T_{kl}^S = [2\alpha v_{v,jj} - a_0 v^2 + \alpha v_{,j} v_{,j} + W_0 + \lambda^S u_{i,i}] \delta_{kl} \\ - 2\alpha v_{,k} v_{,l} + \mu^S (u_{k,l} + u_{l,k}) , \end{aligned} \quad (2.32)$$

where α , a_0 , and W_0 are assumed to be constants. His approach to derive Equation (2.32) was based on ideas of Goodman and Cowin (1971, 1972) who proposed continuum theories for granular materials. Specifically, Ahmadi (1980) assumed that the free energy of the granular media was given by

$$\rho^S \psi^S = [\alpha v_{,k} v_{,k} + a_0 v^2] + W_0 . \quad (2.33)$$

Later, Ahmadi (1982) presented a continuum theory for two-phase media in which the fluid was assumed to be a micropolar fluid [cf. Eringen (1966)] and the particles were assumed to behave like a granular media. Ahmadi (1982) postulated the following constitutive relations:

$$\begin{aligned} T_{kl}^f = (-\pi^f + \lambda^f v_{i,i}) \delta_{kl} + \mu^f (v_{k,l} + v_{l,k}) \\ + K^f (v_{l,k} - \epsilon_{klr} \Lambda_r^f) \end{aligned} \quad (2.34)$$

$$T_{kl}^S = (-\pi^S + \lambda^S u_{i,i}) \delta_{kl} - 2\alpha(v) \Lambda_{i,k} \Lambda_{i,l} - \gamma(v) \phi_{j,k}^S \phi_{j,l}^S + \mu^S (u_{k,l} + u_{l,k}) + K^S (u_{l,k} - \epsilon_{klr} v_r^S) \quad (2.35)$$

where π^f and π^S are the thermodynamic pressures associated with the two phases, ϵ_{ijk} is the alternating tensor, Λ_i is the micro-rotation vector, ϕ_i is the micro-rotation vectors and K^f , K^S , λ are additional material properties. For incompressible grains, Ahmadi (1982) showed that Equation (2.35) reduces to

$$T_{kl}^S = [2\alpha v_{i,i} + \frac{d}{dv} (\alpha v) v_{i,i} - \frac{v^2}{2} \frac{d}{dv} (\frac{\gamma}{v}) \phi_{m,i}^S \phi_{m,i}^S + \lambda^S u_{i,i}] \delta_{kl} - 2\alpha v_{i,k} v_{i,l} - \gamma \phi_{m,k}^S \phi_{m,l}^S + \mu^S (u_{k,l} + u_{l,k}) + K^S (u_{l,k} - \epsilon_{klr} \Lambda_r^S) \quad (2.36)$$

In his paper on multiphase mixture theory for fluid-particle flows, Nunziato (1983) discussed the dilemma of the closure problem for two phase flows. He indicated that, physically, p_f and p_s are the constitutively indeterminate pressures associated with the incompressibility of each phase and π is an additional constraint if the mixture is to remain saturated for all time (p_s , p_f , and π mathematically represent Lagrange multipliers). Nunziato (1983) referred to π as the interface pressure, since it acts at the interface between the particulate and continuous phases. He then assumed [cf. Stuhmiller (1977)] that the interface pressure is associated with the pressure distribution over a particle, i.e., it represents the integrated average of the pressure distribution over a particle. For example, the pressure distribution over an isolated spherical particle in an inviscid fluid is given by

$$\pi = p_f + \omega (v_f - v_s) \cdot (v_f - v_s) \quad (2.37)$$

Referring to $(\pi - p_s)$ and $(\pi - p_f)$ as diffusive pressures, and using the restrictions imposed on the constitutive equations by the dissipation inequality, Nunziato showed that when the mixture is at rest ($v_f = v_s = 0$) and the temperature is uniform ($\nabla \theta = 0$),

$$p_f = \pi , \quad (2.38)$$

$$p_s = p_f + \beta_s , \quad (2.39)$$

where

$$\beta_s = \rho_s \left(\frac{\partial \psi_s}{\partial \phi_s} \right) , \quad (2.40)$$

where ψ_s is the free energy associated with the solid phase. This indicates that even when the two phases are at rest, the pressures are not equal. Therefore, Nunziato (1983), referred to β_s as the equilibrium diffusive pressure, which accounts for interparticle contact forces in mixtures where the particles concentration is very high. For dilute suspensions, he indicated that β_s is related to the Brownian motion of particles.

To model the stress tensor of each phase, Nunziato (1983) assumed

$$\underline{\tau}_a = - \phi_a p_a \underline{1} + \underline{\tau}_a , \quad (2.41)$$

where $\underline{\tau}_a$ is the extra stress associated with phase a. He then assumed

$$\underline{\tau}_f = 2(1 - \phi_s) \mu_f \underline{D}_f , \quad (2.42)$$

$$\underline{\tau}_s = 2\phi_s \mu_s \underline{D}_s , \quad (2.43)$$

where μ_f and μ_s are the constant fluid and solid viscosities, respectively. For the isothermal flows of dilute suspensions, he proposed

$$p_s - p_f = \beta_s - (\lambda_f + \lambda_s)(1 - \phi_s) \operatorname{tr} D_f + \lambda_s \operatorname{grad} \phi_s \cdot (\underline{v}_f - \underline{v}_s) + \omega_f (\underline{v}_f - \underline{v}_s) \cdot (\underline{v}_f - \underline{v}_s), \quad (2.44)$$

where λ_f and λ_s are the bulk viscosities. It should be noted that when both phases are at rest, the above Equation reduces to Equation (2.39). Furthermore, for a dilute suspension ($\phi_s \ll 1$) where the viscous forces in the fluid and the inertial forces in both phases are negligible, Nunziato (1983) showed that

$$\operatorname{grad} (\phi_s \beta_s) = \phi_s \gamma_s \operatorname{grad} c_s, \quad (2.45)$$

where c_s is the chemical potential of the particles per unit mass. To derive the above equation, he neglected effects such as lift forces, dynamic pressure forces, etc. Nunziato (1983) defined a Brownian diffusivity λ as

$$\lambda^* = \frac{1}{\alpha_1} \frac{\partial (\phi_s \beta_s)}{\partial \phi_s}, \quad (2.46)$$

where

$$\alpha_1 = \frac{9 \mu_f (1 + 5.5 \phi_s)}{2a^2}, \quad (2.47)$$

in the limit of low concentration. Nunziato (1983) then showed that using Equation (2.45) in Equation (2.46) gives

$$\lambda^* = \frac{2a^2 (1 - 5.55 \phi_s)}{9\mu_f} \phi_s \gamma_s \frac{\partial c_s}{\partial \phi_s}. \quad (2.48)$$

