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Chapter 18

OVERVIEW OF EULERIAN MODELING METHODS FOR FLUIDIZED SUSPENSIONS

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Abstract

This review is concerned with the mathematical modeling of dense fluidized suspensions, and focuses on the so-called Eulerian or multifluid approach. It introduces newcomers to some of the techniques adopted to model fluidized beds and to the challenges and long-standing problems that these techniques present. We first describe the main approaches used to model fluidized suspensions. Focusing on the multifluid approach, we then overview the principal averaging techniques that consent to turn granular systems into continua; in particular, we discuss statistical, volume and time averages. After, we use volume averages to derive the Eulerian equations of motion for fluidized suspensions of a finite number of monodisperse particle classes. We introduce the closure problem, and overview some closure equations used to express the granular stress and the interaction forces between the phases, giving emphasis to the fluid-particle interaction force and in particular to the buoyancy and drag contributions. We finally extend the modeling to fluidized suspensions of particles continuously distributed over their size, introducing the population balance equation and some numerical techniques that permit to solve it.

PACS 05.45-a, 52.35.Mw, 96.50.Fm.

Keywords: Multiphase flows, Fluidization, Mathematical modeling, CFD.

1. Introduction

Since the 1920s, when it first appeared in commercial use (Geldart, 1973), fluidization has attracted more and more the attention of the industrial world, which did not fail to

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recognize and appreciate the potential offered by this promising technology. This interest, boosted during the Second World War by wartime pressure and a desperate need for aviation gasoline, explains why fluidization achieved soon widespread use in several industrial applications, particularly in the fields of catalytic cracking and coal gasification. Since then, it has lived up to the expectations, turning into a well-established technology employed nowadays in various other areas such as coal combustion, sulphide ores roasting, food processing, biomass gasification, waste disposal and even oyster nursery (Ver & Wang, 1995).

Albeit fluidized beds are used extensively, designing them is still a major challenge to process engineers. The performance of these systems depends strongly on the bed fluid dynamics. This can change significantly and is affected by several variables: the properties of the powder (mainly its size distribution and the particles mean size and density), those of the fluid (mainly its density and viscosity), various process variables such as fluid temperature, pressure and flow rate, and geometrical properties of the vessel containing the bed. Thus, every fluidized system can be described by a fluidization quality map that comprises several fluid dynamic regimes in which the system can operate. By choosing appropriately the values of the variables previously mentioned, one can have the fluidized bed work in any region of the map, attaining the fluid dynamics that best suits the application at hand.

This is the challenge that process engineers have to address. On the one hand, predicting the behavior of industrial fluidized beds is, to quote Geldart (1973), 'tougher than assessing the arrival time of a space probe traveling to Saturn.' On the other hand, being their construction and commissioning quite expensive, design uncertainties must be avoided or at least minimized.

For many years, and partly still today, process engineers have used pilot plants and scaling-up relations to design fluidized beds. But this practice is risky. Whereas laboratory bench-scale experiments might well be used to test the feasibility of basic processes, they do not help much to design industrial plants, inasmuch as the fluidization quality observed in the small-scale prototypes does not indicate what this will be in the respective commercial units (Geldart, 1973). This is all the more true if the bed contains internals, such as heat exchanger tubes, bound to modify the bed fluid dynamics.

When fluidized beds were first employed in the 1920s - 1940s, engineers did not appreciate this problem, most probably because at the time the required plants performance was either not critical (as in FCC plants) or easily achievable (as in roasting and drying). But when later on the problem revealed itself in other, and more demanding, applications, with plants falling far short of the expected conversions formerly achieved in pilot units, it became clear that this matter had to be addressed more thoroughly. So, researchers endeavored to find more reliable methods to predict the dynamics of fluidized suspensions.

In the late 1950s and early 1960s, scientists made major breakthroughs. Using the laws of conservation of mass, linear momentum and energy to analyze nearly any physical and chemical problem, they attained significant theoretical headway. This breakaway approach, fostered by the release of the first edition of the influential textbook *Transport Phenomena* (Bird et al., 1960), bolstered the hope to explain theoretically the behavior of fluidized suspensions, and more generally multiphase fluids, prompting the first trials to develop fluid dynamic models based on conservation equations.

Anderson & Jackson (1967) were among the first to model fluidized beds rigorously.

Starting from the Navier-Stokes equations of continuum mechanics and the Newtonian equations for rigid-body motion, they derived averaged Eulerian equations of conservation for both fluid and particle phases by applying a formal mathematical process of volume averaging. Subsequently, other researchers did the same, refer for instance to Whitaker (1969), Drew (1971) and Drew & Segel (1971). Initially, they employed these models to better understand the complex behavior of multiphase systems, without considering them as a viable alternative to design real systems. But afterwards, when faster computer processors and advanced numerical methods to integrate coupled partial differential equations became available, they realized that a mathematical theory of multiphase flows could be a useful design tool for applications of practical interest.

With the further development of new and more rigorous formulations of multiphase equations of motion (Buyevich, 1971; Hinch, 1977; Nigmatulin, 1979; Drew, 1983; Jenkins & Savage, 1983), the late 1970s and early 1980s witnessed the first endeavors to simulate numerically granular flowing systems (Pritchett et al., 1978; Gidaspow & Ettehadieh, 1983; Gidaspow et al., 1986). The promising results of these few pioneering studies generated an increasing interest in computational fluid dynamics (CFD) and multiphase flows, which rapidly started to turn into research areas in their own right.

Nowadays, CFD has become an almost indispensable tool to solve problems of academic and industrial interest. In the field of fluidization, CFD has assisted to understand fluid-solid interactions and has enabled to predict numerous macroscopic phenomena encountered in particulate systems. Similarly, multiphase flows and fluidization dynamics have become topics of interest not only for the scientific community but also for the industrial world. Over the years, many researchers have developed mathematical models to predict the dynamics of dense fluidized suspensions, proposing several approaches and mathematical schemes; in the next section, we briefly overview some of these strategies (namely, statistical, volume and time averaging), highlighting their advantages and limitations.

2. An Overview of Fluidized Bed Modeling

Fluidized beds can be modeled at various levels of detail. At the most fundamental, the motion of the system is determined by the Newtonian equations for translation and rotation of each particle and the Navier-Stokes transport equations to be satisfied at every point occupied by the interstitial fluid. In this approach, referred to as Eulerian-Lagrangian, the fluid flow field is modeled at a length scale far smaller than the particle size. The Lattice-Boltzmann model (LBM) is a technique that many adopt to compute the fluid-solid interactions at this microscopic length scale (Ladd & Verberg, 2001; Succi, 2001). van der Hoef et al. (2005) used the LBM to compute the mean drag force exerted by a fluid flowing among spheres, finding that the constitutive equations in the literature underestimate the force in the laminar flow regime.

Despite its appeal, the Eulerian-Lagrangian strategy is not the most effective. First, it is very demanding computationally: simulations of this kind have been performed only for diluted systems containing a small number of particles – 1204 in the work of Pan et al. (2002); extending these calculations to dense mixtures, like those found in industrial fluidized beds, is presently inconceivable. Moreover, even if this were feasible, the information provided

by the solution would be too detailed, and one would have to filter or average the results to make them useful.

These observations suggest that it might be convenient to formulate equations of change governing the evolution of these averages directly. In this approach, we renounce to capture all the details described above, satisfying ourselves with a much reduced description of the flow. Although there is no guarantee that these simplified equations can be really found – in closed form, that is – several studies have been conducted in this pursuit (Anderson & Jackson, 1967; Whitaker, 1969; Drew, 1971; Drew & Segel, 1971; Drew, 1983; Drew & Lahey, 1993; Gidaspow, 1994; Zhang & Prosperetti, 1994; Enwald et al., 1996; Jackson, 1997, 1998, 2000). Owing to the complexity of the problem, no one expects, at least for the moment, to derive the exact averaged multiphase equations of motion. The intent is merely to formulate models that can describe satisfactorily phenomena of interest for industrial applications.

Various mathematical techniques yield such equations, and several claims have been advanced as to the superiority of each form of averaging versus the others. However, the resulting transport equations are very similar and present many common features. Two are the most significant. First, they are all written in terms of mean variables defined over the whole physical domain; thus, they resemble those that one would write for imaginary fluids that interpenetrate each other and occupy simultaneously the same physical volume. The model, known as Eulerian-Eulerian or also multifluid, takes therefore the form of coupled differential equations subjected to initial and/or boundary conditions assigned only on the mixture boundaries – that is, no longer on the particle surfaces as in the Eulerian-Lagrangian. Second, the averaging process generates a number of indeterminate terms not directly related to the averaged variables but still associated with details of the motion at the microscopic length scale. These are key terms, represented by the fluid and solid stress tensors and by the interaction forces exchanged by the phases. A closure problem hence arises, which one cannot usually solve analytically; in fact, there is no guarantee that a solution exists. So, one has to resort to empirical correlations, this being the main shortcoming of the method.

Besides these two approaches, there is a third that can be regarded as a hybrid between them. Averaged equations of motion are used for the fluid phase, but rigid-body Newtonian equations are solved for each particle of the system. These do not interact with the fluid through its microscopic velocity field, but with the averaged value of the latter. For instance, the overall force exerted by the fluid on each particle is not computed by integrating over the particle surface the local traction arising from the fluid velocity gradients. The force is instead evaluated in terms of slip velocity between the averaged fluid velocity and the velocity of the particle center of mass and by resorting to empirical correlations. This strategy, called discrete particle modeling, is significantly less demanding computationally than the Eulerian-Lagrangian and has met with resounding success (Tsuji et al., 1993; Hoomans et al., 1996; Xu & Yu, 1997; Ouyang & Li, 1999; Kafui et al., 2002; Lu et al., 2005; Pandit et al., 2005; Ye et al., 2005).

To describe particle collisions, modelers use two approaches: hard and soft sphere. In the first, particles interact via binary, instantaneous, pointwise collisions. Their velocities after an encounter are computed by requiring that linear and angular momentum are conserved in the collision. This approach was pioneered by Allen & Tildesley (1990). Since their publication many authors have found it useful to model the collision dynamics in gran-

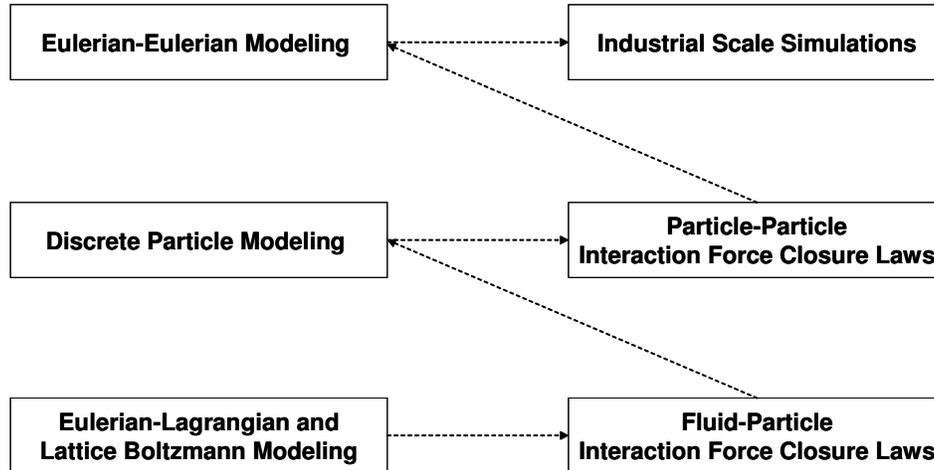


Figure 1. Multiscale modeling strategy.

ular systems. Hoomans et al. (1996) used it in their model for gas-fluidized beds. It was the first time that the technique had been applied to a dense system. Many authors have since published papers using this strategy (see, for instance, Ouyang & Li, 1999). The soft sphere model for fluid beds was instead pioneered by Tsuji et al. (1993), who developed their approach on the basis of earlier work done by Cundall & Strack (1979). Here, during an encounter, particles overlap slightly and the contact forces are calculated from the deformation history of the solids using a linear spring/dashpot model. This has been used by Xu & Yu (1997), Pandit et al. (2005), Ye et al. (2005) and several other researchers.

Among the three modeling approaches presented, the second is often preferred for its valuable feature of being computationally less demanding. Because of the number of particles involved in industrial plants, Eulerian (continuum) modeling is not likely to be replaced by its Lagrangian (discrete) counterparts in the foreseeable future. The role of discrete modeling is yet paramount. The method, to be considered more as an effective research tool than as a practical design instrument, by providing information about the dynamics of multiphase systems at the microscopic length scale, can significantly help develop and improve continuous average-based macroscopic models. In this view, Eulerian-Lagrangian modeling is to multiphase flows what direct numerical simulations are to turbulent flows. This multiscale modeling strategy, clearly described by van der Hoef et al. (2005), is shown schematically in Figure 1. However, how to link the models and extract from each the information needed by the others is still an open challenge.

3. An Overview of Averaging Theory

In the present review, we focus on three techniques that one can adopt to derive averages of point variables: statistical, volume and time averaging.

3.1. Statistical averaging

Statistical averaging is based on a fundamental feature of systems made up of large numbers of particles: it is impossible to know the precise locations and properties (for instance, linear momentum) of each individual constituent of the particle population at any given time. Furthermore, not only are these details impossible to measure, but are also usually unimportant for practical applications, since engineers are only interested in gross features of the system dynamics.

Consider a fluidized suspension of ν identical, spherical, smooth particles, and let $\mathbf{x}_s(t)$ and $\mathbf{v}_s(t)$ be the position vector and linear velocity of the s th particle center, respectively. At the reference time $t = 0$, the s th particle finds itself in the point \mathbf{x}_s^0 with velocity \mathbf{v}_s^0 . If $\mathbf{F}_s(t)$ denotes the unit mass force acting on the particle, as time progresses the latter moves obeying the equations:

$$\dot{\mathbf{x}}_s(t) = \mathbf{v}_s(t); \quad \dot{\mathbf{v}}_s(t) = \mathbf{F}_s(t); \quad \mathbf{x}_s(0) = \mathbf{x}_s^0; \quad \mathbf{v}_s(0) = \mathbf{v}_s^0 \quad (1)$$

Accordingly, if we know the initial conditions for each particle and the functional expressions of the forces $\mathbf{F}_s(t)$, by integrating the above differential equations we can predict with absolute certainty the particle positions and velocities at any future time.

However, solving this initial value problem for a number of particles of realistic order of magnitude, say, $\nu \sim 10^8$, is a prohibitive task, for we would have to know the initial data (that is, the position and velocity) for all the particles, and this appears to be impossible even in principle. In fact, it would involve measuring simultaneously the positions and velocities of all the particles at $t = 0$.

For systems comprising a great number of particles, therefore, we cannot know the initial state of each particle; thus, we cannot assign the initial conditions deterministically, as we have done in equation (1). What we usually know are only *macroscopic* – and therefore *measurable* – properties of the system, such as its local density, temperature or mean velocity. But there are infinite system configurations yielding the same macroscopic properties, each with a certain probability of occurrence. So, we must replace the deterministic initial conditions above with probabilistic initial conditions. To this end, we introduce a probability density function (PDF) defined so that:

$$p_\nu(\mathbf{x}_1, \dots, \mathbf{x}_\nu, \mathbf{v}_1, \dots, \mathbf{v}_\nu, t) d\mathbf{x}_1 \dots d\mathbf{x}_\nu d\mathbf{v}_1 \dots d\mathbf{v}_\nu \quad (2)$$

denotes the joint probability that at time t the first particle has position and velocity in the ranges $d\mathbf{x}_1$ and $d\mathbf{v}_1$ respectively around the points \mathbf{x}_1 and \mathbf{v}_1 , the second in the ranges $d\mathbf{x}_2$ and $d\mathbf{v}_2$ respectively around the points \mathbf{x}_2 and \mathbf{v}_2 and so on up to the ν th particle. Note that the arguments $\mathbf{x}_1, \dots, \mathbf{x}_\nu$ and $\mathbf{v}_1, \dots, \mathbf{v}_\nu$ of the PDF are independent variables identifying the positions and velocities of the particles, respectively. Here, to shorten the notation, we can use the vector $\mathbf{r} \equiv (\mathbf{x}_1, \dots, \mathbf{x}_\nu, \mathbf{v}_1, \dots, \mathbf{v}_\nu)$; then $p_\nu(\mathbf{r}, t) d\mathbf{r}$ represents the probability that at time t the system configuration falls in the range $d\mathbf{r}$ around the point \mathbf{r} . More concisely, $p_\nu(\bar{\mathbf{r}}, t) d\bar{\mathbf{r}} \equiv P\{\bar{\mathbf{r}} \leq \mathbf{r} < \bar{\mathbf{r}} + d\bar{\mathbf{r}}\}$, where $\bar{\mathbf{r}}$ denotes the sample-space variable of the system state vector \mathbf{r} . We usually refer to the function $p_\nu(\mathbf{r}, t)$ as the ν -particle joint PDF or master joint PDF. At any given time, we no longer know the exact configuration of the system, but the master joint PDF states how probable each configuration is. In particular,

for $t = 0$, the quantity $p_\nu(\mathbf{r}, 0) d\mathbf{r}$ yields the probability that the initial system configuration lies in the range $d\mathbf{r}$ around the point \mathbf{r} .

Another useful function is the one-particle marginal PDF, which gives the probability of finding a single particle in a differential neighborhood of a given state, independently of the states of the other $\nu - 1$ particles. There are ν one-particle marginal PDFs, one for each particle of the system. If $p_1^s(\mathbf{x}_s, \mathbf{v}_s, t)$ denotes the one related to the s th particle, by definition $p_1^s(\mathbf{x}_s, \mathbf{v}_s, t) d\mathbf{x}_s d\mathbf{v}_s$ gives the probability of finding the s th particle in the ranges $d\mathbf{x}_s$ and $d\mathbf{v}_s$ respectively around the points \mathbf{x}_s and \mathbf{v}_s .

We can reduce the ν -particle joint PDF to the one-particle marginal PDF by integrating out all the state variables (position and velocity) for the other $\nu - 1$ particles; because the particles are indistinguishable and their labeling is arbitrary, the ν -particle joint PDF is symmetrical with respect to the particle state variables and the ν one-particle marginal PDFs associated with each particle coincide. If we denote these ν identical functions by $p_1(\mathbf{x}_1, \mathbf{v}_1, t)$, it is:

$$p_1(\mathbf{x}_1, \mathbf{v}_1, t) \equiv \int_{\Omega_x} \dots \int_{\Omega_x} \int_{\Omega_v} \dots \int_{\Omega_v} p_\nu(\mathbf{r}, t) \prod_{s=2}^{\nu} d\mathbf{x}_s d\mathbf{v}_s \quad (3)$$

where Ω_x and Ω_v represent the ranges of variation of the particle positions and velocities, respectively; the former coincides with the region of physical space enclosed by the vessel containing the suspension, whereas the latter is unbounded and coincides with \mathbb{R}^3 .

Knowing the master joint PDF allows calculating any average associated with the fluidized suspension. A particularly important average function is the *expected* number density function (NDF). If we denote this by $f(\mathbf{x}, \mathbf{v}, t)$, by definition $f(\mathbf{x}, \mathbf{v}, t) d\mathbf{x} d\mathbf{v}$ represents the expected number of particles located in the range $d\mathbf{x}$ around \mathbf{x} with velocity in the range $d\mathbf{v}$ around \mathbf{v} at time t . Here *expected* means averaged statistically over all the possible system configurations. To find the expected NDF, we reason as follows. Given a system configuration with particle positions $\mathbf{x}_1, \dots, \mathbf{x}_\nu$ and velocities $\mathbf{v}_1, \dots, \mathbf{v}_\nu$, the associated actual NDF takes the following mathematical form:

$$\varphi(\mathbf{x}, \mathbf{v}, \mathbf{r}) \equiv \varphi(\mathbf{x}, \mathbf{v}, \mathbf{x}_1, \dots, \mathbf{x}_\nu, \mathbf{v}_1, \dots, \mathbf{v}_\nu) \equiv \sum_{s=1}^{\nu} \delta(\mathbf{x} - \mathbf{x}_s) \delta(\mathbf{v} - \mathbf{v}_s) \quad (4)$$

To derive the expected NDF we now have to average this function over all the possible system configurations, weighting each with its probability of occurrence; so, we write:

$$\begin{aligned} f(\mathbf{x}, \mathbf{v}, t) &\equiv \int_{\Omega_r} \varphi(\mathbf{x}, \mathbf{v}, \mathbf{r}) p_\nu(\mathbf{r}, t) d\mathbf{r} = \sum_{s=1}^{\nu} \int_{\Omega_r} \delta(\mathbf{x} - \mathbf{x}_s) \delta(\mathbf{v} - \mathbf{v}_s) p_\nu(\mathbf{r}, t) d\mathbf{r} \\ &= \sum_{s=1}^{\nu} \int_{\Omega_x} \int_{\Omega_v} \delta(\mathbf{x} - \mathbf{x}_s) \delta(\mathbf{v} - \mathbf{v}_s) p_1(\mathbf{x}_s, \mathbf{v}_s, t) d\mathbf{x}_s d\mathbf{v}_s = \nu p_1(\mathbf{x}, \mathbf{v}, t) \end{aligned} \quad (5)$$

where Ω_r denotes the range of variation of \mathbf{r} . So, the expected NDF coincides with the one-particle marginal PDF times the number of particles present in the fluidized suspension. This function allows calculating local averages of properties attached to single particles. At

any given time, the statistical average of any particle property at a specified spatial location \boldsymbol{x} is defined as the arithmetic mean of the instantaneous point property evaluated at \boldsymbol{x} at the time in question for each of the infinite number of system configurations. So, for a given particle property $\boldsymbol{\xi}(\boldsymbol{x}, \boldsymbol{v}, t)$, the average $\langle \boldsymbol{\xi} \rangle_s(\boldsymbol{x}, t)$ is defined so that:

$$\langle \boldsymbol{\xi} \rangle_s(\boldsymbol{x}, t) \int_{\Omega_v} f(\boldsymbol{x}, \boldsymbol{v}, t) d\boldsymbol{v} \equiv \int_{\Omega_v} \boldsymbol{\xi}(\boldsymbol{x}, \boldsymbol{v}, t) f(\boldsymbol{x}, \boldsymbol{v}, t) d\boldsymbol{v} \quad (6)$$

the integral on the left representing the particle number per unit volume, or equivalently the particle number density, at the chosen location \boldsymbol{x} . Note that the function $\langle \boldsymbol{\xi} \rangle_s(\boldsymbol{x}, t)$ is defined everywhere within Ω_x , even if the particulate system is discrete.

To find how the property $\langle \boldsymbol{\xi} \rangle_s(\boldsymbol{x}, t)$ evolves in time and space, we can either derive a transport equation for the expected NDF (the so-called Boltzmann equation) and then use equation (6) or try to find a transport equation for the mean property directly. We shall say more about the first strategy in §6., referring for further details to the abundant literature on the subject (Chapman & Cowling, 1970; Cercignani, 1988; Gidaspow, 1994; Brilliantov & Poschel, 2004; Marchisio & Fox, 2007).

3.2. Volume averaging

Volume averages are computed over spatial domains large enough to contain several particles but small compared with the scale of point-to-point macroscopic variations in which the modelers are interested and which they wish to capture. A volume $V(\boldsymbol{x})$ bounded by a surface $S(\boldsymbol{x})$ is attached to each spatial point \boldsymbol{x} ; within such a volume, the property of interest is averaged out by using the mean value theorem of integral calculus. When this averaging scheme is adopted, all the property values within $V(\boldsymbol{x})$ are accounted for and given the same weight in the average; conversely, the values outside $V(\boldsymbol{x})$ are ignored, that is, are given zero weight. These averages are usually referred to as hard volume averages to distinguish them from soft volume averages. The latter are based on an alternative technique, more elegant and convenient from a mathematical standpoint, that uses a radial weighting function: a continuous, monotone, decreasing function of the radial distance from the point wherein the average is evaluated. This mathematical device ascribes a weight to the property values within the whole physical domain. As a consequence, the average is no longer calculated by considering only the values attached to a well-defined averaging volume; on the contrary, all values are taken in consideration, each one suitably weighted.

Volume averaged variables thus defined appear to depend on the specific form chosen for the weighting function and, in particular, on its radius (that is, the size of the region over which the averaging effectively takes place; we shall give a formal definition of weighting function radius later on in §4.1.). Nevertheless, the larger the ratio between the smallest macroscopic length scale and the particle size, the more this dependence dwindles provided that the weighting function radius is properly chosen. If this radius is denoted by r_a , the particle radius by r_s and the aforementioned macroscopic length scale by r_m , the local average is expected to be insensitive to the particular form of the weighting function provided that $r_s \lll r_a \lll r_m$. Such a choice of r_a is, of course, possible only if r_m is far greater than r_s ; in such a case, there is said to be *separation of scales* between the macroscopic fluid dynamic problem and the detailed motion at the scale of a single particle. Only in this

instance the locally averaged variables do possess an unambiguous physical meaning and can be employed in the development of averaged equations of motion.

In section §4., we will derive the locally averaged transport equations for both the fluid and the particles using soft volume averages.

3.3. Time averaging

To explain time averaging, let us consider a field $\xi(\mathbf{x}, t)$; for any spatial fixed position $\bar{\mathbf{x}}$, $\xi(\bar{\mathbf{x}}, t)$ is a pure function of time that fluctuates irregularly. The time scale τ_t that represents these fluctuations is sometimes referred to as turbulent time scale. At each given location, we can obtain a mean value of $\xi(\mathbf{x}, t)$ by time averaging over a large number of fluctuations, considering a time interval τ_a much longer than the turbulent time scale. Again, we resort to the mean value theorem:

$$\langle \xi \rangle_t(\mathbf{x}, t) \equiv \frac{1}{\tau_a} \int_{t-\tau_a/2}^{t+\tau_a/2} \xi(\mathbf{x}, \tau) d\tau \quad (7)$$

where $\langle \xi \rangle_t(\mathbf{x}, t)$ denotes the time average and τ is a dummy integration variable. Also now, the mean value is expected to be insensitive to the averaging time scale provided that $\tau_t \lll \tau_a \lll \tau_m$, where τ_m represents the time scale of the mean flow fluctuations (refer to Delhaye & Achard, 1977, 1978).

4. Volume Averaged Equations of Motion for Fluidized Suspensions

Using soft volume averages, we now derive the Eulerian-Eulerian averaged equations of motion for dense fluidized suspensions. In doing so, we use the method advanced by Jackson (1997, 1998) for monodisperse fluidized suspensions and recently generalized by Owoyemi et al. (2007) and Mazzei (2008) for suspensions of any number of monodisperse particle classes. In this derivation, we consider only two particle classes, since further generalization is immediate; we also assume that the particles are inert (they neither react nor break, aggregate, etc.) and rigid, and that the fluid is incompressible.

4.1. Weighting functions

As mentioned, to define locally averaged variables, we use weighting functions. These are characterized by the following mathematical properties:

- 1) The weighting function ψ is a scalar function of r defined for $r > 0$, where r denotes the distance of a point \mathbf{y} from a point \mathbf{x} in the Euclidean space:

$$\psi = \psi(r); \quad r \equiv \|\mathbf{x} - \mathbf{y}\| \quad (8)$$

- 2) $\psi(r)$ is positive for any value of r , decreases monotonically with r and possesses continuous derivatives of any order.

- 3) $\psi(r)$ is normalized so that, if Ω_x denotes the spatial domain occupied by the system of interest (assumed here to stretch out to infinity), it is:

$$\int_{\Omega_x} \psi(\|\mathbf{x} - \mathbf{y}\|) d\mathbf{y} = 4\pi \int_0^{+\infty} \psi(r) r^2 dr = 1 \quad (9)$$

In the integral on the left-hand side, \mathbf{y} is the spatial variable of integration, whereas \mathbf{x} is the spatial position in which the volume average is computed.

4.2. Overall locally averaged variables

Given a generic point variable $\xi(\mathbf{x}, t)$ function of spatial position \mathbf{x} and time t , we define its overall local average calculated in \mathbf{x} at time t as:

$$\langle \xi \rangle_o(\mathbf{x}, t) \equiv \int_{\Omega_x} \xi(\mathbf{y}, t) \psi(\|\mathbf{x} - \mathbf{y}\|) d\mathbf{y} \quad (10)$$

To clarify this definition, we use two examples. Take the function $\psi(r) = 1/V_x$, where V_x is the volume of Ω_x assumed to be bounded. This expression is unacceptable, for $\psi(r)$ does not decrease monotonically with r as required; all the same, we adopt it temporarily to render the meaning of equation (10) more apparent. With this weighting function, equation (10) reduces to:

$$\langle \xi \rangle_o(t) = \frac{1}{V_x} \int_{\Omega_x} \xi(\mathbf{y}, t) d\mathbf{y} \quad (11)$$

This equation merely expresses the mean value theorem of integral calculus. The problem with $\psi(r)$ is that it averages over the *entire system* without differentiating between points that are near \mathbf{x} and points that are far; thus, the averaged value of $\xi(\mathbf{x}, t)$ computed in \mathbf{x} does not depend on \mathbf{x} ! In a proper definition of *local average*, points near \mathbf{x} should count more. To obtain a meaningful average, we set $\psi(r) = 1/V_a$ for $r \leq r_a$ and $\psi(r) = 0$ for $r > r_a$, where V_a denotes the volume of the spherical domain Ω_a of radius r_a centered in \mathbf{x} . With this choice, equation (10) yields:

$$\langle \xi \rangle_o(\mathbf{x}, t) = \frac{1}{V_a} \int_{\Omega_a} \xi(\mathbf{y}, t) d\mathbf{y} \quad (12)$$

This other definition of $\psi(r)$ ensures that only points within the spherical neighborhood Ω_a of \mathbf{x} of radius r_a are considered in the average; the remaining ones are neglected, so that the average is local. This weighting function defines hard spatial averages, inasmuch as $\psi(r)$ is discontinuous in r_a . Soft spatial averages, based on continuous weighting functions that meet the requirements 1) to 3), are more convenient, for they allow us to use all the theorems of analytical calculus. For continuous weighting functions, the averaging radius is defined as the scalar r_a that satisfies the equation:

$$4\pi \int_0^{r_a} \psi(r) r^2 dr = 4\pi \int_{r_a}^{+\infty} \psi(r) r^2 dr = \frac{1}{2} \quad (13)$$

Hence, r_a is a measure of the linear size of the spherical neighborhood of \mathbf{x} wherein the spatial points have appreciable weight in the averaging process. As already mentioned, we must choose $\psi(r)$ so that the value of r_a yielded by equation (13) fulfills the condition $r_s \lll r_a \lll r_m$.

4.3. Fluid phase locally averaged variables

The void fraction or fraction of space occupied by the fluid (here void means solid-free) and the fluid phase average of $\xi(\mathbf{x}, t)$ calculated in \mathbf{x} at time t are so defined:

$$\varepsilon(\mathbf{x}, t) \equiv \int_{\Omega_e} \psi(\|\mathbf{x} - \mathbf{y}\|) d\mathbf{y} ; \quad \varepsilon(\mathbf{x}, t) \langle \xi \rangle_e(\mathbf{x}, t) \equiv \int_{\Omega_e} \xi(\mathbf{y}, t) \psi(\|\mathbf{x} - \mathbf{y}\|) d\mathbf{y} \quad (14)$$

In the equations above, Ω_e represents the domain occupied by the fluid phase at time t (we have left out the explicit dependence on t to simplify the notation). Note that, since both the fluid and the solid particles are incompressible, the volume that they occupy is constant and therefore time-independent. But Ω_e is not a physical volume: it represents a domain of integration; accordingly, its shape and position are important and need to be taken into account. Since both fluid and solid particles move continuously, the shape and position of the domains that they occupy vary in time. These changes affect the integrals above, especially their time derivatives. To distinguish between particle volume and domain of integration, we shall denote the former by V_i and the latter by Ω_i . Consequently, V_i is a property of the particles of phase \mathcal{F}_i and is time-independent; conversely, Ω_i is the time-dependent domain of integration bounded by the surface $\partial\Omega_i$ marking the region of physical space occupied by a particle of phase \mathcal{F}_i at the time of interest.