Using results from the statistical-mechanical theory of dilute solutions [Batchelor (1976)], Nunziato (1983) showed

$$\lambda^* = \frac{\kappa \theta}{6\pi \mu_f a} (1 + 1.45 \phi_s). \quad (2.49)$$

In the above equation, κ is Boltzman's constant, a is the radius of a particle, and α_s is related to Stokes drag. Nunziato (1983) defined the mixture pressure and viscosity as

$$p = (1-\phi_s) p_f + \phi_s p_s , \quad (2.50)$$

$$\mu = (1-\phi_s) \mu_f + \phi_s \mu_s . \quad (2.51)$$

Gidaspow and his co-workers (1983, 1986) in their formulation of fluidization in two-dimensional beds, in addition to selecting the isotropic parts of stress tensors similar to those of Equations (2.15) and (2.16), assumed the constitutive equation for the normal component of the solid stress s_τ is of the form

$$s_\tau = \tau(\epsilon) . \quad (2.52)$$

It should be noted that they assumed the fluid to be an inviscid fluid, thus the only term appearing in the fluid stress tensor is one proportional to p^f . Furthermore, they also assumed that

$$p^f = p^s = p . \quad (2.53)$$

Applying the chain rule to Equation (2.52),

$$\frac{\partial \tau}{\partial y} = \frac{\partial \tau}{\partial \epsilon} \frac{\partial \epsilon}{\partial y} , \quad (2.54)$$

where the y -direction is assumed to be the vertical direction. The particle-to-particle interaction coefficient (or what is referred to by some researchers as the modulus of elasticity (Gidaspow 1986a) was defined to be

$$G(\epsilon) = - 10^{-8.76 \epsilon + 5.43} \frac{N}{m^2} . \quad (2.55)$$

Gidaspow (1986a) explains: "This term becomes of numerical significance only when the void fractions go below the minimum fluidization void fraction. It also helps to make the system numerically stable, because it

converts the imaginary characteristics into real values. For some calculations, it was necessary to adjust this stress to prevent the void fraction from reaching impossibly low values."

It should be pointed out that the Equation (2.55) is based on the experimental results of Rietema and Mutsers (1973), who studied the effect of interparticle forces on the expansion of a homogeneous gas-fluidized bed. In a later work, Mutsers and Rietema (1977) indicated that the experimental data reported earlier by Rietema and Mutsers (1973) contained an error, which was subsequently rectified. It appears, however, that studies performed by Gidaspow and co-workers (1983; Ettehadieh, et al., 1984; 1986a,b; Symalal, 1985), constantly quote the original experimental work of Rietema and Mutsers (1973) that contains the error. A critical evaluation of the use of $G(\epsilon)$ and its derivation from experimental data can be found in Massoudi, et al. (1990).

In their study of slow flow of a dilute suspension of solid particles in a viscous fluid, McTigue, et al. (1986) proposed the following constitutive equation for p_s :

$$p_s = p_f + \bar{\tau} + \omega (\underline{v}_f - \underline{v}_s)^2 + \lambda \text{tr } \underline{D}_f \quad (2.56)$$

where p_f is considered to be the mean fluid pressure and not the fluid pressure in the vicinity of each particle, $\bar{\tau}$ the Brownian pressure which is a function of particle concentration, and ω and λ are assumed to be constants. They also defined the mixture pressure p as

$$p = (1-\phi_s) p_f + \phi_s p_s \quad (2.57)$$

where ϕ_s is the volume fraction of the solid particles. To obtain a value for $\bar{\tau}$, they defined the Brownian diffusivity as

$$\zeta = \frac{1}{\alpha_1} \frac{\partial}{\partial \phi_s} (\phi_s \bar{\tau}) \quad (2.58)$$

where α_1 is the coefficient of drag related to the Stokes flow. McTigue, et al. (1986) use the Nemst-Einstein relation

$$\zeta = \frac{\kappa \theta}{6\pi\mu_f a} \quad (2.59)$$

where κ is the Boltzmann's constant, θ is the absolute temperature, and a is the radius of the particles.

Givler (1987) has given a "quantitative description" for p_s in the slow steady flow of a dilute suspension of particles. By "slow flow" it is implied that the local fluid inertia is important on a length scale compared to the particle diameter. He used the suggestion proposed by Nunziato (1983) and related through p_s and p_f through

$$p_s - p_f = c_0 \gamma_f (\underline{v}_f - \underline{v}_s) \cdot (\underline{v}_f - \underline{v}_s) \quad (2.60)$$

where c_0 is constant and γ_f is the mass density of the fluid. Givler (1987) then sought to mathematically justify the functional form of the above relation and also obtain a value for c_0 . To this end, he used the results of Chester and Breach (1969). By viewing the solid-phase pressure as an average of the disturbance pressure in the vicinity of a single particle and integrating the expression given in Chester and Breach (1969) for $(p-p_f)$ over the particle surface, he obtained

$$p_s - p_f = \frac{1}{S} \int (p - p_f) dS = \frac{9}{32} \text{Re} + O(\text{Re}^2), \quad (2.61)$$

where

$$\text{Re} = \frac{aU\gamma_f}{\mu_f}, \quad (2.62)$$

where U is the uniform streaming velocity. By comparing Equations (2.61) and (2.60), Givler concluded that

$$p_s = p_f + \frac{9}{32} \gamma_f (\underline{v}_f - \underline{v}_s)^2, \quad (2.63)$$

It should be observed that if no relative motion exists between the phases, the contribution due to the local fluid inertia vanishes and $p_s = p_f$. As indicated by Givler (1987), the solid-phase pressure defined by (2.63) is a dynamic effect and its value is greater than the fluid-phase pressure. His results, however, do not agree with Prosperetti & Jones (1984), who proposed the following equation

$$p_s - p_f = - \frac{\gamma_f}{4} (\underline{v}_f - \underline{v}_s)^2, \quad (2.64)$$

which indicates that the solid pressure due to the presence of the solid particles is less than the fluid pressure. Givler (1987) observes that this assumption leads to results that contradict experimental observations.