We can obtain the fluid phase averages of spatial and time derivatives of point variables by generalizing the relationships originally derived by Anderson & Jackson (1967) for monodisperse suspensions; they are given by (proof is provided in the appendix):

$$\begin{aligned} \varepsilon(\mathbf{x}, t) \left\langle \frac{\partial \xi}{\partial x_a} \right\rangle_e(\mathbf{x}, t) &= \frac{\partial}{\partial x_a} \left[\varepsilon(\mathbf{x}, t) \langle \xi \rangle_e(\mathbf{x}, t) \right] - \varphi_x \left[\xi(\mathbf{x}, t), a \right] ; \\ \varepsilon(\mathbf{x}, t) \left\langle \frac{\partial \xi}{\partial t} \right\rangle_e(\mathbf{x}, t) &= \frac{\partial}{\partial t} \left[\varepsilon(\mathbf{x}, t) \langle \xi \rangle_e(\mathbf{x}, t) \right] + \varphi_t \left[\xi(\mathbf{x}, t) \right] \end{aligned} \quad (15)$$

where it is:

$$\begin{aligned} \varphi_x \left[\xi(\mathbf{x}, t), a \right] &\equiv \sum_{i=1}^2 \sum_{\mathcal{F}_i} \int_{\partial\Omega_i} \xi(\mathbf{y}, t) k_a(\mathbf{y}, t) \psi(\|\mathbf{x} - \mathbf{y}\|) dS_y ; \\ \varphi_t \left[\xi(\mathbf{x}, t) \right] &\equiv \sum_{i=1}^2 \sum_{\mathcal{F}_i} \int_{\partial\Omega_i} \xi(\mathbf{y}, t) \mathbf{u}(\mathbf{y}, t) \cdot \mathbf{k}(\mathbf{y}, t) \psi(\|\mathbf{x} - \mathbf{y}\|) dS_y \end{aligned} \quad (16)$$

In these equations, the vectors $\mathbf{k}(\mathbf{x}, t)$ and $\mathbf{u}(\mathbf{x}, t)$ are the outward unit normal to $\partial\Omega_i$ and the fluid velocity, respectively, and $k_a(\mathbf{x}, t)$ is the a th component of $\mathbf{k}(\mathbf{x}, t)$ with respect to a generic orthonormal vector basis. Also, the summation is over all the particles of phase \mathcal{F}_i .

4.4. Solid phase locally averaged variables

The volume fraction of the generic particle species \mathcal{F}_i and the solid phase average of $\xi(\mathbf{x}, t)$ calculated in \mathbf{x} at time t are so defined:

$$\phi_i(\mathbf{x}, t) \equiv \sum_{\mathcal{F}_i} \int_{\Omega_i} \psi(\|\mathbf{x} - \mathbf{y}\|) d\mathbf{y};$$

$$\phi_i(\mathbf{x}, t) \langle \xi \rangle_s^i(\mathbf{x}, t) \equiv \sum_{\mathcal{F}_i} \int_{\Omega_i} \xi(\mathbf{y}, t) \psi(\|\mathbf{x} - \mathbf{y}\|) d\mathbf{y} \quad (17)$$

This average, employed by several researchers (Enwald et al., 1996; Crowe et al., 1997; Drew & Passman, 1998; Fan & Zhu, 1998), operates on the microscopic properties of the particle material, considering point fields $\xi(\mathbf{x}, t)$ that vary within the particles. It is an average that exactly parallels the one just given for the fluid. This approach, however, is not always straightforward and presents conceptual difficulties when the particles are rigid (as in the present treatment) or massless (for instance, in bubble columns); in these cases, artifices are necessary to overcome these difficulties (Batchelor, 1970; Hinch, 1977; Sangani & Didwania, 1993; Zhang & Prosperetti, 1994). Thus, to derive the averaged transport equations for the particles, we do not use this average, but favor an other based on properties $\xi_i(t)$ of the particles *as a whole*, as opposed to the point properties $\xi(\mathbf{x}, t)$ varying *within the particles* used in equation (17).

4.5. Particle phase locally averaged variables

Since the particles are rigid, their motion is determined by the translation of their centers of mass and by the rotation of their bodies about instantaneous axes of rotation. Thus, the resultant forces and torques acting on the particles suffice to establish their motion. We can then introduce a different kind of local average that depends only on the properties of the particles as a whole. We define the number density of particles of class \mathcal{F}_i calculated in \mathbf{x} at time t as follows:

$$n_i(\mathbf{x}, t) \equiv \sum_{\mathcal{F}_i} \psi(\|\mathbf{x} - \mathbf{x}_i\|) \quad (18)$$

where $\mathbf{x}_i(t)$ denotes the position occupied at time t by the center of mass of a generic particle of the phase under consideration. The volume fraction $\phi_i(\mathbf{x}, t)$ is related to the number density $n_i(\mathbf{x}, t)$ by the following relationship (Mazzei, 2008):

$$\phi_i(\mathbf{x}, t) = n_i(\mathbf{x}, t) V_i + O(r_s^2/r_m^2) \quad (19)$$

r_s being the particle radius and r_m the macroscopic length scale of interest. The last term on the right-hand side can be safely neglected if there is separation of scale between the macroscopic motion of the suspension and the microscopic motion of the particles.

Generalizing the averaging scheme of Jackson (1997), we define the particle phase average for the phase \mathcal{F}_i of a particle property $\xi_i(t)$ calculated in \mathbf{x} at time t as:

$$n_i(\mathbf{x}, t) \langle \xi \rangle_p^i(\mathbf{x}, t) \equiv \sum_{\mathcal{F}_i} \left[\xi_i(t) \psi(\|\mathbf{x} - \mathbf{x}_i\|) \right] \quad (20)$$

The particle phase average of a particle property time derivative is:

$$n_i(\mathbf{x}, t) \langle \dot{\xi} \rangle_p^i(\mathbf{x}, t) = \frac{\partial}{\partial t} \left[n_i(\mathbf{x}, t) \langle \xi \rangle_p^i(\mathbf{x}, t) \right] + \frac{\partial}{\partial x_a} \sum_{\mathcal{F}_i} \left[n_i(\mathbf{x}, t) \langle \xi v_a \rangle_p^i(\mathbf{x}, t) \right] \quad (21)$$

where $\langle \xi v_a \rangle_p^i(\mathbf{x}, t)$ is the average of the product of $\xi_i(t)$ and the a th component of the center velocity $v_i(t)$. Here and in what follows, we adopt the convention that repeated lower indices are summed over the values one to three, with the exception of i and r , used as phase indices, and of e and p , used to specify the average type. Until now, we have shown the dependence on time and position; from now on, to simplify the notation, we shall sometimes leave it out.

4.6. Averaged equations of motion for the fluid phase

We now apply the definitions and mathematical relationships just presented to derive the macroscopic locally averaged transport equations for the fluid phase.

Continuity equation

To derive this equation, we set $\xi = u_a$ and $\xi = 1$ in the first and second equations in (15), respectively, and then add the results. Because the fluid is incompressible, doing so yields:

$$\frac{\partial \varepsilon}{\partial t} + \frac{\partial}{\partial x_a} \left(\varepsilon \langle u_a \rangle_e \right) = 0 \quad (22)$$

In this equation, as expected, the fluid volume fraction takes the role that the fluid density has in single-phase compressible fluids.

Dynamical equation

Microscopically, the fluid satisfies the Navier-Stokes equation:

$$\rho_e \left[\frac{\partial u_a}{\partial t} + \frac{\partial}{\partial x_b} (u_a u_b) \right] = \frac{\partial T_{ab}}{\partial x_b} + \rho_e g_a \quad (23)$$

where ρ_e is the fluid density, $T_{ab}(\mathbf{x}, t)$ is the ab th component of the point fluid stress tensor and g_a is the a th component of the gravitational field. To average this equation, we multiply both sides by $\psi(\|\mathbf{x} - \mathbf{y}\|)$ and integrate over Ω_e with respect to \mathbf{y} . To treat the left-hand side of equation (23), we write equations (15) with $\xi = u_a u_b$ in the first and $\xi = u_a$ in the second; to treat the right-hand side, we use the first of equations (14) and (15) with $\xi = T_{ab}$. The result is:

$$\rho_e \left[\frac{\partial}{\partial t} \left(\varepsilon \langle u_a \rangle_e \right) + \frac{\partial}{\partial x_b} \left(\varepsilon \langle u_a u_b \rangle_e \right) \right] = \frac{\partial}{\partial x_b} \left(\varepsilon \langle T_{ab} \rangle_e \right) + \varepsilon \rho_e g_a - \varphi_x \left[T_{ab}(\mathbf{x}, t), b \right] \quad (24)$$

The last term on the right-hand side is the sum over all particle classes of the mean resultant traction force exerted by the fluid on the particles of each class. The force:

$$\sum_{\mathcal{F}_i} \int_{\partial\Omega_i} T_{ab}(\mathbf{y}, t) k_b(\mathbf{y}, t) \psi(\|\mathbf{x} - \mathbf{y}\|) dS_y \quad (25)$$

is the sum of the average resultant forces exerted by the fluid on the i th phase particles. In order to compute this force for each particle, we firstly weight the differential traction forces acting on each infinitesimal part dS_y of the particle surface using $\psi(\|\mathbf{x} - \mathbf{y}\|)$ at the locations under consideration, and then we sum all the contributions. The fluid-solid interaction force, defined by equation (25), couples the dynamical equation of the fluid to those of each particle class.

It is convenient to transform equation (25) from a situation where the differential traction forces exerted on every infinitesimal surface element dS_y of the fluid-particle interface are firstly weighted using the values $\psi(\|\mathbf{x} - \mathbf{y}\|)$ of the weighting function at the locations of the elements dS_y and then summed, to one where the overall force acting on the entire surface $\partial\Omega_i$ of each particle is first calculated and then weighted using the value $\psi(\|\mathbf{x} - \mathbf{x}_i\|)$ of the weighting function at the particle center. This second average interprets better the fluid-particle interaction force and *fulfills the principle of action and reaction*. To this end, we expand $\psi(\|\mathbf{x} - \mathbf{y}\|)$ in a Taylor series about the particle center $\mathbf{x}_i(t)$. Doing so gives:

$$\begin{aligned} \psi(\|\mathbf{x} - \mathbf{y}\|) &= \psi(\|\mathbf{x} - \mathbf{x}_i\|) - \frac{\partial\psi}{\partial x_b}(\|\mathbf{x} - \mathbf{x}_i\|)(y_b - x_{i,b}) \\ &\quad + \frac{1}{2} \frac{\partial^2\psi}{\partial x_b \partial x_c}(\|\mathbf{x} - \mathbf{x}_i\|)(y_b - x_{i,b})(y_c - x_{i,c}) - \dots \quad (26) \end{aligned}$$

At any spatial location within each particle, we can truncate the Taylor series at the second order term with acceptably small error. Since it is:

$$\forall \mathbf{y} \in \partial\Omega_i : \mathbf{y} - \mathbf{x}_i = r_i \mathbf{k}(\mathbf{y}) \quad (27)$$

where $\partial\Omega_i$ represents the surface bounding the particle, $\mathbf{k}(\mathbf{y})$ is the outward unit normal to $\partial\Omega_i$ and r_i is the radius of the particles of species \mathcal{F}_i , we can write:

$$\begin{aligned} \forall \mathbf{y} \in \partial\Omega_i : \psi(\|\mathbf{x} - \mathbf{y}\|) &\approx \psi(\|\mathbf{x} - \mathbf{x}_i\|) \\ &\quad - \frac{\partial\psi}{\partial x_b}(\|\mathbf{x} - \mathbf{x}_i\|) r_i k_b(\mathbf{y}) + \frac{1}{2} \frac{\partial^2\psi}{\partial x_b \partial x_c}(\|\mathbf{x} - \mathbf{x}_i\|) r_i^2 k_b(\mathbf{y}) k_c(\mathbf{y}) \quad (28) \end{aligned}$$

Using this relation, we approximate the force in equation (25) as:

$$n_i \langle f_a \rangle_p^i - \frac{\partial}{\partial x_b} \left(n_i \langle A_{ab} \rangle_p^i \right) + \frac{1}{2} \frac{\partial^2}{\partial x_b \partial x_c} \left(n_i \langle B_{abc} \rangle_p^i \right) \quad (29)$$

where it is:

$$n_i \langle f_a \rangle_p^i(\mathbf{x}, t) \equiv \sum_{\mathcal{F}_i} \left[\psi(\|\mathbf{x} - \mathbf{x}_i\|) \int_{\partial\Omega_i} T_{ad}(\mathbf{y}, t) k_d(\mathbf{y}, t) dS_y \right] \quad (30)$$

$$n_i \langle A_{ab} \rangle_p^i(\mathbf{x}, t) \equiv r_i \sum_{\mathcal{F}_i} \left[\psi(\|\mathbf{x} - \mathbf{x}_i\|) \int_{\partial\Omega_i} T_{ad}(\mathbf{y}, t) k_d(\mathbf{y}, t) k_b(\mathbf{y}, t) dS_y \right] \quad (31)$$

$$n_i \langle B_{abc} \rangle_p^i(\mathbf{x}, t) \equiv r_i^2 \sum_{\mathcal{F}_i} \left[\psi(\|\mathbf{x} - \mathbf{x}_i\|) \int_{\partial\Omega_i} T_{ad}(\mathbf{y}, t) k_d(\mathbf{y}, t) k_b(\mathbf{y}, t) k_c(\mathbf{y}, t) dS_y \right] \quad (32)$$

The quantities defined above are the components of a vector, a second-order tensor and a third-order tensor, respectively. Using the Reynolds decomposition for the average of the dyadic product on the left-hand side of equation (24), we can write:

$$\langle u_a u_b \rangle_e \approx \langle u_a \rangle_e \langle u_b \rangle_e + \langle \hat{u}_a \hat{u}_b \rangle_e \quad (33)$$

$\hat{u}_a(\mathbf{x}, t)$ and $\hat{u}_b(\mathbf{x}, t)$ representing the deviations of the velocity components $u_a(\mathbf{x}, t)$ and $u_b(\mathbf{x}, t)$ from their respective means $\langle u_a \rangle_e(\mathbf{x}, t)$ and $\langle u_b \rangle_e(\mathbf{x}, t)$, respectively. Note that this relation is not exact, but holds only when point variations can be decomposed into contributions of length scales much smaller and much larger than the weighting function radius (Anderson & Jackson, 1967). Introducing equations (29) and (33) into equation (24), after few manipulations, we obtain:

$$\rho_e \left[\frac{\partial}{\partial t} (\varepsilon \langle u_a \rangle_e) + \frac{\partial}{\partial x_b} (\varepsilon \langle u_a \rangle_e \langle u_b \rangle_e) \right] = \frac{\partial \langle S_{ab} \rangle_e}{\partial x_b} - \sum_{i=1}^2 \left(n_i \langle f_a \rangle_p^i \right) + \varepsilon \rho_e g_a \quad (34)$$

where, by definition, it is:

$$\langle S_{ab} \rangle_e(\mathbf{x}, t) \equiv \varepsilon \langle T_{ab} \rangle_e + \sum_{i=1}^2 \left[n_i \langle A_{ab} \rangle_p^i - \frac{1}{2} \frac{\partial}{\partial x_c} \left(n_i \langle B_{abc} \rangle_p^i \right) \right] - \varepsilon \rho_e \langle \hat{u}_a \hat{u}_b \rangle_e \quad (35)$$

This term is the fluid phase effective stress tensor. The quantity $\langle \hat{u}_a \hat{u}_b \rangle_e(\mathbf{x}, t)$ is analogous to the Reynolds stresses of turbulent flows and is related to the deviations of the fluid velocity from its average value. Finding an analytical closure for the effective fluid stress tensor is extremely complex; however, a closure has been derived by Jackson (1997) on theoretical grounds for the limiting case of diluted, Stokesian monodisperse suspensions fluidized by Newtonian fluids. We shall present suitable closures for the terms on the right-hand side of equation (34) later on in §5.