Green and Homsy (1987) in their study of instabilities in a self fluidized-bed assumed the following constitutive equations for the stresses:

$$\underline{T}_{ij}^f = -p^f \epsilon \delta_{ij}, \quad (2.65)$$

$$\begin{aligned} \underline{T}_{ij}^s = & -p^s (1 - \epsilon) \delta_{ij} \\ & + \beta(\epsilon) D_{kk}^s \delta_{ij} + 2\eta(\epsilon) (D_{ij}^s - D_{kk}^s \delta_{ij}), \end{aligned} \quad (2.66)$$

where p_f and p_s are the fluid and solids pressure, and β and η are the bulk and shear viscosity of the solid phase, respectively.

Recently, Massoudi (1986, 1988) advocated the modeling of the stress in the solid constituent of the mixture by a constitutive expression appropriate to flowing granular particles, and the stress in the fluid constituent of the mixture by a linearly viscous fluid. In particular, he assumed:

$$\underline{T} = -p^f(\epsilon) \underline{1} + \lambda_f(\epsilon) \text{tr } \underline{D}^f \underline{1} + 2\mu_f(\epsilon) \underline{D}^f, \quad (2.67)$$

$$\begin{aligned} \underline{T} = & [\beta_0 + \beta_1 \text{grad } v \cdot \text{grad } v + \lambda_s \text{tr } \underline{D}^s] \underline{1} \\ & + 2\mu_s(v) \underline{D}^s - 2\alpha \text{grad } v \otimes \text{grad } v, \end{aligned} \quad (2.68)$$

where all the material properties are functions of the volume fraction v . Thus, the mixture is assumed to be composed of a Newtonian fluid and a granular solid. A detailed review of the continuum theories of granular materials is given by Massoudi and Boyle (1988).

One of the inherent problems in assuming a fluid-like behavior for the solid phase is defining the solid pressure term. For the fluid, $-p^f \delta_{ij}$ represents the constitutively indeterminate spherical stress due to the constraint of incompressibility, whereas no such analogy exists for the corresponding term for the solid.

It should be pointed out that the basic set of equations is not closed. That is, the conservation of mass and linear momentum for the two phases provides a system of eight equations from which the nine unknowns, $\underline{u}$, $\underline{y}$, ϵ , p^f , and p^s , need to be calculated. This is the reason for the various assumptions on p_s by all the earlier investigators.

Finally, a fundamental shortcoming of assuming a fluid-like behavior for solid particles [cf. Anderson and Jackson (1967), Drew and Segel (1971), and Homsy, et al. (1980)] or a behavior based on ad-hoc assumptions [cf. Gidaspow (1986)] is that, theoretically, in the limit, as the solid volume fraction v approaches zero, the mixture of fluid and solid particles behaves as a fluid, while in the other limit, as the (fluid) void fraction ϵ becomes very small, the mixture does not behave as a granular material. This second limiting case is not described by the majority of the existing constitutive relations for the solid stress tensor in two-phase flows. This issue is discussed in the next section.

3.0 Theory of Interacting Continua and Equations of Motion for the Mixture

3.1 Background

To describe the interactions between bodies composed of different constituents, we will set up the mathematical theory of mixtures. The theory, in its general form, attempts to represent the phenomena of diffusion, dissociation, combination, and chemical reaction [Truesdell (1984)], as well as electromagnetic effects, couple stresses, and singular surfaces [Kelly (1964)].

Atkin and Craine (1976b) have applied the continuum theories of mixtures to a variety of problems such as mixtures of fluids, mixtures of an elastic solid and a viscous fluid, and mixtures of solids, but they did not solve any boundary value problems. An inherent difficulty in applying the mixture theory is that it is formulated in terms of partial stresses. There does not seem to be a physically straightforward way of specifying boundary conditions on the partial stresses in a boundary value problem. This issue was addressed for a wide class of boundary value problems by Shi et al. (1981) and Rajagopal et al. (1986). Using mixture theory and using the boundary conditions generated by Shi, et al., (1981) and Rajagopal, et al. (1986), several boundary value problems have been solved recently involving the diffusion of fluid through isotropic and anisotropic solids undergoing large deformation [cf. Gandhi, et al. (1986), Dai and Rajagopal (1990a,b), Dai, et al. (1990)]. The results agree both qualitatively and quantitatively with experimental results where available [cf. Paul and Ebra-Lima (1970)]. Also, recently Munaff, et al. (1990) have studied the problem of diffusion of fluids through porous rock-like solid and the predictions of the theory, in addition to providing the effects of inertia and the viscosity of the fluid on the diffusion process, a feature absent in a Darcy's law type of approach, agrees well with the original experimental work of Darcy (1856). Thus, the theory of interacting continua has proved to be quite successful in describing the mechanics of fluid-solid mixtures.

Here, we propose to gainfully employ the theory of interacting continua for a mixture of fluids and solid particles.

In the theory of interacting continua, it is assumed a priori that each of the components that make up the mixture are present at each point in space. In other words, each point in space is occupied by a particle belonging to each constituent homogenized over the current configuration. The governing equations are written for each constituent and the interactions between the constituent, e.g., the conversion of one constituent into the other or the supply of mass, momentum, and energy from one constituent to the other, are incorporated into the equations through an appropriate constitutive theory.