4.7. Averaged equations of motion for the particle phases

We now use a similar strategy to derive the averaged transport equations for each particle class in the system. To do so, we refer to the generic particle phase \mathcal{F}_i .

Continuity equation

We derive the equation by simply setting $\xi_i(t) = 1$ in equation (21); this yields:

$$\frac{\partial n_i}{\partial t} + \frac{\partial}{\partial x_a} \left(n_i \langle v_a \rangle_p^i \right) = 0 \quad (36)$$

Here the particle number density (or equivalently its volume fraction) takes the role that the fluid density has in single-phase compressible fluids.

Dynamical equation

To derive this equation, we start from the linear momentum balance equation written for a generic particle of species \mathcal{F}_i . The forces at play are the traction force exerted by the fluid on the particle, the forces resulting from the collisions between the particle in question and particles of the same and of different species at their mutual points of contact and the effect of gravity. So, we write:

$$\rho_i V_i \dot{v}_{i,a}(t) = \int_{\partial\Omega_i} T_{ab}(\mathbf{y}, t) k_b(\mathbf{y}, t) dS_y + \sum_{r=1}^2 \sum_{\mathcal{F}_r} f_{ir,a}(t) + \rho_i V_i g_a \quad (37)$$

where ρ_i is the density of the particles of the i th phase, $\dot{v}_{i,a}(t)$ is the a th component of the acceleration of the particle center of mass and $f_{ir,a}(t)$ is the a th component of the force exerted by the generic particle r of phase \mathcal{F}_r on the particle i under consideration when a collision takes place. Note that $f_{ir,a}(t) \neq 0$ only if particles i and r are in direct contact.

To average equation (37), we multiply both sides by $\psi(\|\mathbf{x} - \mathbf{x}_i\|)$ and sum over all the particles i that belong to the phase under consideration. Doing so gives:

$$\begin{aligned} \rho_i V_i \sum_{\mathcal{F}_i} \left[\psi(\|\mathbf{x} - \mathbf{x}_i\|) \dot{v}_{i,a}(t) \right] &= \sum_{\mathcal{F}_i} \left[\psi(\|\mathbf{x} - \mathbf{x}_i\|) \int_{\partial\Omega_i} T_{ab}(\mathbf{y}, t) k_b(\mathbf{y}, t) dS_y \right] \\ &+ \sum_{\mathcal{F}_i} \left[\psi(\|\mathbf{x} - \mathbf{x}_i\|) \sum_{r=1}^2 \sum_{\mathcal{F}_r} f_{ir,a}(t) \right] + \rho_i V_i g_a \sum_{\mathcal{F}_i} \psi(\|\mathbf{x} - \mathbf{x}_i\|) \end{aligned} \quad (38)$$

We now use equations (18), (20), (21) and (30), choosing in the second $\xi_i(t) = \dot{v}_{i,a}(t)$ and in the third $\xi_i(t) = v_{i,a}(t)$, and employ the relation:

$$\sum_{\mathcal{F}_i} \left[\psi(\|\mathbf{x} - \mathbf{x}_i\|) \sum_{r=1}^2 \sum_{\mathcal{F}_r} f_{ir,a}(t) \right] = \sum_{r=1}^2 \sum_{\mathcal{F}_i} \left[\psi(\|\mathbf{x} - \mathbf{x}_i\|) \sum_{\mathcal{F}_r} f_{ir,a}(t) \right] \quad (39)$$

Then equation (38) becomes:

$$\begin{aligned} \rho_i V_i \left[\frac{\partial}{\partial t} \left(n_i \langle v_a \rangle_p^i \right) + \frac{\partial}{\partial x_b} \left(n_i \langle v_a v_b \rangle_p^i \right) \right] &= n_i \langle f_a \rangle_p^i \\ &+ n_i \rho_i V_i g_a + \sum_{r=1}^2 \sum_{\mathcal{F}_i} \left[\psi(\|\mathbf{x} - \mathbf{x}_i\|) \sum_{\mathcal{F}_r} f_{ir,a}(t) \right] \end{aligned} \quad (40)$$

The first term on the right-hand side is the fluid-particle force of interaction, which has its exact analogue in equation (34), while the last term combines the resultant forces arising from the particle-particle contacts between particles belonging to the same phase ($r = i$) and particles belonging to different phases ($r \neq i$). These contributions are conceptually different: the former is a self-interaction term that represents the stress internal to the phase under examination, while the latter is a contact force acting between the Eulerian solid phases. In order for the collisional solid stress tensor associated to phase \mathcal{F}_i to appear explicitly in equation (40), further mathematical manipulations are required. We start by considering the following double sum over the particles i and r of the i th phase:

$$\sum_{\mathcal{F}_i} \sum_{\mathcal{F}_i} \left[\psi(\|\mathbf{x} - \mathbf{x}_{ir}\|) f_{ir,a}(t) \right] \quad (41)$$

where $\mathbf{x}_{ir}(t)$ denotes the position vector of the point of mutual contact between the rigid particles i and r , both belonging to *the same* phase \mathcal{F}_i . This double sum clearly vanishes, because $\mathbf{x}_{ir}(t) = \mathbf{x}_{ri}(t)$ and, for the principle of action and reaction, $f_{ir,a}(t) = -f_{ri,a}(t)$. By expanding the function $\psi(\|\mathbf{x} - \mathbf{x}_{ir}\|)$ in a Taylor series about the center $\mathbf{x}_i(t)$ of the particles, we find:

$$\sum_{\mathcal{F}_i} \left[\psi(\|\mathbf{x} - \mathbf{x}_i\|) \sum_{\mathcal{F}_i} f_{ir,a}(t) \right] \approx \frac{\partial}{\partial x_b} \left[n_i \langle M_{ab} \rangle_p^i - \frac{1}{2} \frac{\partial}{\partial x_c} \left(n_i \langle N_{abc} \rangle_p^i \right) \right] \quad (42)$$

where, with $\mathbf{k}_{ir}(t)$ denoting the unit vector of $\mathbf{x}_{ir}(t) - \mathbf{x}_i(t)$, it is:

$$n_i \langle M_{ab} \rangle_p^i(\mathbf{x}, t) \equiv r_i \sum_{\mathcal{F}_i} \left\{ \psi(\|\mathbf{x} - \mathbf{x}_i\|) \sum_{\mathcal{F}_i} \left[f_{ir,a}(t) k_{ir,b}(t) \right] \right\} \quad (43)$$

$$n_i \langle N_{abc} \rangle_p^i(\mathbf{x}, t) \equiv r_i^2 \sum_{\mathcal{F}_i} \left\{ \psi(\|\mathbf{x} - \mathbf{x}_i\|) \sum_{\mathcal{F}_i} \left[f_{ir,a}(t) k_{ir,b}(t) k_{ir,c}(t) \right] \right\} \quad (44)$$

The second-order tensor so defined:

$$\langle C_{ab} \rangle_p^i(\mathbf{x}, t) \equiv n_i \langle M_{ab} \rangle_p^i(\mathbf{x}, t) - \frac{1}{2} \frac{\partial}{\partial x_c} \left[n_i \langle N_{abc} \rangle_p^i(\mathbf{x}, t) \right] \quad (45)$$

is the collisional stress tensor of the i th particle phase which accounts for the transfer of linear momentum at collisions between alike particles over the distance $2r_i$ separating their centers. This physical phenomenon is important in dense fluidized suspensions, where the total volume occupied by the particles is not negligible compared with the volume of the vessel containing them. For rarefied granular gases, which one can model adopting the Boltzmann-Grad limit defined as:

$$r_i \rightarrow 0; \quad \nu_i \rightarrow \infty; \quad \nu_i r_i^2 \text{ bounded} \quad (46)$$

where ν_i denotes the overall number of particles belonging to the i th phase, since $\mathbf{x}_i(t) \rightarrow \mathbf{x}_{ir}(t) \rightarrow \mathbf{x}_r(t)$, the collisional stress vanishes. This is consequence of the principle of

action and reaction, inasmuch as in the Boltzmann-Grad limit we find:

$$\sum_{\mathcal{F}_i} \left[\psi(\|\mathbf{x} - \mathbf{x}_i\|) \sum_{\mathcal{F}_i} f_{ir,a}(t) \right] \rightarrow \sum_{\mathcal{F}_i} \sum_{\mathcal{F}_i} \left[\psi(\|\mathbf{x} - \mathbf{x}_{ir}\|) f_{ir,a}(t) \right] = 0 \quad (47)$$

Let us now consider the second contribution to the overall particle-particle contact force that appears on the right-hand side of equation (40). We express this term, which represents the contact forces acting between the particles i of the phase \mathcal{F}_i under examination and the particles r of the other phase \mathcal{F}_r of the fluidized suspension, as:

$$\sum_{\mathcal{F}_i} \left[\psi(\|\mathbf{x} - \mathbf{x}_i\|) \sum_{\mathcal{F}_r} f_{ir,a}(t) \right] \quad (48)$$

where $r \neq i$ and with particles i and r belonging to phases \mathcal{F}_i and \mathcal{F}_r , respectively. Given its definition, this force should fulfill the principle of action and reaction, so that:

$$\sum_{\mathcal{F}_i} \left[\psi(\|\mathbf{x} - \mathbf{x}_i\|) \sum_{\mathcal{F}_r} f_{ir,a}(t) \right] = - \sum_{\mathcal{F}_r} \left[\psi(\|\mathbf{x} - \mathbf{x}_r\|) \sum_{\mathcal{F}_i} f_{ri,a}(t) \right] \quad (49)$$

Clearly, this condition is not met, for even if Newton's third law of mechanics states that $f_{ir,a}(t) = -f_{ri,a}(t)$, it is $\psi(\|\mathbf{x} - \mathbf{x}_i\|) \neq \psi(\|\mathbf{x} - \mathbf{x}_r\|)$. Only for $r_i \rightarrow 0$ and $r_r \rightarrow 0$ this equation holds. We conclude that the force on the left-hand side of the above equation cannot be regarded as the interaction force between the two particle phases, but has to encompass an additional contribution that does not satisfy the action and reaction principle. To find this force, which will prove to be the divergence of the collisional stress tensor of the i th particle phase accounting for the transfer of linear momentum at collisions between particles i and r over the distance $r_i + r_r$ separating their centers, we expand $\psi(\|\mathbf{x} - \mathbf{x}_{ir}\|)$ in a Taylor series about the point $\mathbf{x}_i(t)$ in which the center of particle i is located. This gives:

$$\sum_{\mathcal{F}_i} \left[\psi(\|\mathbf{x} - \mathbf{x}_i\|) \sum_{\mathcal{F}_r} f_{ir,a}(t) \right] \approx n_i \langle f_a \rangle_p^{ir} + \frac{\partial}{\partial x_b} \left[n_i \langle P_{ab} \rangle_p^i - \frac{1}{2} \frac{\partial}{\partial x_c} \left(n_i \langle Q_{abc} \rangle_p^i \right) \right] \quad (50)$$

where it is:

$$n_i \langle f_a \rangle_p^{ir} \equiv \sum_{\mathcal{F}_i} \left\{ \psi(\|\mathbf{x} - \mathbf{x}_{ir}\|) \sum_{\mathcal{F}_r} f_{ir,a}(t) \right\} \quad (51)$$

$$n_i \langle P_{ab} \rangle_p^i(\mathbf{x}, t) \equiv r_i \sum_{\mathcal{F}_i} \left\{ \psi(\|\mathbf{x} - \mathbf{x}_i\|) \sum_{\mathcal{F}_r} \left[f_{ir,a}(t) k_{ir,b}(t) \right] \right\} \quad (52)$$

$$n_i \langle Q_{abc} \rangle_p^i(\mathbf{x}, t) \equiv r_i^2 \sum_{\mathcal{F}_i} \left\{ \psi(\|\mathbf{x} - \mathbf{x}_i\|) \sum_{\mathcal{F}_r} \left[f_{ir,a}(t) k_{ir,b}(t) k_{ir,c}(t) \right] \right\} \quad (53)$$

The second-order tensor so defined:

$$\langle D_{ab} \rangle_p^i(\mathbf{x}, t) \equiv n_i \langle P_{ab} \rangle_p^i(\mathbf{x}, t) - \frac{1}{2} \frac{\partial}{\partial x_c} \left[n_i \langle Q_{abc} \rangle_p^i(\mathbf{x}, t) \right] \quad (54)$$

is the collisional stress tensor previously mentioned, related to momentum transfer at collisions between the two granular phases. Introducing equations (42) and (50) into equation (40), and applying the Reynolds decomposition to the advection term, we obtain:

$$\begin{aligned} \rho_i V_i \left[\frac{\partial}{\partial t} \left(n_i \langle v_a \rangle_p^i \right) + \frac{\partial}{\partial x_b} \left(n_i \langle v_a \rangle_p^i \langle v_b \rangle_p^i \right) \right] \\ = \frac{\partial \langle S_{ab} \rangle_p^i}{\partial x_b} + n_i \langle f_a \rangle_p^i + n_i \langle f_a \rangle_p^{ir} + n_i \rho_i V_i g_a \end{aligned} \quad (55)$$

where, by definition, it is:

$$\langle S_{ab} \rangle_p^i(\mathbf{x}, t) \equiv \langle C_{ab} \rangle_p^i(\mathbf{x}, t) + \langle D_{ab} \rangle_p^i(\mathbf{x}, t) - n_i \rho_i V_i \langle \hat{v}_a \hat{v}_b \rangle_p^i \quad (56)$$

The first two terms on the right-hand side together represent the total collisional stress tensor, while the last term represents the kinetic stress tensor, related to the deviations of the particle velocities about their mean. $\langle S_{ab} \rangle_p^i(\mathbf{x}, t)$ is the particle phase effective stress tensor, while $n_i \langle f_a \rangle_p^{ir}(\mathbf{x}, t)$, which fulfills the principle of action and reaction, is the effective interaction force between the granular phases i and r .

Table 1 reports, in absolute notation, the multifluid equations of motion for mixtures of n particle classes, which are an immediate generalization of the equations just derived for bidisperse mixtures. To write them, we have used equation (19). Note that in the dynamical equation for phase \mathcal{F}_i , the index r appearing in the summation must be different from the index i .

5. The Problem of Closure

As previously said, the process of averaging generates a number of indeterminate terms not directly related to the mean variables but still associated with details of the motion at the microscopic length scale. A clear example is given by the fluid-particle interaction force:

$$n_i \langle \mathbf{f} \rangle_p^i(\mathbf{x}, t) \equiv \sum_{\mathcal{F}_i} \left[\psi(\|\mathbf{x} - \mathbf{x}_i\|) \int_{\partial\Omega_i} \mathbf{T}(\mathbf{y}, t) \cdot \mathbf{k}(\mathbf{y}) dS_y \right] \quad (57)$$

To compute this force, we need to know the point fluid stress tensor $\mathbf{T}(\mathbf{x}, t)$, related to the microscopic fluid velocity field $\mathbf{u}(\mathbf{x}, t)$ and not its mean value $\langle \mathbf{u} \rangle_e(\mathbf{x}, t)$. The closure problem that arises is extremely difficult to tackle on purely theoretical grounds and has to be overcome by means of semitheoretical expressions. In this section, we overview this subject.

Table 1. Eulerian-Eulerian averaged equations of motion for a system of ν particle classes

Continuity equation - Fluid phase $\frac{\partial \varepsilon}{\partial t} + \nabla \cdot \varepsilon \langle \mathbf{u} \rangle_e = 0$
Continuity equation - Particle phase i $\frac{\partial \phi_i}{\partial t} + \nabla \cdot \phi_i \langle \mathbf{v} \rangle_p^i = 0$
Dynamical equation - Fluid phase $\rho_e \left[\frac{\partial}{\partial t} (\varepsilon \langle \mathbf{u} \rangle_e) + \nabla \cdot (\varepsilon \langle \mathbf{u} \rangle_e \langle \mathbf{u} \rangle_e) \right] = \nabla \cdot \langle \mathbf{S} \rangle_e - \sum_{i=1}^{\nu} n_i \langle \mathbf{f} \rangle_p^i + \varepsilon \rho_e \mathbf{g}$
Dynamical equation - Particle phase i $\rho_i \left[\frac{\partial}{\partial t} (\phi_i \langle \mathbf{v} \rangle_p^i) + \nabla \cdot (\phi_i \langle \mathbf{v} \rangle_p^i \langle \mathbf{v} \rangle_p^i) \right] = \nabla \cdot \langle \mathbf{S} \rangle_p^i + n_i \langle \mathbf{f} \rangle_p^i + \sum_{r=1}^{\nu} n_i \langle \mathbf{f} \rangle_p^{ir} + \phi_i \rho_i \mathbf{g}$

5.1. Guiding principles

Some important basic principles which provide valuable guidance in the development of acceptable closure laws should be fulfilled. We mention the principles of frame indifference, well-posedness and fulfillment of the entropy inequality. The first asserts that constitutive equations should be objective and must not depend on any specific physical observer (Astarita & Marrucci, 1974; Truesdell, 1977; Lai et al., 1993). The second requires that the fluid dynamics should be described by a set of equations with unique solution and dependent continuously on their initial and boundary conditions (Drew & Passman, 1998). The third demands that the second law of thermodynamics be met: entropy generation must never be negative (Astarita, 1989; Sandler, 1989). These principles cannot tell us what the closure equations are, but by restricting their allowable form, they tell us what the closures cannot possibly be.

The problem of closure consists in finding constitutive expressions for the effective stress tensors of the fluid and particle phases and for the average interaction forces between the phases. As already pointed out, deriving analytical closures based on purely theoretical arguments is prohibitively difficult; in fact, there is no guarantee that such equations even exist. The goal here is far less ambitious and is finding equations that consent to analyze the systems of interest with the desired accuracy; hence, the problem becomes deriving closures that are as simple as possible, whilst still being able to capture enough physics to describe the fluid dynamics satisfactorily. This is the opposite of the principle of equipresence, which states that in functional dependencies any possible variable should be included, unless it can be proved that a particular dependence cannot occur. Although in theory the idea is correct, its weakness lies in the multitude of parameters present in these equipresence equations and in the impossibility of measuring them experimentally. Thus, especially in the framework

of multiphase flows, this principle is seldom observed.

In what follows, we first present some strategies for modeling the effective fluid and solid stress tensors; we then analyze the mean fluid-particle interaction force, laying emphasis on the buoyancy and drag forces, and the mean particle-particle interaction force.

5.2. Effective stress

Equations (35) and (56), presented in §4., reveal quite clearly the complexity of the Eulerian stress; this arises from the many contributions that make up the effective stress tensors yielded by the averaging process. Closing these quantities is further complicated by the absence of experimental measurements having a direct bearing on them. Several attempts to investigate the viscous behavior of particulate mixtures are found in the literature. In these studies, the suspensions are usually treated as Newtonian pseudofluids, and quantitative measurements of their effective viscosities are provided (Rutgers, 1962; Prudhoe & Whitmore, 1964; Grace, 1970; King et al., 1981; Reiling, 1992; Poletto & Joseph, 1995; Colafigli et al., 2009). This problem has also been the subject of various theoretical (Einstein, 1906, 1911; Vand, 1948; Brinkman, 1952; Frankel & Acrivos, 1967; Batchelor & Green, 1972; Graham, 1981) and semitheoretical (Mooney, 1951; Thomas, 1965; Barnea & Mizrahi, 1973; Metzner, 1985; Gibilaro et al., 2007) investigations. However, as mentioned earlier, all these analyses are concerned with, and provide insight into, the rheology of the mixtures and not of their Eulerian constituent phases. Relating the two is difficult and research is still needed.

Jackson (1997, 1998) and Zhang & Prosperetti (1997) derived complete theoretical closures for dilute monodisperse fluidized suspensions characterized by small Reynolds and Stokes numbers (for more details concerning the degree of dilution and the definition of Stokes number, we refer to the cited articles). Jackson adopted volume averages, whereas Zhang & Prosperetti adopted ensemble averages; the results, however, are similar, and therefore we report only Jackson's. In the limit considered, the effective solid stress tensor vanishes, whereas the effective fluid stress tensor takes the form:

$$\begin{aligned} \langle S_{ab} \rangle_e = & -\delta_{ab} \langle p \rangle_e + \mu_e \left(\frac{\partial \langle u_a \rangle_o}{\partial x_b} + \frac{\partial \langle u_b \rangle_o}{\partial x_a} \right) + \frac{5}{2} \mu_e \phi \left(\frac{\partial \langle u_a \rangle_e}{\partial x_b} + \frac{\partial \langle u_b \rangle_e}{\partial x_a} \right) \\ & - \frac{\partial}{\partial x_b} \left[\frac{3}{4} \mu_e \phi \left(\langle u_a \rangle_e - \langle v_a \rangle_p \right) \right] + 3 \mu_e \phi \left[\frac{1}{2} \left(\frac{\partial \langle u_a \rangle_e}{\partial x_b} - \frac{\partial \langle u_b \rangle_e}{\partial x_a} \right) - \varepsilon_{acb} \langle \omega_c \rangle_p \right] \end{aligned} \quad (58)$$

Here δ_{ab} denotes the Kronecker delta symbol, $\langle p \rangle_e(\mathbf{x}, t)$ the mean fluid pressure, μ_e the fluid shear viscosity, $\langle u_a \rangle_o(\mathbf{x}, t)$ the a th component of the overall average suspension velocity defined as per equation (11), ε_{acb} the Kronecker permutation symbol and $\langle \omega_c \rangle_p(\mathbf{x}, t)$ the c th component of the average angular velocity of the particles. The first term on the right-hand side is the ab th component of the isotropic effective stress tensor; the second and third are viscous contributions, the third presenting the Einstein (1906, 1911) correction for dilute suspensions; the fourth and fifth involve the average linear and rotational slip velocities between the fluid and particle phases.

The closure above has limited validity, but is insightful. It tells us that, even in the simple case of low particle concentrations and small Reynolds and Stokes numbers, the fluid

deviatoric effective stress tensor is not simply related to the fluid average rate of deformation tensor (represented by the third contribution shown in the equation), but depends also on terms related to the average angular and linear velocity fields of the particle phase, since $\langle u_a \rangle_o$ combines both fluid and particle average velocities, and to the angular velocity associated with the fluid average velocity field. Thus, at least in principle, to fully characterize the system dynamics we must also solve an averaged balance equation for the particle angular momentum; for more details about how to derive this equation, we refer to Jackson (1997).

Despite these considerations, researchers often use very simple closures for the effective stress tensors, assuming that each phase is Newtonian and writing:

$$\mathbf{S}_e = - \left[p_e - \left(\kappa_e - \frac{2}{3} \mu_e \right) \text{tr} \mathbf{D}_e \right] \mathbf{I} + 2 \mu_e \mathbf{D}_e ;$$

$$\mathbf{S}_i = - \left[p_i - \left(\kappa_i - \frac{2}{3} \mu_i \right) \text{tr} \mathbf{D}_i \right] \mathbf{I} + 2 \mu_i \mathbf{D}_i \quad (59)$$

where $p_e, p_i, \kappa_e, \kappa_i, \mu_e$ and μ_i are the averaged pressures, dilatational viscosities and shear viscosities of the fluid and solid phases, respectively; furthermore, \mathbf{I} is the identity tensor, whereas \mathbf{D}_e and \mathbf{D}_i are the rate of deformation (or strain) tensors, defined as:

$$\mathbf{D}_e \equiv \frac{1}{2} (\nabla \mathbf{u}_e + \nabla \mathbf{u}_e^T) ; \quad \mathbf{D}_i \equiv \frac{1}{2} (\nabla \mathbf{v}_i + \nabla \mathbf{v}_i^T) \quad (60)$$

From now on, as done in these last expressions, we simplify the notation by leaving out the angular brackets that imply averaging.

The problem of closure then reduces to finding suitable constitutive expressions for the pressure, shear viscosity and dilatational viscosity of each phase. Neglecting the compressibility of the fluid, we do not need to specify its pressure constitutively. Furthermore, often μ_e is assumed to be constant, whilst κ_e is neglected. For the solid phases, elaborate expressions have been derived from the granular kinetic theory (Gidaspow, 1994; Brilliantov & Poschel, 2004), a generalization of the mathematical theory of dense non-uniform gases (Chapman & Cowling, 1970). The main idea is that, since fluidized suspensions resemble in several ways a dense molecular gas, the constitutive equations that govern the two should be derivable, at least in part, from the same theoretical framework. Similarly to a molecular gas, particle pressure and viscosities are functions of a granular temperature, which is governed by a balance equation for a pseudointernal energy related to the particle peculiar velocity. For dense fluidized suspensions, this equation differs from the classical internal energy balance equation because of a sink term $S_c(\mathbf{x}, t)$ representing energy degradation caused by inelastic collisions, a source term $G_d(\mathbf{x}, t)$ representing the generation of particle velocity fluctuations by fluctuating fluid-particle forces and a sink term $S_v(\mathbf{x}, t)$ representing their dampening by viscous resistance to particle motion. Therefore, for the i th solid phase, the balance equation is:

$$\rho_i \left[\frac{\partial}{\partial t} (\phi_i U_i) + \nabla \cdot (\phi_i U_i \mathbf{v}_i) \right] = - \nabla \cdot \mathbf{q}_i + \mathbf{S}_i : \nabla \mathbf{v}_i + G_{d,i} - S_{v,i} - S_{c,i} \quad (61)$$

where $U_i \equiv 3/2 \Theta_i$ is the pseudointernal energy per unit mass, Θ_i being the granular temperature of particle phase i , and \mathbf{q}_i is the pseudothermal heat flux. The closure problem

requires finding expressions for \mathbf{q}_i , $G_{d,i}$, $S_{v,i}$ and $S_{c,i}$. For this we refer to Gidaspow (1994).

Various closures for the solid pressure are available in the literature, all derived from the granular kinetic theory. Here, as an example, we report the expression advanced by Lun et al. (1984), suitably extended to cater for polydisperse suspensions:

$$p_i = \left[1 + 2 \sum_{r=1}^n \left(\frac{s_{ir}}{s_i} \right)^3 (1 + e_{ir}) \phi_r g_{ir} \right] \phi_i \rho_i \Theta_i \quad \text{where} \quad s_{ir} \equiv \frac{s_i + s_r}{2} \quad (62)$$

where s_i and s_r are the particle diameters for phases i and r , respectively, e_{ir} is the coefficient of restitution for collisions between particles of phases i and r , and g_{ir} is a radial distribution function that we obtain by combining the radial distribution functions g_i and g_r of the i th and r th solid phases, respectively. For these functions, there are many possible closures, one being:

$$g_i = \frac{s_i}{2} \sum_{r=1}^n \frac{\phi_r}{s_r} + \left[1 - \left(\frac{\phi}{\phi_{max}} \right)^{1/3} \right]^{-1}; \quad g_{ir} = \frac{s_i g_r + s_r g_i}{s_i + s_r} \quad (63)$$

Here ϕ is the overall solid volume fraction, and ϕ_{max} is the maximum solid compaction, *i.e.*, the maximum value of ϕ . For the dilatational and shear viscosities, Gidaspow (1994) reports:

$$\begin{aligned} \kappa_i &= \frac{4}{3} \phi_i^2 \rho_i s_i g_i (1 + e_i) \left(\frac{\Theta_i}{\pi} \right)^{1/2}; \\ \mu_i &= \frac{10 \rho_i s_i \sqrt{\pi \Theta_i}}{96(1 + e_i) g_i} \left[1 + \frac{4}{5} (1 + e_i) \phi_i g_i \right]^2 + \frac{3}{5} \kappa_i \end{aligned} \quad (64)$$

These expressions are those originally developed for monodisperse suspensions and do not directly account for the presence of the other solid phases.

Two parameters characterize a polydisperse powder: the maximum solid packing ϕ_{max} and the frictional solid packing ϕ_f . The first is a geometrical property of the particles and, as previously mentioned, indicates the maximum volume fraction that the solid can reach; the second marks the transition from the viscous to the plastic flow regime. In the first regime, the particles undergo transient contacts, momentum transfer is translational and collisional, and the granular kinetic theory holds, while in the second, the particles undergo enduring contacts, momentum transfer is mainly frictional and other models, empirical, phenomenological or based on soil mechanics theory, apply.

The plastic granular stress is often modeled with arbitrary functions, which have no theoretical basis but correctly describe qualitatively how dense granular media behave (Syamlal et al., 1993). The prime feature that must be captured is that such materials cannot reach compactations that are unphysically high. A closure for the plastic solid pressure that some modelers use is:

$$p_i^p = \phi_i p^* \quad \text{where} \quad p^* \equiv 10^A (\phi - \phi_f)^B \quad (65)$$

where the coefficients A and B are very high, with typical values of 25 and 10, respectively. This equation is extremely sensitive to the deviation of ϕ from ϕ_f and this may lead to big pressure fluctuations and violent numerical instabilities (Schaeffer, 1987; Schaeffer & Pitman, 1988).

Equation (65) is often employed with radial distribution functions that are bounded and do not diverge positively when ϕ approaches ϕ_{max} . For instance, the model of Syamlal et al. (1993), implemented in the CFD code MFIX, adopts the expression of Lebowitz (1964):

$$g_i = \frac{1}{\varepsilon} \left(1 + \frac{3s_i}{2\varepsilon} \sum_{r=1}^n \frac{\phi_r}{s_r} \right) \quad (66)$$

where, as opposed to equation (63), ϕ_{max} does not feature. Since in equation (63) g_i does diverge when ϕ approaches ϕ_{max} , the viscous solid pressure already prevents the mixture from overpacking, and therefore there is no motive to replace the closure (62) with any arbitrary divergent function, qualitatively sound but theoretically unfounded.

These considerations induce to employ the so-called KTGF-based model, a plastic model partly based on the kinetic theory of granular flows which accounts only for the plastic shear viscosity μ_i^p , neglecting the plastic solid pressure p_i^p and the plastic dilatational viscosity κ_i^p . When ϕ exceeds ϕ_f , it keeps on using the viscous closure for the solid pressure, equation (62), but increases the shear solid viscosity by adding to the viscous contribution, equation (64), a frictional one, whose expression is (Schaeffer, 1987):

$$\mu_i^p = \frac{p_i \sin \vartheta_i}{2\sqrt{I_2(\mathbf{D}_i)}}; \quad I_2(\mathbf{D}_i) \equiv \frac{1}{2} [(\text{tr} \mathbf{D}_i)^2 - \text{tr} \mathbf{D}_i^2] \quad (67)$$

where ϑ_i is the angle of internal friction of the i th granular material, while $I_2(\mathbf{D}_i)$ is the second invariant of the rate of deformation tensor \mathbf{D}_i . In the balance equation of the pseudointernal energy, the higher viscosity generates a higher dissipation of mechanical energy into pseudointernal energy, thus increasing the granular temperature and in turn the viscous solid pressure; therefore, the plastic shear viscosity indirectly affects the solid pressure, despite our setting p_i^p to zero. By slowing down the solid packing, this mechanism increases numerical stability and allows us to use bigger time steps compared with simulations where we neglect the plastic stress entirely (refer to Mazzei et al., 2010).

5.3. Fluid-particle interaction force

There are five main contributors to the fluid-particle interaction force. The first is the buoyancy force, whose definition in the context of multiphase flows is not unique and needs to be discussed. The second acts in the direction of the fluid-particle slip velocity – that is, the fluid velocity relative to an observer moving with the same local mean velocity as the particles. The third is normal to the slip velocity, the fourth is parallel to the relative acceleration between the phases and the fifth is proportional to the local mean acceleration of the fluid. The last four terms are commonly referred to as drag force, lift force, virtual mass force and local fluid acceleration force, respectively. As we shall see, the local fluid acceleration force is not always present, but features only when one definition of buoyancy force is used – in particular, the classical definition presented later on. For this reason, some researchers group the two forces and regard their combination as the buoyancy force; doing so is not incorrect, but we prefer to preserve their individuality. Among these five terms, often the buoyancy and drag forces are dominant; consequently, particular effort has been put into finding reliable equations of closure for such contributions.