3.2 Preliminaries

We shall consider the interaction of two continua, a solid S_1 and a fluid S_2 . Let Ω and Ω_t denote the reference configuration and current configuration of the mixture at time t , respectively. At any instant of time t , each point in Ω is occupied by a particle belonging to both the solid S_1 and the fluid S_2 . Let $\underline{x}$ and $\underline{y}$ denote reference positions of particles in S_1 and S_2 , respectively. The motion of the constituents S_1 and S_2 are defined by

$$\underline{x} = \underline{x}_s(\underline{x}, t) , \quad (3.1)$$

and

$$\underline{y} = \underline{x}_f(\underline{y}, t) , \quad (3.2)$$

respectively. We shall assume that these motions are one-to-one invertible, and sufficiently smooth and that all the derivatives defined later make sense.*

The velocity fields associated with the solid and fluid are defined through†

$$\underline{u}^s = \frac{d\underline{x}_s}{dt}, \quad (3.3)$$

$$\underline{u}^f = \frac{d\underline{x}_f}{dt}, \quad (3.4)$$

and the respective accelerations are

$$\underline{a}^s = \frac{d^2\underline{x}_s}{dt^2}, \quad (3.5)$$

and

$$\underline{a}^f = \frac{d^2\underline{x}_f}{dt^2}. \quad (3.6)$$

*We shall assume that $\Omega, \Omega_t \subset E$, where E is a three-dimensional Euclidean space. For a function defined on $\Omega \times \mathbb{R}$ we denote by grad and d/dt , partial derivatives with respect to the first and second place, respectively. For a function defined on $\Omega \times \mathbb{R}$, we shall denote by grad and $\partial/\partial t$, partial derivatives with respect to the first and second place. The divergence operator associated with grad is denoted by div. The Laplacian associated with grad is denoted by ∇^2 .

†We shall use suffixes or superscripts s and f to denote quantities associated with the solids and fluid, respectively.

The deformation gradient associated with the motion $\underline{x}_S$ is defined through

$$\underline{F} = \frac{\partial \underline{x}_S}{\partial \underline{x}} = \text{grad } \underline{x}_S . \quad (3.7)$$

The velocity gradients associated with the solid and fluid motion are

$$\underline{L}_S = \text{grad } \underline{u}^S , \quad (3.8)$$

$$\underline{L}_f = \text{grad } \underline{u}^f , \quad (3.9)$$

and the symmetric and skew-symmetric parts of the velocity gradients are defined through

$$\underline{D}_S = \underline{L}_S + \underline{L}_S^T , \quad (3.10)$$

$$\underline{D}_f = \underline{L}_f + \underline{L}_f^T , \quad (3.11)$$

$$\underline{W}_S = \underline{L}_S - \underline{L}_S^T , \quad (3.12)$$

and

$$\underline{W}_f = \underline{L}_f - \underline{L}_f^T . \quad (3.13)$$

3.3 Conservation of Mass

Let ρ_{10} and ρ_{20} represent the densities of the solid and fluid in their respective reference states. Let ρ_1 and ρ_2 denote the density of the solid and fluid in the mixed state, respectively. The total density of the mixture ρ is then defined through

$$\rho = \rho_1 + \rho_2 . \quad (3.14)$$

In the absence of interconversion of mass between the solid and fluid, the conservation of mass for the solid is given in the Lagrangean form by

$$\rho_1 |\det \underline{E}| = \rho_{10} . \quad (3.15)$$

The conservation of mass for the fluid is given in the Eulerian form by

$$\frac{\partial \rho_2}{\partial t} + \operatorname{div}(\rho_2 \underline{u}_f) = 0 . \quad (3.16)$$

3.4 Balance of Linear Momentum

Let $\underline{I}^s$ and $\underline{I}^f$ denote the partial Cauchy stress associated with the solid and fluid, respectively. The total Cauchy stress $\underline{I}$ is defined through

$$\underline{I} = \underline{I}^s + \underline{I}^f . \quad (3.17)$$

The interaction between the two constituents gives rise to forces which are denoted by the Interaction force $\underline{I}$. $\underline{I}$ is essentially the momentum supplied by the second constituent to the first. Let $\underline{b}^1$ and $\underline{b}^2$ denote the external body force fields on the solid and fluid, respectively. The balance of linear momentum of the solid and fluid are given by

$$\operatorname{div} \underline{I}_s - \underline{I} + \underline{b}^1 = \rho_1 \underline{a}_s , \quad (3.18)$$

$$\operatorname{div} \underline{I}_f + \underline{I} + \underline{b}^2 = \rho_2 \underline{a}_f , \quad (3.19)$$

respectively.

3.5 Balance of Angular Momentum

The balance of the moment of momentum implies that

$$\underline{I}_s + \underline{I}_f = (\underline{I}_s)^T + (\underline{I}_f)^T . \quad (3.20)$$

Thus, while the total stress is symmetric, the individual partial stresses need not be symmetric.

3.6 Surface Conditions

Let $\underline{n}$ denote the outward unit normal at a point on Ω_t . Let $\underline{t}_s$ and $\underline{t}_f$ denote the surface traction supported by the solid and fluid, respectively. The partial stress tensors $\underline{I}_s$ and $\underline{I}_f$ are related to $\underline{t}_s$ and $\underline{t}_f$ through

$$\underline{t}_s = \underline{I}_s \underline{n} , \quad \text{and} \quad \underline{t}_f = \underline{I}_f \underline{n} . \quad (3.21)$$

3.7 Volume Additivity Constraint

We shall assume that the volume of the mixture is the sum of the volumes of the individual constituents in their reference states. This assumption places a restriction on the motion of the mixture [cf. Mills (1966)]:

$$\frac{\rho_1}{\rho_{10}} + \frac{\rho_2}{\rho_{20}} = 1 . \quad (3.22)$$

(Balance of energy for the mixture and the entropy inequality are not discussed here as we are interested in purely mechanical interactions.)

3.8 Constitutive Relations for the Stresses

The equations presented in the previous sections express properties common to all bodies and motions. The differences among the materials that make up these bodies are reflected in the theory by constitutive relations. Specifically, Equations (3.14) through (3.22) describe the motion of two interacting continua where the effects of temperature and interconversion of mass have been neglected.

Based on our knowledge of modeling in the theory of granular solids and linearly viscous fluid [cf. Rajagopal, et al. (1990)], it would be natural to assume all the constitutive functions A^a , q^a , $\underline{I}^a$, $\underline{I}$, etc., depend on

$$\rho^1, \rho^2, \nabla\rho^1, \nabla\rho^2, \nabla\nabla\rho^1, \nabla\nabla\rho^2, \theta, \nabla\theta, \underline{u}^f - \underline{u}^s, \underline{D}^s, \underline{D}^f, \quad (3.23)$$

where A is the specific Helmholtz free energy, $\underline{q}$ is the heat flux vector, $\underline{I}$ is the stress, $\underline{I}$ is the interaction, ρ is the density, θ is the temperature, $\underline{u}$ is the velocity, and $\underline{D}$ is the stretching tensor, and the superscripts f (or 2) and s (or 1) denote the properties associated with the fluid and solid, respectively. In addition to the quantities in Equation (3.21), the interaction term $\underline{I}$ could depend on a frame-indifferent quantity derivable from the accelerations $\underline{a}^s$ and $\underline{a}^f$ of the solid and the fluid acceleration, respectively, to account for the virtual mass effect. Then, using methods which are by now standard in continuum mechanics [cf. Shi, Rajagopal and Wineman (1981), Atkin and Craine (1976)], we can obtain restrictions and forms for such constitutive expressions. Here, we shall not discuss such an approach. Another approach to modeling the mixture is to postulate the constitutive expressions by simply generalizing the structure of the constitutive relations from single constituent theory. This is what we shall do here. In this case,

$$\underline{I}^s = \underline{I}^s (\rho^1, \rho^2, \nabla\rho^1, \nabla\rho^2, \nabla\nabla\rho^1, \nabla\nabla\rho^2, \underline{u}^f - \underline{u}^s, \underline{D}^s, \underline{D}^f), \quad (3.24)$$

$$\underline{I}^f = \underline{I}^f (\rho^1, \rho^2, \nabla\rho^1, \nabla\rho^2, \nabla\nabla\rho^1, \nabla\nabla\rho^2, \underline{u}^f - \underline{u}^s, \underline{D}^s, \underline{D}^f), \quad (3.25)$$

In the case of a single constituent granular solid, the Cauchy stress $\underline{I}^s$ would depend on the arguments that have the superscript s associated with them. Such a model would be a generalization of the model derived by Goodman and Cowin (1972) to describe the mechanics of granular solids. Due to the presence of the term $\nabla\nabla\rho^1$ such a theory would include the model proposed by Kortweg to describe the effects of capillarity [cf. Truesdell and Noll (1965)]. It is also worth observing that such models exhibit the phenomena of normal stress differences [cf. Rajagopal and Massoudi (1989)].

In general, the constitutive expressions for $\underline{I}^s$ and $\underline{I}^f$ in Equations (3.24) and (3.25) would depend on the kinematical quantities associated with both constituents. However, we shall assume that $\underline{I}^s$ and $\underline{I}^f$ depend only on the kinematical quantities associated with the solid and fluid, respec-

tively. This assumption is sometimes called the "principle of phase separation" [cf. Drew (1976)]. This is not a principle but just an assumption [cf. Adkins (1963)]. For the sake of simplicity, we shall assume that $\mathbb{I}^S$ and $\mathbb{I}^f$ do not depend on $\nabla\nabla\rho^1$ and $\nabla\nabla\rho^2$, respectively. We shall assume that

$$\begin{aligned} \mathbb{I}^S = & a_0 \underline{1} + a_1 \underline{D} + a_2 \underline{N} + a_3 \underline{D}^2 + a_4 \underline{N}^2 + a_5 (\underline{D}\underline{N} + \underline{N}\underline{D}) \\ & + a_6 (\underline{D}^2 \underline{N} + \underline{N} \underline{D}^2) + a_7 (\underline{D}\underline{N}^2 + \underline{N}^2 \underline{D}) + a_8 (\underline{D}^2 \underline{N}^2 + \underline{N}^2 \underline{D}^2), \end{aligned} \quad (3.26)$$

where

$$\underline{N} = \nabla\rho^1 \otimes \nabla\rho^1, \quad (3.27)$$

and the coefficients a_i , $i = 0, 1, \dots, 8$ depend on ρ^1 , $\text{tr } \underline{D}$, $\text{tr } \underline{D}^2$, $\text{tr } \underline{D}^3$, $\text{tr } \underline{N}$, $\text{tr } \underline{N}^2$, $\text{tr } \underline{N}^3$, $\text{tr}(\underline{D}\underline{N})$, $\text{tr}(\underline{D}\underline{N}^2)$, $\text{tr}(\underline{N}\underline{D}^2)$, $\text{tr}(\underline{N}^2 \underline{D}^2)$.

If we require that $\mathbb{I}^S$ depends linearly on $\underline{D}^S$ and $\underline{N}$, this model simplifies considerably:

$$\begin{aligned} \mathbb{I}^S = & \{ \beta_0(\rho^1) + \beta_1(\rho^1) \text{grad } \rho^1 \cdot \text{grad } \rho^1 + \beta_2(\rho^1) \text{tr} \underline{D} \} \underline{1} \\ & + \beta_3(\rho^1) \nabla\rho^1 \otimes \nabla\rho^1 + \beta_4(\rho^1) \underline{D}. \end{aligned} \quad (3.28)$$

The spherical part of stress in Equation (3.28) can be interpreted as the solid pressure p^S . However, here $p^S = p^S(v)$, and the term within the parenthesis is the constitutive expression for p^S . The material moduli β_1 and β_3 are material parameters that reflect the distribution of the granular particles, and β_0 plays the role akin to pressure in a compressible fluid and is given by an equation of state. The material modulus β_2 is again a viscosity akin to the second coefficient of viscosity in a compressible fluid, and β_4 denotes the viscosity (i.e., the resistance of the material to flow) of the granular solids. Recently, Rajagopal and Massoudi (1990) have outlined an experimental/theoretical program to determine these material moduli.

In the majority of fluid-solid mixtures the fluid is either a gas or water. Therefore, it seems appropriate to assume that the fluid behaves as a linearly viscous fluid (Newtonian), whose constitutive equation is

$$\underline{\underline{I}}^f = -p^f \underline{\underline{1}} + \lambda_f (\text{tr } \underline{\underline{D}}^f) \underline{\underline{1}} + 2\mu_f \underline{\underline{D}}^f. \quad (3.29)$$

In the above equation, p^f is the fluid pressure, λ_f and μ_f are the viscosities, and $\underline{\underline{D}}^f$ is the stretching tensor for the fluid. If the fluid is incompressible, then p^f is one of the unknown quantities in the problem that would have to be calculated. If the fluid is compressible, an equation of state is needed. In general, p^f , λ_f , and μ_f are functions of ρ_f .