5.3.1. Buoyancy force

The definition of buoyancy force, normally considered fairly obvious for single objects in pure fluids, is in our setting quite ambiguous and has been the subject of heated debates for several years. A comprehensive treatment of the topic can be found in Jackson (2000). For fluidized suspensions, at least three alternative definitions have been put forward. The first regards the force as equal to the weight of the fluid displaced by the solid, as suggested for instance by Richardson & Zaki (1954), Wen & Yu (1966), Epstein (1984), Clift et al. (1987), Fan et al. (1987), Kunii & Levenspiel (1989), Jean & Fan (1992) and Mazzei & Lettieri (2007). Accordingly, if we refer the force to the unit volume of suspension, this takes the form:

$$n_i \mathbf{f}_{b,i} \equiv -\phi_i \rho_e \mathbf{g} \quad (68)$$

This definition coincides with the original formulation of the Archimedes's principle (*i.e.*, force equal to the weight of the fluid displaced by the particles); therefore, we refer to it as classical definition. According to equation (68), the buoyancy force is constant, regardless of the local mean pressure and velocity fields that surround the particles. The second definition relates the force to the effective fluid stress tensor, as reported by Jackson (2000). Per unit volume of suspension, it is:

$$n_i \mathbf{f}_{b,i}^\bullet \equiv \phi_i \nabla \cdot \mathbf{S}_e \quad (69)$$

The last definition, endorsed by Foscolo et al. (1983), Gibb (1991), Di Felice (1994) and Gibilaro (2001), to mention just a few, deems the force proportional to the gradient of the average fluid pressure. The constant of proportionality is again the solid volume fraction:

$$n_i \mathbf{f}_{b,i}^\circ \equiv -\phi_i \nabla p_e \quad (70)$$

To better grasp the meaning of these definitions, let us consider homogeneous, monodisperse suspensions. The second and third definitions here clearly coincide, because $\mathbf{D}_e(\mathbf{x}, t)$ vanishes. In uniform mixtures, the solid particles are motionless and equally distributed in space; furthermore, whereas the point fluid velocity is an inhomogeneous field owing to the fluid flow around the particles occurring at the microscopic length scale, the locally averaged velocity field is uniform (in the bulk of the suspension, sufficiently far from the system boundaries). Therefore, both rate of deformation tensors vanish and the stress tensors of both phases are isotropic. Moreover, since no collisions are present between the particles, the solid pressure is zero and, from equation (59), no stress arises in the solid phase. The expression for \mathbf{S}_e in equation (59), along with the definitions (69) and (70), instead gives:

$$\mathbf{S}_e = -p_e \mathbf{I} \quad \Rightarrow \quad n \mathbf{f}_b^\bullet = n \mathbf{f}_b^\circ \equiv n \mathbf{f}_b^* \quad (71)$$

where n is the particle number density. We can derive an expression for $n \mathbf{f}_b^*(\mathbf{x}, t)$ from the mean dynamical equations reported in Table 1; in this case, these reduce to:

$$n \mathbf{f} = -\nabla p_e + \varepsilon \rho_e \mathbf{g}; \quad n \mathbf{f} = -\phi \rho_s \mathbf{g} \quad (72)$$

where ϕ denotes the solid volume fraction, ρ_s the particle density and $n \mathbf{f}$ the fluid-particle interaction force, which according to the equation on the right must counterbalance the

weight of the particles. Subtracting the two equations and using equations (70) and (71) gives:

$$\nabla p_e = (\varepsilon\rho_e + \phi\rho_s)\mathbf{g} ; \quad n\mathbf{f}_b^* = -\phi(\varepsilon\rho_e + \phi\rho_s)\mathbf{g} \quad (73)$$

Therefore, for uniform, monodisperse fluidized mixtures the difference between the first and the other two definitions reduces to the density choice in the force expression: the first requires the fluid density, while the second the suspension bulk density. How we define the buoyancy force is important in the development, or simply the choice, of drag force closures.

In our opinion, the classical definition of buoyancy force, equation (68), should be favored, for it is the only definition that preserves the distinctive feature of this force: being constant and unrelated to the specific characteristics of the flow (Bird et al., 1960). We believe that in our setting relating the buoyancy force to the suspension bulk density, as the other two definitions do, is physically incorrect. In a fluidized suspension of identical particles, or more generally of particles whose diameters do not differ by orders of magnitude, each single particle moves *through the fluid* and displaces its own volume of fluid as it flows, not of suspension. If it were not for the collisions that inevitably take place among the particles, these would not feel the presence of the surrounding others. This is evident in diluted mixtures. In light of these considerations, the definition of buoyancy force that most naturally suggests itself is the one that relates the force to the fluid density. This conclusion, as already pointed out, holds true *as long as all the particles have similar dimensions*. For large objects in suspensions of small particles the situation is quite different: here the motion of such bodies takes place *through the suspension* and the volume displaced comprises both fluid and particles. It makes therefore physical sense to relate the buoyancy force to the bulk density of the mixture. In this regard, we refer to the work of Poletto & Joseph (1995), where the motion of single test spheres in monodisperse suspensions was investigated. The dispersion was modeled as a pseudofluid with effective density and viscosity. The analysis aimed to determine the values of these properties that allowed predicting the settling velocity of test spheres in sedimenting and fluidized suspensions using appropriate modifications of the equation of Francis (1933) for the settling of single spheres in pure Newtonian fluids. The article, in particular, intended to establish the limits of applicability of the idea that the mixture effective density, on which the expression of the buoyancy force is based, should be the mixture bulk density. Experimental evidence showed that this approach is valid as long as the test spheres are rather larger than the suspended particles and provided the solid concentration in the mixture is sufficiently high (more quantitative data can be found in the paper). For dilute suspensions and test particles of similar or smaller size than the suspended particles the model failed. This corroborates the idea that the suspension bulk density should be used only when large objects move through suspensions of much smaller particles.

5.3.2. Local fluid acceleration force

If the classical definition of buoyancy force is adopted, equation (68), the interaction force between fluid and particles must include an additional term known as local fluid acceleration force. This is true not only for solid suspensions but also for single bodies in pure fluids. The origin of this force is clearly presented by Maxey & Riley (1983) in the analysis of

the motion of small rigid spheres in non-uniform flows. Per unit volume of suspension, the force is taken to be:

$$n_i \mathbf{f}_{a,i} \equiv \phi_i \rho_e \frac{D_e \mathbf{u}_e}{Dt} \quad (74)$$

Here the derivative on the right-hand side is a material derivative relative to a Lagrangian observer moving with the locally averaged velocity of the fluid. An analogous material derivative can also be introduced for the particle phases; it is:

$$\frac{D_e \mathbf{u}_e}{Dt} \equiv \frac{\partial \mathbf{u}_e}{\partial t} + \mathbf{u}_e \cdot \nabla \mathbf{u}_e ; \quad \frac{D_i \mathbf{v}_i}{Dt} \equiv \frac{\partial \mathbf{v}_i}{\partial t} + \mathbf{v}_i \cdot \nabla \mathbf{v}_i \quad (75)$$

Often the fluid acceleration is much less than the gravitational acceleration; if so, the local fluid acceleration force is negligible compared with the buoyancy force. Nevertheless, the force plays an important conceptual role; to prove this point, we resort to a thought experiment reported by Jackson (2000). A uniform assembly of particles is at rest in a body of fluid. The fluid is also at rest in a vertical container placed on a horizontal plane. The whole system resides in a uniform gravitational field. At a specified time, the plane that supports the container and the constraints that keep the particles at rest are removed. So, the whole system falls freely with an acceleration equal to the gravitational field. As the mean velocity fields of both phases are uniform and no pressure gradients are present, the fluid and particle effective stress tensors vanish, and the averaged dynamical equations reported in Table 1 reduce to:

$$\varepsilon \rho_e \frac{D_e \mathbf{u}_e}{Dt} = -n \mathbf{f} + \varepsilon \rho_e \mathbf{g} ; \quad \phi \rho_s \frac{D_s \mathbf{v}_s}{Dt} = n \mathbf{f} + \phi \rho_s \mathbf{g} \quad (76)$$

For convenience, we have used the non-conservative formulation of the equations; to obtain them, we must combine the dynamical and continuity equations (Bird et al., 1960). Both material derivatives are equal to the gravitational acceleration; hence, the two equations of motion lead to the same result: the fluid-particle interaction force must vanish. Clearly, this condition can be met only if the contribution owing to the local fluid acceleration force is accounted for. In fact, as the two phases undergo the same motion, no slip velocity and acceleration are present between them; consequently, the drag, the virtual mass and the lift forces are all zero. The buoyancy force, conversely, is constant; accordingly, the overall interaction force can go to zero only if the local fluid acceleration force is considered:

$$n \mathbf{f} = n \mathbf{f}_b + n \mathbf{f}_a = -\phi \rho_e \mathbf{g} + \phi \rho_e \mathbf{g} = \mathbf{0} \quad (77)$$

Since the averaged fluid acceleration is often far less than the local gravitational field, this ideal experiment does not reflect the normal conditions wherein fluidized beds usually operate; as pointed out, the local fluid acceleration force can be often neglected.

5.3.3. Virtual mass and lift forces

If a body immersed in a fluid accelerates, some of the surrounding medium must also accelerate; this results in a force named virtual mass force. For an insightful analysis on this topic, we refer to Birkhoff (1950). If an object moves in a fluid that is in shearing flow, it experiences a force transverse to the direction of relative motion. This force is called lift force.

Exact expressions for these forces have been derived analytically by some authors for single particles of spherical or nearly spherical shape in non-uniform flows (Maxey & Riley, 1983; Nadim & Stone, 1991). They, nevertheless, lack general validity and only apply to specific fluid dynamic conditions (*e.g.*, vanishing or extremely low Reynolds numbers). Researchers have used these equations to develop theoretical closures for monodisperse fluidized suspensions; we mention, for instance, the work of Zhang & Prosperetti (1994, 1997) and of Jackson (1997, 1998). The latter, however, are valid for low particle concentrations and under very restricting assumptions, such as vanishing viscosity (Zhang & Prosperetti, 1994) or very small Stokes and Reynolds numbers (Jackson, 1997, 1998; Zhang & Prosperetti, 1997). Their generalization to other fluid dynamic conditions is not simple and raises conceptual issues (related, for example, to frame indifference and objectivity). For instance, the lift force on an isolated spherical particle takes quite different functional forms in the inviscid and low Reynolds number cases (Saffman, 1965; Auton et al., 1988; Jackson, 2000); finding a general expression for this term appears to be difficult.

An established closure for the virtual mass force is:

$$n_i \mathbf{f}_{v,i} \equiv \phi_i \rho_e C_v(\phi_i) \left(\frac{D_e \mathbf{u}_e}{Dt} - \frac{D_i \mathbf{v}_i}{Dt} \right) \quad (78)$$

Theoretical, and even empirical, expressions for the coefficient $C_v(\phi_i)$ are still to be found. For very dilute mixtures of spherical particles, $C_v(\phi_i)$ is taken to be 1/2, for this is the calculated value for a single sphere in an infinite fluid (Maxey & Riley, 1983; Nadim & Stone, 1991). A similar result was also found analytically by Zhang & Prosperetti (1994), who derived an exact analytical expression of the force for inviscid fluids at low particle concentrations.

The lift force is usually taken to be:

$$n_i \mathbf{f}_{l,i} \equiv \phi_i \rho_e C_l(\phi_i) (\nabla \times \mathbf{u}_e) \times (\mathbf{u}_e - \mathbf{v}_i) \quad (79)$$

Equations (78) and (79) are not frame independent if taken separately, yet their sum fulfills the principle of material objectivity provided that $C_v(\phi_i)$ and $C_l(\phi_i)$ are equal (Drew & Passman, 1998). For this reason, in dilute flows of spherical particles, one normally puts $C_l(\phi_i) = 1/2$.

5.3.4. Drag force

By definition, the drag force is parallel to the fluid-particle slip velocity; so, it is:

$$n_i \mathbf{f}_{d,i} \equiv \beta_i (\mathbf{u}_e - \mathbf{v}_i) \quad (80)$$

where β_i is the drag force coefficient for the i th particle phase. Finding a closure for the drag force amounts to finding a constitutive expression for β_i .

As previously mentioned, the definition of buoyancy force is important inasmuch as it affects the closure for the drag force. To prove this point, let us consider a monodisperse, uniform mixture. If we denote by $n \mathbf{f}_d$ and $n \mathbf{f}_d^\circ$ the drag forces per unit volume of suspension consistent with the buoyancy force definitions (68) and (70), respectively, it is:

$$n \mathbf{f}_d^\circ = \varepsilon n \mathbf{f}_d \quad (81)$$

This is because in uniform fluidized suspensions the local fluid acceleration force (if present) and the virtual mass and lift forces vanish; so, the second of equations (72) gives:

$$n\mathbf{f} = n\mathbf{f}_b + n\mathbf{f}_d = n\mathbf{f}_b^\circ + n\mathbf{f}_d^\circ = -\phi\rho_s\mathbf{g} \quad (82)$$

Then, from equations (68) and (73), it is:

$$n\mathbf{f}_d = -\phi(\rho_s - \rho_e)\mathbf{g}; \quad n\mathbf{f}_d^\circ = -\varepsilon\phi(\rho_s - \rho_e)\mathbf{g} \quad (83)$$

whence equation (81) ensues. The latter relation is required to render drag force closures based on different buoyancy force definitions consistent with one another.

We now report some of the most well-known expressions used for modeling the drag force in fluidized beds. All the closures below are consistent with the classical definition of buoyancy force, or are rendered so by resorting to equation (81).

The Ergun (1952) empirical correlation was developed to assess the unrecoverable pressure drop through packed beds. Extending its range of validity to uniform fluidized suspensions, we get:

$$\beta_i = 150 \frac{\mu_e \phi_i (1 - \varepsilon)}{s_i^2 \varepsilon^2} + 1.75 \frac{\rho_e \|\mathbf{u}_e - \mathbf{v}_i\| \phi_i}{s_i \varepsilon} \quad (84)$$

Other well-known closures are those of Lewis et al. (1949), Wen & Yu (1966) and Kmiec (1982), the first being the default correlation in most CFD codes when the suspension void fraction exceeds 0.80. All three relations can be put in the form:

$$\beta_i = \frac{3}{4} C_D(Re_i) \frac{\rho_e \|\mathbf{u}_e - \mathbf{v}_i\| \phi_i}{s_i} \varepsilon^{-\alpha} \quad (85)$$

where $\alpha = 2.65$ for Lewis et al., $\alpha = 2.70$ for Wen & Yu and $\alpha = 2.78$ for Kmiec. Here the drag coefficient is calculated using the expression of Schiller & Naumann (1935):

$$Re_i \equiv \frac{\rho_e \varepsilon \|\mathbf{u}_e - \mathbf{v}_i\| s_i}{\mu_e};$$

$$C_D(Re_i) = \begin{cases} \frac{24}{Re_i} (1 + 0.15 Re_i^{0.687}) & \text{for } Re_i < 1000 \\ 0.44 & \text{for } Re_i \geq 1000 \end{cases} \quad (86)$$

Di Felice (1994) modified equation (85) considering a functional dependence of the coefficient α on the particle Reynolds number, suggesting:

$$\alpha = 2.7 - 0.65 \exp \left[-\frac{1}{2} (1.5 - \log_{10} Re_i)^2 \right] \quad (87)$$

We finally report the closure recently advanced by Mazzei & Lettieri (2007); this can be put in a form similar to equation (85), where the coefficient α is a function of both particle

Reynolds number and suspension void fraction. In particular, it is:

$$\begin{aligned} \alpha(\varepsilon, Re_i) &\equiv -\frac{\ln \varphi(\varepsilon, Re_i)}{\ln \varepsilon} ; \quad \varphi(\varepsilon, Re_i) \equiv \frac{\bar{C}_D(\bar{Re}_i)}{C_D(Re_i)} \varepsilon^{2(1-n_r)} \\ C_D(Re_i) &= \left(0.63 + 4.8 Re_i^{-1/2}\right)^2 ; \quad \bar{C}_D(\bar{Re}_i) = \left(0.63 + 4.8 \bar{Re}_i^{-1/2}\right)^2 \\ Re_i &\equiv \frac{\rho_e \varepsilon \|\mathbf{u}_e - \mathbf{v}_i\| s_i}{\mu_e} ; \quad \bar{Re}_i(\varepsilon, Re_i) \equiv \frac{Re_i}{\varepsilon^{n_r}} ; \\ n_r(\bar{Re}_i) &= \frac{4.8 + 2.4 \cdot 0.175 \bar{Re}_i^{3/4}}{1 + 0.175 \bar{Re}_i^{3/4}} \end{aligned} \quad (88)$$

Here the drag coefficients C_D and \bar{C}_D are evaluated with the relation of Dallavalle (1948), whilst n_r is the Richardson & Zaki (1954) coefficient evaluated with the equation of Rowe (1987). Notice that, since n_r is a function of \bar{Re}_i and \bar{Re}_i in turn depends on n_r , to calculate these quantities we need to solve a nonlinear system. Nevertheless, since n_r has a very narrow range of variation (being bounded between 2.40 and 4.80), finding the solution requires few iterations.