The above constitutive relations, Equations (3.28) and (3.29), express the behavior of the granular materials and the fluid phase, respectively, in their reference states. However, since we are interested in the motion of behavior of the mixture in the current configuration, these equations somehow would have to be modified.

Further, let

$$\nu = \phi = \frac{\rho_1}{\rho_{10}}, \quad \bar{\phi} = \frac{\rho_2}{\rho_{20}} = \frac{\rho_2}{\rho_f}, \quad (3.30)$$

where ρ_1 denotes the density of solid in the current state in the mixture, and ρ_{10} the density of the granular solid. Also, ρ_2 and ρ_{20} denote the density of the fluid in the current and reference state, respectively, by the volume additivity constraint.

Departing from previous approaches, we shall assume that the stresses $\underline{\underline{I}}^s$ and $\underline{\underline{I}}^f$ in the pure state are weighted by ϕ and $(1-\phi)$, respectively. Thus, we shall assume that

$$\underline{\underline{I}}_1 = \underline{\underline{I}}^s \quad (3.31)$$

and

$$\underline{I}_2 = \bar{\phi} \underline{I}^f = (1-\phi) \underline{I}^f, \quad (3.32)$$

and thus the total Cauchy stress in the mixture $\underline{I}$ is given by

$$\underline{I} = \underline{I}^S + (1-\phi) \underline{I}^f. \quad (3.33)$$

When $\phi \rightarrow 0$, then $\underline{I} \rightarrow \underline{I}^f$, while when $\phi \rightarrow 1$, then $\underline{I} \rightarrow \underline{I}^S$, as we would expect.

In previous approaches, the weighting of the stresses of the components were done according to the ratio of their current density in the mixture to their pure density. Thus, for instance if ρ_s and ρ_f represented the pure density, and ρ_1 and ρ_2 the current density (in the mixture), then $\underline{I}^S$ was weighted by ρ_1/ρ_s and $\underline{I}^f$ by ρ_2/ρ_f . However, the weighting could also be done with regard to the mass fractions of the constituents in the mixture rather than their mass fractions with regard to their respective pure state. Thus, we could weight the first constituent by ρ_1/ρ , and the second constituent by ρ_2/ρ . In this sense, the former is a weighting in a Lagrangean manner with weighting done with regard to the reference mass fractions while the latter could be viewed as an Eulerian approach in which the weighting is done with respect to the ratio of the current mass fractions. Notice that in our modeling we have neglected the dependence of stresses on $\nabla \nabla \rho^1$ and $\nabla \nabla \rho^2$. Introducing higher gradients in the theory always introduces the attendant difficulty of specifying additional boundary conditions. Later, we shall see that even dependence of ∇v introduces difficulties with regard to boundary condition. In the above model, Equation (3.28), we shall assume that β_0 , β_2 and $\beta_4 \rightarrow 0$ as $v \rightarrow 0$, and thus $\underline{I}^S \rightarrow 0$ as $v \rightarrow 0$. This simply implies that when there are no particles, i.e., $v = 0$, the stress in the granular materials, $\underline{I}^S$, should vanish. Also, notice that $\underline{I}_2 \rightarrow 0$ as $v \rightarrow 1$.

3.9 Governing Equations

Since we are treating the mixture of fluid and solid particles as a continuum, the balance laws valid for a single continuum would also apply here. Furthermore, inherent in the theory of interacting continua are the following ideas, attributed to Truesdell (1984):

- "1. All properties of the mixture must be mathematical consequences of properties of the constituents.
2. So as to describe the motion of a constituent, we may in imagination isolate it from the rest of the mixture, provided we allow properly for the actions of the other constituents upon it.
3. The motion of the mixture is governed by the same equations as is a single body."

With these, we proceed to regard the governing equations. The conservation of mass for each constituent is given by

$$\frac{\partial \rho_1}{\partial t} + \text{div} (\rho_1 \underline{u}) = 0 , \quad (3.34)$$

$$\frac{\partial \rho_2}{\partial t} + \text{div} (\rho_2 \underline{v}) = 0 , \quad (3.35)$$

where $\partial/\partial t$ denotes the partial derivative with respect to time, and ρ_1 and ρ_2 represent the density of the solid and fluid in the mixture, respectively. If ρ_s and ρ_f represent the densities of the solid and fluid in the reference state, and furthermore, if the solid and fluid are assumed to be incompressible in the reference state, i.e., $\rho_s = \text{constant}$ and $\rho_f = \text{constant}$, then Equations (3.34) and (3.35) can be rewritten as

$$\frac{\partial \phi}{\partial t} + \text{div} [\phi \underline{u}] = 0 , \quad (3.36)$$

$$\frac{\partial}{\partial t} (1-\phi) + \text{div} [(1-\phi) \underline{v}] = 0 . \quad (3.37)$$

The conservation of linear momentum takes the form

$$\rho_1 \frac{d\underline{u}}{dt} = \text{div} \underline{I}_1 - \underline{I} + \rho_1 \underline{b} . \quad (3.38)$$

$$\rho_2 \frac{d\underline{v}}{dt} = \text{div} \underline{I}_2 + \underline{I} + \rho_2 \underline{b} . \quad (3.39)$$

where

$$\underline{I}_1 = \underline{I}_s \quad \text{and} \quad \underline{I}_2 = (1-\phi) \underline{I}_f . \quad (3.40)$$

To obtain the conservation of mass for the mixture, we add Equations (3.34) and (3.35). Therefore,

$$\frac{\partial}{\partial t} (\rho_1 + \rho_2) + \text{div} (\rho_1 \underline{u} + \rho_2 \underline{v}) = 0 . \quad (3.41)$$

The mass-weighted velocity of the mixture is defined [cf. Bowen (1976)]

$$\rho \underline{w} = \rho_1 \underline{u} + \rho_2 \underline{v} . \quad (3.42)$$

Thus, substituting Equations (3.42) and (3.14) in Equation (3.41) we obtain the conservation of mass for the mixture:

$$\frac{\partial \rho}{\partial t} + \text{div} (\rho \underline{w}) = 0 . \quad (3.43)$$