The accuracy of the closure laws reported above should be checked case by case against experimental evidence. Here, as an alternative, we use as benchmark the correlation of Richardson & Zaki (1954), which allows determining the expansion of non-cohesive fluidized powders for any given value of the superficial fluid velocity. Following the method devised by Mazzei & Lettieri (2007), we report in Figure 2 the voidage ratio (defined below) as a function of the particle Reynolds number parametric in the void fraction for all the closures described above and assuming that the powder is monodisperse; for equation (85) we select the expression of Wen & Yu, choosing α equal to 2.70.

The void ratio $R_\varepsilon(Re, \varepsilon_r)$ is defined as follows. Consider a monodisperse, uniform suspension fluidized by a *liquid* with superficial velocity equal to u . Let ε_r and ε_c be the equilibrium bed void fractions predicted at this velocity by the Richardson & Zaki equation and a generic drag force closure, respectively. If we use the classical definition of buoyancy force, to compute ε_c we need to solve the first of equations (83), which expresses a simple linear momentum balance. Then the void ratio is $R_\varepsilon \equiv \varepsilon_c / \varepsilon_r$. This quantity is a function of ε_r (or equivalently u) and Re . The closer R_ε is to one, the better the match between the Richardson & Zaki equation and the drag force closure examined for the given values of u and Re .

The Ergun equation agrees with the Richardson & Zaki correlation in the viscous and intermediate fluid dynamic regimes; in the first, the agreement improves as the void fraction decreases, whereas in the second an opposite trend is observed. For high particle Reynolds numbers, the equations agree only for fixed or nearly fixed beds. The Wen & Yu closure yields the same results as the Richardson & Zaki equation in both viscous and inertial limits for any void fraction; in the intermediate region, nevertheless, a good agreement is found only at high dilutions. The predictions improve if we employ the expression of Di Felice, whereas the match is perfect if we employ the closure of Mazzei & Lettieri; this is because the latter is based on the Richardson & Zaki correlation. The reader should keep in mind that this correlation is not perfect; here we adopt it as a benchmark because,

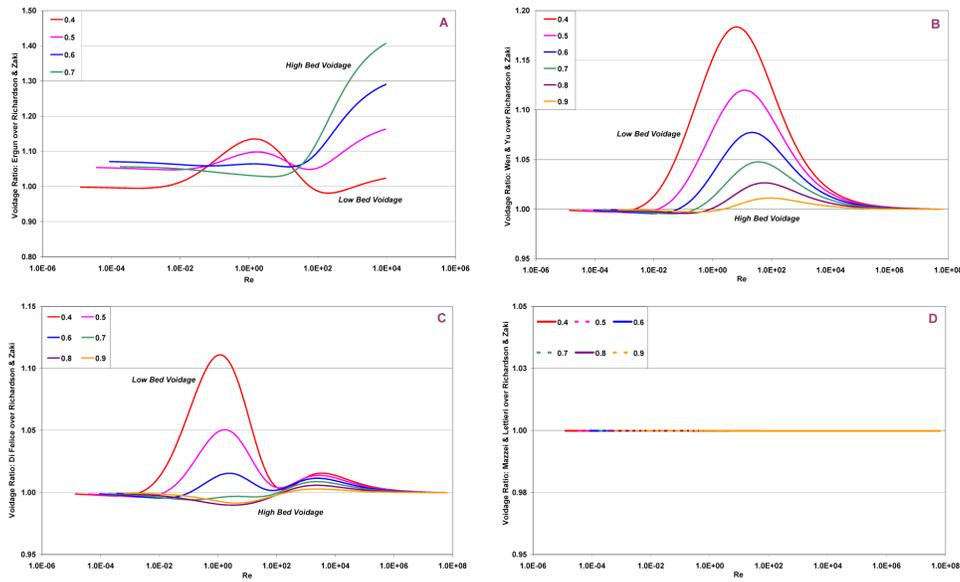


Figure 2. Ratio between the equilibrium void fractions based on the closures of A) Ergun (1952), B) Wen & Yu (1966), C) Di Felice (1994) and D) Mazzei & Lettieri (2007) and on Richardson & Zaki (1954) empirical correlation. The curves are parametric in the Richardson & Zaki void fraction.

in absence of specific experimental data, this equation is the most reliable. However, as already mentioned, the accuracy of the various closures varies from case to case, and in each one the modeler should validate the predictions against experimental findings.

The importance of accounting for the dependence of α on the void fraction and particle Reynolds number as suggested by Mazzei & Lettieri (2007) can be appreciated in Figure 3A, in which α is plotted versus Re , parametric in ε (for further details, we refer the reader to the cited article). As we can observe, albeit in the extremes of purely viscous and purely inertial regimes α does tend to the constant value of 2.80 unaffected by the value of the fluid volume fraction, in the intermediate fluid dynamic region the behavior is radically different. Indeed, far from being constant, α drops significantly reaching values as low as 1.90. Furthermore, the exponent does not depend solely on the particle Reynolds number, since a strong dependence is also found on the suspension void fraction. Hence, the minimum value of α is not uniquely defined (as it was in the correlation advanced by Di Felice) but results to be an increasing function of ε varying roughly between 1.90 and 2.30. These results are also confirmed by the comparison with the experimental data of Happel & Epstein (1954) and Rumpf & Gupte (1971) reported on the diagram.

To complete the analysis, we compare the predictions of equation (88) with those of the other closures examined. Once again, we reason in terms of exponent α . In Figure 3B, we compare the theoretical values of α in the various equations (for that of Ergun, where no exponent appears, we define an effective exponent; refer to Mazzei & Lettieri, 2007) with the corresponding empirical values based on the experimental data of Richardson & Zaki (1954) and Wilhelm & Kwauk (1948), which refer to studies on expanding homogeneous

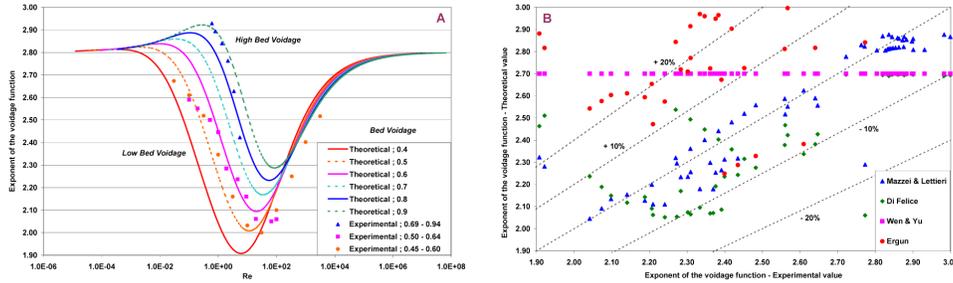


Figure 3. Comparison between theoretical and experimental values of the exponent of the voidage function. The empirical data are of Happel & Epstein (1954) and Rumpf & Gupte (1971). A) Closure of Mazzei & Lettieri (2007). B) Closures of Ergun (1952), Wen & Yu (1966), Di Felice (1994) and Mazzei & Lettieri (2007).

beds. The figure shows that the experimental values of α and those predicted by equation (88) agree well; whereas the error of equation (88) is usually less than $\pm 10\%$, that of the other relations is often greater, especially for equations (84) and (85), where the error sometimes exceeds $\pm 20\%$.

Figure 3 clearly highlights the dependence of α on Re and ε . To assess well the drag force magnitude and predict accurately the bed expansion of uniform, non-cohesive fluidized mixtures, we have to account for this variability, which the other closures entirely or partly neglect.

5.3.5. Other forces

Other contributions to the fluid-particle force could, at least in principle, be considered. A comprehensive overview can be found in Drew & Passman (1998). Here we cite only the Faxen force, the elastic force and a history-dependent term analogous to the Basset force for the motion of isolated particles (Basset, 1888). For the latter, we can reasonably believe that for dense fluidized suspensions the averaging of history-dependent forces should result into a vanishing contribution: averaging would most probably erase any historical effect of the motion of the particles on the fluid in their immediate neighborhood. The Faxen force instead arises from viscous effects in the fluid phase (Brenner, 1964), whereas the elastic force is related to suspension void fraction gradients; in uniform fluidized beds it vanishes, but is important in the study of their stability. Foscolo & Gibilaro (1987), in particular, used this concept to explain the physical origin of stability on fluid dynamical grounds (for a detailed analysis, we refer to Gibilaro, 2001), as opposed to other researchers who ascribed it to the effect of interparticle forces (see for instance Massimilla et al., 1972; Mutsers & Rietema, 1977; Rietema & Piepers, 1990).

5.4. Particle-particle interaction force

In fluidized mixtures of several monodisperse particle classes, each class exchanges linear momentum with all the others; this momentum transfer arises from particle collisions and results into a particle-particle drag force. Soo (1967) was among the first to quantify it,

deriving a theoretical expression for the force acting on a single particle of species i in a cloud of colliding particles of species r . Nakamura & Capes (1976) and Arastoopour et al. (1982) made similar efforts. Many authors have since then put forward other correlations, most of them being variations of earlier works. The force is expressed as the product of a drag coefficient and the slip velocity between the particle classes; thus, it is:

$$n_i \mathbf{f}_{ir} \equiv \zeta_{ir} (\mathbf{v}_r - \mathbf{v}_i) \quad (89)$$

where $n_i \mathbf{f}_{ir}$ is the force exerted by phase r on phase i per unit volume of suspension (see Table 1) and ζ_{ir} is the particle-particle drag coefficient for the two particle classes involved. The closure problem reduces to finding a constitutive expression for ζ_{ir} . Gidaspow et al. (1985) advanced the relation:

$$\zeta_{ir} = C_{ir} (1 + e_{ir}) \frac{\rho_i \rho_r \phi_i \phi_r (s_i + s_r)^2}{\rho_i s_i^3 + \rho_r s_r^3} \|\mathbf{v}_r - \mathbf{v}_i\| \quad (90)$$

where ρ_i , ρ_r , s_i and s_r are the densities and diameters of the particles of classes i and r , respectively, e_{ir} is their coefficient of restitution and C_{ir} is given by:

$$C_{ir} \equiv \frac{3A_{ir} + B_{ir}}{4(A_{ir} - B_{ir})}; \quad A_{ir} \equiv \Phi_{ir}^{1/3}; \quad B_{ir} \equiv (\phi_i + \phi_r)^{1/3} \quad (91)$$

Here the quantity Φ_{ir} is defined to be:

$$\Phi_{ir} \equiv (1 - s_{ir}) \left[\Phi_i + (1 - \Phi_i) \Phi_r \right] (1 - X_{ir}) + \Phi_i \quad \text{for} \quad X_{ir} \geq \frac{\Phi_i}{\Phi_i + (1 - \Phi_i) \Phi_r} \quad (92)$$

Otherwise, it is:

$$\Phi_{ir} \equiv \left[(\Phi_i - \Phi_r) + (1 - s_{ir})(1 - \Phi_i) \Phi_r \right] \left[\Phi_i + (1 - \Phi_i) \Phi_r \right] \frac{X_{ir}}{\Phi_i} + \Phi_r \quad (93)$$

In the relations above, Φ_i and Φ_r are the particle volume fractions at maximum packing for the phases i and r , respectively; moreover, it is:

$$X_{ir} \equiv \frac{\phi_i}{\phi_i + \phi_r}; \quad s_{ir} \equiv \left(\frac{s_r}{s_i} \right)^{1/2} \quad \text{if} \quad s_i \geq s_r \quad \text{and} \quad s_{ir} \equiv \left(\frac{s_r}{s_i} \right)^{-1/2} \quad \text{if} \quad s_r < s_i \quad (94)$$

The closure of Syamlal (1987) instead reads:

$$\zeta_{ir} = \frac{3}{4} (1 + e_{ir}) \left(1 + \frac{\pi}{4} F_{ir} \right) \frac{\rho_i \rho_r \phi_i \phi_r g_{ir} (s_i + s_r)^2}{\rho_i s_i^3 + \rho_r s_r^3} \|\mathbf{v}_r - \mathbf{v}_i\| \quad (95)$$

where F_{ir} denotes a coefficient of friction for phases i and r , whereas g_{ir} the radial distribution function of Lebowitz (1964), given by equation (66). Gera et al. (2004) suggested that the equations above should include an additional term that is necessary to prevent the particle phases from segregating when they are fully packed. Without it, equations (90) and (95) permit packed particles of different size to segregate, a phenomenon that is not observed experimentally. To avoid this, they recommended adding to the coefficient ζ_{ir} the

term ΨP^* , where Ψ must be adjusted for different particle mixtures to match the actual segregation rate, and P^* is given by:

$$P^* = \begin{cases} 10^{25}(\phi - \phi^*)^{10} & \text{for } \phi \geq \phi^* \\ 0 & \text{for } \phi < \phi^* \end{cases} \quad (96)$$

where ϕ^* is a threshold greater than the frictional solid packing ϕ_f which should coincide with the packed bed solid compaction. ΨP^* is added so that when the powder nears maximum packing the particle-particle drag increases sufficiently to make the two solid phases move together as if they were one phase, hindering segregation. Similarly to equation (65), this one as well is extremely sensitive to the deviation of ϕ from ϕ^* and may lead to violent numerical instabilities.

6. Eulerian Modeling of polydisperse Fluidized Suspensions

In the previous sections, we have presented the Eulerian equations of motion for dense fluidized suspensions constituted of ν classes of particles, the i th class being characterized by a density ρ_i and a diameter s_i . Even if powerful, this modeling does not allow the particle diameter to vary. This is a serious restriction that limits the model flexibility: particles can mix and segregate, but variations in their diameters are not allowed for. In reality, conversely, particles can grow, shrink, aggregate, break and nucleate; hence, their size distribution modifies continuously in time and space. Predicting this evolution, which depends on the local conditions wherein the system operates, is essential for a reliable description of the mixture behavior. To this end, we now briefly introduce a different modeling approach, based on the so-called generalized population balance equation, an extension of the Boltzmann equation (Cercignani, 1988).

A fluidized suspension is formed by several particles. To each particle we can associate a spatial position \mathbf{x} , a velocity \mathbf{v} and a size s . The first is usually referred to as external coordinate, whilst the others as internal coordinates. Therefore, we can regard each particle as a material point moving in a seven-dimensional phase space where the position vector is $\mathbf{r} \equiv (\mathbf{x}, \mathbf{v}, s)$. This vector is different from the one previously defined in §3.1., which represented the state of the entire suspension in its associated phase space. As done in §3.1., we can demonstrate that there exists a number density function $f(\mathbf{x}, \mathbf{v}, s, t)$ defined so that $f(\mathbf{x}, \mathbf{v}, s, t)d\mathbf{x}d\mathbf{v}ds$ represents the expected (in a statistical sense) number of particles located in physical space in the volume $d\mathbf{x}$ around \mathbf{x} having velocity in the range $d\mathbf{v}$ around \mathbf{v} and size in the range ds around s . The NDF depends on t because, as the particles collide, break, aggregate, grow and undergo similar phenomena, their distribution in each point \mathbf{x} changes as time progresses.