To obtain the conservation of linear momentum equation for the mixture, we add Equations (3.38) and (3.39). Therefore

$$\rho_1 \frac{du}{dt} + \rho_2 \frac{dv}{dt} = \text{div} (\underline{I}_1 + \underline{I}_2) + (\rho_1 + \rho_2) \underline{b}$$

or

(3.44)

$$\rho_1 \frac{du}{dt} + \rho_2 \frac{dv}{dt} = \text{div} \underline{I} + \rho \underline{b} ,$$

where $\underline{I}$ is given in Equation (3.33). Notice that we do not "force" the equation of linear momentum for the mixture, Equation (3.44), to look like the equation

$$\rho \frac{dw}{dt} = \text{div} \underline{I}^* + \rho \underline{b} , \quad (3.45)$$

where now the stress of the mixture, $\underline{I}^*$, is no longer defined as the sum of the partial stresses [cf. Equation (3.33)]. Both approaches have been used in mixture theory. Green and Naghdi (1969) provide an interesting explanation as to why the form of the equation of linear momentum, given by Equation (3.44), may be more appropriate than the form given by Equation (3.45).

Substituting Equations (3.28), (3.29), and (3.40) into Equation (3.44), we obtain

$$\begin{aligned} \rho_s v \frac{du}{dt} + \rho_f (1-v) \frac{dv}{dt} = & \text{div} [(\beta_0 + \beta_1 \text{grad } v \cdot \text{grad } v + \beta_2 \text{tr} \underline{D}^s) \underline{1} \\ & + \beta_3 \underline{D} + \beta_4 \text{grad } v \otimes \text{grad } v] \end{aligned} \quad (3.46)$$

$$+ \text{div} [-(1-v)p^f \underline{1} + \lambda_f (1-v) (\text{tr} \underline{D}^f) \underline{1} + 2\mu_f (1-v) \underline{D}^f]$$

$$+ \rho_s v \underline{b} + \rho_f (1-v) \underline{b}$$

Taking the limit of this equation as $\nu \rightarrow 0$ (no particles), we should obtain the Navier-Stokes equations. This is only possible if

$$\begin{aligned} \beta_0 &\rightarrow 0 \\ \beta_2 &\rightarrow 0 \\ \beta_3 &\rightarrow 0 \end{aligned} \quad \text{as } \nu \rightarrow 0 \quad (3.47)$$

which was mentioned earlier. Therefore, Equation (3.46), as $\nu \rightarrow 0$, becomes

$$\rho_f \frac{dv}{dt} = \text{div} [(-p^f + \lambda_f (\text{tr} \underline{D})) \underline{1} + 2\mu_f \underline{D}^f] + \rho_f \underline{b} . \quad (3.48)$$

Taking the limit of Equation (3.46) as $\nu \rightarrow 1$ (i.e., dense flow of granular materials), we obtain the equation of motion for flowing granular materials where the effects or presence of the interstitial fluid is neglected. Therefore, Equation (3.46), as $\nu \rightarrow 1$, becomes

$$\begin{aligned} \rho_s \nu \frac{du}{dt} = \text{div} [(\beta_0 + \beta_1 \text{grad } \nu \cdot \text{grad } \nu + \beta_2 \text{tr} \underline{D}^s) \underline{1} \\ + \beta_3 \underline{D} + \beta_4 \text{grad } \nu \otimes \text{grad } \nu] + \rho_s \nu \underline{b} \end{aligned} \quad (3.49)$$

To obtain explicit dependence of β_i 's on ν , we use a Taylor series expansion. Thus,

$$\beta_i(\nu) = \beta_{i0} + \beta_i(0) \nu + O(|\nu^2|) , \quad i = 0, 1, 2, 3, 4 \quad (3.50)$$

where $O(|\nu^2|)$ indicates terms of higher order than ν . Equation (3.47) implies

$$\beta_{i0} = 0 , \quad i = 0, 2, 3, \quad (3.51)$$

Therefore, as a first approximation, the material properties β_0 through β_4 must have the following forms:

$$\beta_i = k_i v, \quad i = 0, 2, 3, \quad (3.52)$$

$$\beta_j = k_{0j} + k_j v \quad j = 1, 4$$

Through a simple analysis, Rajagopal and Massoudi (1989) have shown that if the constitutive Equation (3.28) is to apply at equilibrium (static) cases, then

$$k_0 < 0. \quad (3.53)$$

Therefore, we shall also assume

$$\beta_0 = -\kappa v. \quad (3.54)$$

With these restrictions, the stress tensor of the mixture, Equation (3.33), becomes:

$$\begin{aligned} \underline{T} = & [-\kappa v - (1-v) p + (k_{01} + k_1 v) \text{grad } v \cdot \text{grad } v + (k_2 v) \text{tr} \underline{D}^S \\ & + \lambda_f (1-v) \text{tr} \underline{D}^f] \underline{1} + k_3 v \underline{D}^S + (k_{04} + k_4 v) \text{grad } v \otimes \text{grad } v \\ & + 2\mu_f (1-v) \underline{D}^f. \end{aligned} \quad (3.55)$$

In the next section, we will discuss the implications of modeling a mixture of fluid-solid particles as being composed of granular materials and a linearly viscous fluid.

4.0 Comments and Conclusions

Historically, the problem of the theoretical determination of the viscosity of an infinitely dilute suspension consisting of an incompressible Newtonian fluid and rigid spherical particles was studied by Einstein (1956). He derived the classical formula for the effective viscosity of the suspension

$$\mu^* = \mu_f (1 + 2.5 \phi) , \quad (4.1)$$

where μ_f is the viscosity coefficient of the fluid and ϕ is the volume fraction of spheres in the suspension. Einstein assumed that ϕ is very small compared with unity. This indicates that the particles are far apart from each other and are not interacting with each other. Taylor (1932) showed that if the spheres are small drops of another fluid, then the viscosity of the suspension is given by

$$\mu^* = \mu_f \left[1 + 2.5 \phi \left(\frac{\mu' + \frac{2}{5} \mu_f}{\mu' + \mu_f} \right) \right] , \quad (4.2)$$

where μ' is the viscosity of the liquid drops and μ_f is the viscosity of the host fluid.

Later, Batchelor and Green (1972) derived a formula for the effective viscosity for the term of order ϕ^2 . They showed that

$$\mu^* = \mu_f (1 + 2.5 \phi + 7.6 \phi^2) , \quad (4.3)$$

where again $\phi \ll 1$. For a recent review of this subject, the reader is referred to Hsu (1987).

Sampaio and Williams (1977) used a modified version of mixture theory to obtain an expression for the effective viscosity of a mixture of two

liquids. They showed that if the velocities of the two liquids are equal, then the effective viscosity of the mixture is given by

$$\mu = \mu_A + \mu_B + \mu_{AB} , \quad (4.4)$$

where μ_A and μ_B are viscosities of the two fluids and μ_{AB} is defined as "mutual viscosity," and arises due to the interactions of the molecules of liquid A and liquid B and all the viscosities are assumed to be functions of ϕ .

If this is the case, then it is not clear how μ_{AB} could exist [cf. Equation (4.4)] if the two liquids have the same velocity. Sampaio and Williams (1977) compared Equation (4.4) with the expression given by Dolezalek and Schulze (1913), where

$$\mu = \phi^2 \eta_A + 2\phi(1-\phi)\eta_{AB} + (1-\phi)^2 \eta_B , \quad (4.5)$$

where η_A and η_B are the pure (unmixed) viscosities of the two liquids. Sampaio and Williams (1977) concluded that

$$\mu_A = \phi^2 \eta_A , \quad \mu_B = (1-\phi)^2 \eta_B , \quad \mu_{AB} = 2\phi(1-\phi)\eta_{AB} . \quad (4.6)$$

Nunziato (1983), using mixture theory, defined the viscosity of a mixture of a fluid and particle as

$$\mu = (1-\phi) \mu_f + \phi \mu_s , \quad (4.7)$$

where μ_f and μ_s are the viscosities of the fluid and solid constituents, respectively. Again, for the case of no slip between the two phase (i.e., equal velocities), Nunziato (1983) suggested that the solid viscosity, in the low concentration limit [cf. Equation (4.1)], can be estimated as

$$\mu_s = 3.5 \mu_f . \quad (4.8)$$

If the mixture of fluid-solids is assumed to be composed of a saturated granular media infused with a linearly viscous fluid, then the stress tensor for the mixture is given in Equation (3.55). It can be seen that it is not an easy matter to define a mixture viscosity by simply looking at Equation (3.55). That is, if somehow we can rewrite this equation as

$$\underline{T} = -p_m \underline{1} + 2\mu_m \underline{D}_m + \alpha \text{grad } v \otimes \text{grad } v . \quad (4.9)$$

where p_m can be thought of as a mixture pressure and $\underline{D}_m$ as the stretching rate for the mixture, then we can consider the meaning of μ_m (the mixture viscosity) and how it can be measured. This, however, is not an easy task. Instead, if we consider the case of equal velocities (i.e., particles are simply being carried by the host fluid), where

$$\underline{u} = \underline{v} , \quad (4.10)$$

the Equation (3.55) is rewritten as

$$\begin{aligned} \underline{T} = & [-k_v - (1-v)p + (k_{01} + k_1 v) \text{grad } v \cdot \text{grad } v \\ & + (k_2 v + \lambda_f (1-v)) \text{tr} \underline{D}] \underline{1} + [k_3 v + 2\mu_f (1-v)] \underline{D} \\ & + (k_{04} + k_4 v) \text{grad } v \otimes \text{grad } v . \end{aligned} \quad (4.11)$$

This indicates that the flow characteristics of a suspension can be described, in this special case, by a constitutive relation similar to that of Korteweg-type fluid [cf. Truesdell and Noll (1965)]. (The previous approaches have all indicated that the suspension behaves like a Newtonian fluid where the effective viscosity is a function of the volume concentration.) Therefore, if we perform a simple shear flow experiment, the viscosity of the mixture

$$\mu_m = k_3 v + \mu_f (1-v) , \quad (4.12)$$

is measured. If at the same time, the volume fraction of the solids and the viscosity of fluid are independently measured, we can obtain the viscosity

of solid particles k_s . Equation (4.12) simply indicates a relationship between the viscosity of the mixture and the viscosities of the two constituents provided:

1. The fluid is modeled as a linearly viscous fluid, Equation (3.29),
2. The solid particles are modeled as a granular media represented by a constitutive relation, Equation (3.28),
3. The total stress of the mixture is weighted according to Equation (3.33), and
4. The material moduli of the granular media are approximated as linear functions, Equation (3.52).

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6.0

Nomenclature

a	Particle radius
$\underline{a}$	Acceleration vector
A	Specific Helmholtz free energy
$\underline{b}$	Specific body force
$\underline{D}$	Rate of deformation
$\underline{E}$	Deformation gradient
$\underline{I}$	Interaction force
k_i	Material coefficients
$\underline{L}$	Velocity gradient
$\underline{n}$	Unit normal vector
p	Pressure
$\underline{q}$	Heat flux vector
R	Gas constant
T	Temperature
$\underline{T}$	Stress tensor
$\underline{t}$	Traction vector
$\underline{u}, \underline{v}$	Velocity vectors
$\underline{w}$	Mixture velocity

Greek Characters

v	Volume fraction of solids
ϵ	Void fraction of fluid
ϕ	Volume fraction
ρ	Density
μ	Shear viscosity
λ	Bulk viscosity
β_i	Material moduli of the granular media
η	Viscosity
θ	Temperature
π	Pressure
ψ	Free energy
λ^*	Brownian diffusivity

τ	Shear stress
χ	Mapping function
$\underline{\Delta}$	Micro-gyration vector
δ_{ij}	Kronecker delta

Superscripts (Subscripts) and Other Symbols

f	Referring to the fluid phase
s	Referring to the solid phase
'	Prime, differentiation
~	A tensorial quantity
$\otimes$	Outer (dyadic) product
∇	Gradient symbol
tr	Trace of a tensor
T	Transpose
1 $\circ$ or 2 $\circ$	Referring to the reference states
div	Divergence operator
Σ	Summation sum
1 or 2	Referring to the solid or fluid phases

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