The generalized population balance equation (GPBE) is the transport equation, written in the phase space of one particle, governing the evolution of the NDF. In the phase space, the particle velocity is $\dot{\mathbf{r}} \equiv (\mathbf{v}, \dot{\mathbf{v}}, \dot{s})$, where \dot{s} is the rate of change of the particle size owing to continuous phenomena such as reaction or wearing, and $\dot{\mathbf{v}}$ is the particle acceleration, that is, the force acting on the particle per unit mass. To derive the GPBE, we consider an arbitrary fixed volume $\Lambda_r \equiv \Lambda_x \cup \Lambda_v \cup \Lambda_s$ in the one-particle phase space. The net numbers

of particles that accumulate in Λ_r and enter it per unit time are:

$$Acc = \int_{\Lambda_r} \frac{\partial f}{\partial t} d\mathbf{r}; \quad In - Out = - \int_{\Lambda_r} \nabla_r \cdot f \dot{\mathbf{r}} d\mathbf{r} \quad (97)$$

where ∇_r is the nabla operator in the multidimensional space. The difference between Acc and $In - Out$ must equate the net number of particles generated per unit time within Λ_r . Here particles generate because of collisions, breakage, aggregation and alike *discontinuous phenomena*. For instance, if two particles located outside Λ_v collide, their velocities change abruptly and after the collision one or even both particles might be located inside Λ_v , having thus entered Λ_r without crossing its boundaries. Using the notation of suggested by Chapman & Cowling (1970), we write:

$$Gen = \int_{\Lambda_r} \frac{\partial f_e}{\partial t} d\mathbf{r} \quad (98)$$

where $\partial f_e / \partial t$ denotes the net number of particles generated per unit volume of multidimensional space and unit time owing to discontinuous (that is, instantaneous) phenomena. If we now equate $Acc - (In - Out)$ to Gen , we find an integral equation over the arbitrary control volume Λ_r . Since this equation must vanish for any Λ_r , its integrand must be zero; therefore, we obtain:

$$\frac{\partial f}{\partial t} + \nabla_r \cdot f \dot{\mathbf{r}} = \frac{\partial f_e}{\partial t} \quad (99)$$

This is the GPBE. We find it convenient to rewrite it in terms of the velocities that the particles have in the physical, velocity and size spaces, respectively. Now, since it is $\nabla_r \cdot f \dot{\mathbf{r}} \equiv \nabla_x \cdot f \mathbf{v} + \nabla_v \cdot f \dot{\mathbf{v}} + \partial f \dot{s} / \partial s$, where ∇_x and ∇_v denote the nabla operators in physical space and velocity space, respectively, substituting into equation (99) yields:

$$\frac{\partial f}{\partial t} + \nabla_x \cdot f \mathbf{v} + \nabla_v \cdot f \dot{\mathbf{v}} + \frac{\partial f \dot{s}}{\partial s} = \frac{\partial f_e}{\partial t} \quad (100)$$

Solving this equation would give the function $f(\mathbf{x}, \mathbf{v}, s, t)$, which describes how the particles are distributed in space and time over velocity and size. Doing so, however, is difficult for two reasons. First, the equation is unclosed, because the generation term $\partial f_e / \partial t$ involves velocity and size correlations between two particles (Marchisio & Fox, 2007). Second, even if we close the equation constitutively, this involves more than three spatial dimensions – because \mathbf{r} is seven-dimensional – and therefore no conventional CFD code would be able to integrate it. However, if we were able to solve the GPBE, we would know everything about the state of single particles; in particular, we would be able to calculate the local mean value $\langle \xi \rangle_s(\mathbf{x}, t)$ of any property $\xi(\mathbf{x}, \mathbf{v}, s, t)$ attached to single particles by writing:

$$\langle \xi \rangle_s(\mathbf{x}, t) \int_{\Omega_v} \int_{\Omega_s} f(\mathbf{x}, \mathbf{v}, s, t) d\mathbf{v} ds \equiv \int_{\Omega_v} \int_{\Omega_s} \xi(\mathbf{x}, \mathbf{v}, s, t) f(\mathbf{x}, \mathbf{v}, s, t) d\mathbf{v} ds \quad (101)$$

This equation generalizes equation (6). Here Ω_v and Ω_s denote the ranges of variation of the coordinates \mathbf{v} and s , respectively, while the integral on the left-hand side represents the

particle number per unit volume, or equivalently the particle number density, at the spatial location \boldsymbol{x} .

Deriving constitutive expressions for $\partial f_e / \partial t$ is extremely complex; we do not address this problem here, but refer to the literature (Chapman & Cowling, 1970; Marchisio & Fox, 2007). There exist various methods that can solve GPBEs numerically; for a comprehensive review we refer to Ramkrishna (2000). A powerful one is the Monte Carlo technique, based on artificial realizations of the system behavior. Initially, the particle population is in a state known statistically; as it evolves, the population undergoes deterministic (continuous) changes in particle state, described by ordinary differential equations, and random (discontinuous) changes in particle state, owing to processes, such as aggregation or breakage, with specified probabilities. To create a realization of the system behavior, we artificially generate random variables satisfying specified probability laws of change. By generating numerous realizations, we can determine the expected behavior of the system by averaging the sample paths. This technique, founded on ensemble averaging, requires a lot of computing power and is impractical for real applications.

Often engineers are only interested in some integral properties of the density function. These properties, called moments, might be important because they control the product quality or because they are simple to measure and monitor. The idea behind the so-called method of moments is to derive transport equations for the moments of interest by integrating out all the internal coordinates from the GPBE (Randolph & Larson, 1971). For any given function $\varphi(\boldsymbol{v}, s)$, we can write:

$$\begin{aligned} \frac{\partial}{\partial t} \int_{\Omega_v} \int_{\Omega_s} \varphi(\boldsymbol{v}, s) f d\boldsymbol{v} ds + \int_{\Omega_v} \int_{\Omega_s} \varphi(\boldsymbol{v}, s) \nabla_r \cdot \boldsymbol{f} \dot{\boldsymbol{r}} d\boldsymbol{v} ds \\ = \int_{\Omega_v} \int_{\Omega_s} \varphi(\boldsymbol{v}, s) \frac{\partial f_e}{\partial t} d\boldsymbol{v} ds \quad (102) \end{aligned}$$

The integral on the left-hand side is the moment of f associated with the function $\varphi(\boldsymbol{v}, s)$, and depends only on \boldsymbol{x} and t . As a consequence, the equation above, which governs the evolution in time and physical space of the moment of f associated with $\varphi(\boldsymbol{v}, s)$, can be dealt with by any normal CFD numerical code. We have therefore overcome the dimensionality issue. The problem with the equation above, nevertheless, is that it is usually unclosed, because for any set of moments that the modeler wishes to track, obtained with a finite set of functions $\varphi_1, \dots, \varphi_n$, the equations involve also moments external to the set (Marchisio & Fox, 2007). For this reason, the method in the present form can be rarely applied.

To overcome the closure problem, we can operate as follows. As just said, the method of moments aims to overcome the dimensionality issue by turning a problem involving one higher-dimensional differential equation in a problem involving an infinite number of four-dimensional differential equations. This system of infinitely many relations is equivalent to the original GPBE. Nevertheless, because we neither want nor can solve an infinite number of equations, the idea behind the method of moments is to satisfy only a finite number of transfer equations. This leaves the NDF largely undetermined, inasmuch as only the infinite set can yield the correct NDF. This means that we can choose – to a certain extent – the NDF arbitrarily and then let the moment equations determine the details that we have not specified. The different moment methods differ in the choice of functions $\varphi(\boldsymbol{v}, s)$ and

in the arbitrary input for the NDF. Their common feature is to choose the latter so that $f(\mathbf{x}, \mathbf{v}, s, t)$ is a given function of \mathbf{v} and s containing $3n$ undetermined parameters, two scalar and one vectorial, depending on \mathbf{x} and t . So, if we take $5n$ scalar moment transport equations, we find $5n$ differential equations for the unknown parameters. One hopes that, for n sufficiently high, the result is accurate and independent of the form chosen for the NDF.

The quadrature methods of moments are examples of this approach; they overcome the closure problem assuming that the NDF has the form:

$$f(\mathbf{x}, \mathbf{v}, s, t) = \sum_{i=1}^n n_i(\mathbf{x}, t) \delta[\mathbf{v} - \mathbf{v}_i(\mathbf{x}, t)] \delta[s - s_i(\mathbf{x}, t)] \quad (103)$$

This is a quadrature formula, where n is the number of nodes, $s_i(\mathbf{x}, t)$ and $\mathbf{v}_i(\mathbf{x}, t)$ are the i th quadrature nodes and $n_i(\mathbf{x}, t)$ is the i th quadrature weight. Equation (103) represents the population through n granular classes, the i th having number density $n_i(\mathbf{x}, t)$ and being made up of particles with size $s_i(\mathbf{x}, t)$ and velocity $\mathbf{v}_i(\mathbf{x}, t)$. The difference between this representation and that used in §4. is that here the size of each particle class is not fixed, but evolves in time and space. Hence, the model can handle continuous and discontinuous processes, such as growth, breakage or aggregation. Here the $3n$ parameters that one must determine through the moment transport equations are $n_i(\mathbf{x}, t)$, $\mathbf{v}_i(\mathbf{x}, t)$ and $s_i(\mathbf{x}, t)$. For more details about this powerful, but complex, approach, we refer the reader to the cited literature.

7. Conclusion

This review has presented strategies for modeling fluidized suspensions. Discrete methods are too expensive computationally and yield too many details of the flow, being therefore unsuitable for industrial applications. For these reasons, we focused on Eulerian modeling, describing the various averaging techniques that turn granular systems in continuous media. The averaged equations directly yield the macroscopic information in which we are interested, but present the disadvantage of being unclosed. We addressed the closure problem, describing the main constitutive equations used by modelers to express the fluid-particle and particle-particle interaction forces and the fluid dynamic stress. This kind of modeling represents the fluidized suspension as a set of classes of particles of fixed size. This is a limitation, since physical and chemical phenomena alter the particle sizes. To conclude the review, we briefly overviewed a more flexible modeling approach, based on the generalized population balance equation, that consents to handle changes in particle size or any other internal coordinate needed to characterize the particle state.

8. Appendix

8.1. Fluid phase local averages of point variable spatial derivatives

We intend to find an expression for the fluid phase local averages of point variable spatial derivatives. To this end, we start by considering the derivative:

$$\frac{\partial}{\partial x_a} \left[\varepsilon(\mathbf{x}, t) \langle \xi \rangle_e(\mathbf{x}, t) \right] \quad (104)$$

Then, using the definition of fluid phase local average given in equations (14) and the derivation chain rule, we rewrite the quantity above as:

$$\begin{aligned} \frac{\partial}{\partial x_a} \int_{\Omega_e} \xi(\mathbf{y}, t) \psi(\|\mathbf{x} - \mathbf{y}\|) d\mathbf{y} &= \int_{\Omega_e} \xi(\mathbf{y}, t) \frac{\partial \psi}{\partial x_a}(\|\mathbf{x} - \mathbf{y}\|) d\mathbf{y} \\ &= - \int_{\Omega_e} \xi(\mathbf{y}, t) \frac{\partial \psi}{\partial y_a}(\|\mathbf{x} - \mathbf{y}\|) d\mathbf{y} \\ &= \int_{\Omega_e} \frac{\partial \xi}{\partial y_a}(\mathbf{y}, t) \psi(\|\mathbf{x} - \mathbf{y}\|) d\mathbf{y} - \int_{\Omega_e} \frac{\partial}{\partial y_a} \left[\xi(\mathbf{y}, t) \psi(\|\mathbf{x} - \mathbf{y}\|) \right] d\mathbf{y} \end{aligned} \quad (105)$$

For the first integral, we can write:

$$\int_{\Omega_e} \frac{\partial \xi}{\partial y_a}(\mathbf{y}, t) \psi(\|\mathbf{x} - \mathbf{y}\|) d\mathbf{y} = \varepsilon(\mathbf{x}, t) \left\langle \frac{\partial \xi}{\partial x_a} \right\rangle_e(\mathbf{x}, t) \quad (106)$$

For the second, the Gauss's theorem allows writing:

$$\begin{aligned} \int_{\Omega_e} \frac{\partial}{\partial y_a} \left[\xi(\mathbf{y}, t) \psi(\|\mathbf{x} - \mathbf{y}\|) \right] d\mathbf{y} \\ = \int_{\partial\Omega_v} \xi(\mathbf{y}, t) m_a(\mathbf{y}, t) \psi(\|\mathbf{x} - \mathbf{y}\|) dS_y - \varphi_x \left[\xi(\mathbf{x}, t), a \right] \end{aligned} \quad (107)$$

where $\partial\Omega_v$ is the surface bounding the domain containing the mixture, $m_a(\mathbf{x}, t)$ is the a th component of the unit vector normal to $\partial\Omega_e$ pointing away from the mixture and φ_x is defined by equation (16). Provided the shortest distance from the generic point $\mathbf{x} \in \partial\Omega_v$ is considerably larger than the weighting function radius, the first term of the right-hand side of the above equation is much smaller than the second. Neglecting it, we obtain equation (15).

8.2. Fluid phase local averages of point variable time derivatives

Similarly, to derive an expression for the fluid phase local averages of point variable time derivatives, we start by considering the derivative:

$$\frac{\partial}{\partial t} \left[\varepsilon(\mathbf{x}, t) \langle \xi \rangle_e(\mathbf{x}, t) \right] \quad (108)$$

Using the definition of fluid phase local average given in equations (14) and applying the Leibnitz's theorem allows writing the quantity above as:

$$\begin{aligned} \frac{\partial}{\partial t} \int_{\Omega_e} \xi(\mathbf{y}, t) \psi(\|\mathbf{x} - \mathbf{y}\|) d\mathbf{y} &= \int_{\Omega_e} \frac{\partial \xi}{\partial t}(\mathbf{y}, t) \psi(\|\mathbf{x} - \mathbf{y}\|) d\mathbf{y} - \varphi_t [\xi(\mathbf{x}, t)] \\ &+ \int_{\partial\Omega_v} \xi(\mathbf{y}, t) \mathbf{u}_v(\mathbf{y}, t) \cdot \mathbf{m}(\mathbf{y}, t) \psi(\|\mathbf{x} - \mathbf{y}\|) dS_y \end{aligned} \quad (109)$$

where $\mathbf{u}_v(\mathbf{x}, t)$ is the velocity of the points $\mathbf{x} \in \partial\Omega_v$ bounding the (time dependent) domain containing the mixture and φ_t is defined by equation (16). Note that in writing φ_t we have used the no-slip boundary condition holding on the surfaces of the particles:

$$\forall \mathbf{x} \in \partial\Omega_i : \mathbf{v}_s(\mathbf{x}, t) = \mathbf{u}(\mathbf{x}, t) \quad (110)$$

which allows replacing the solid velocity $\mathbf{v}_s(\mathbf{x}, t)$ with the fluid velocity $\mathbf{u}(\mathbf{x}, t)$. The integral on $\partial\Omega_v$ can be neglected for the same reasons given in §8.1.. Now, since it is:

$$\int_{\Omega_e} \frac{\partial \xi}{\partial t}(\mathbf{y}, t) \psi(\|\mathbf{x} - \mathbf{y}\|) d\mathbf{y} = \varepsilon(\mathbf{x}, t) \left\langle \frac{\partial \xi}{\partial t} \right\rangle_e(\mathbf{x}, t) \quad (111)$$

equation (109) yields equation (15). Note that if Ω_v is time independent, then $\mathbf{u}_v(\mathbf{x}, t) = \mathbf{0}$ and the integral on $\partial\Omega_v$ in equation (109) rigorously vanishes.

8.3. Particle phase local averages of point variable time derivatives

We intend to derive an expression for the particle phase local averages of point variable time derivatives. In this case, consider the derivative:

$$\frac{\partial}{\partial t} \left[n_i(\mathbf{x}, t) \langle \xi \rangle_p^i(\mathbf{x}, t) \right] \quad (112)$$

Employing the definition of particle phase local average given in equation (20), we can express the partial derivative above as:

$$\begin{aligned} \sum_{\mathcal{F}_i} \frac{\partial}{\partial t} \left[\xi_i(t) \psi(\|\mathbf{x} - \mathbf{x}_i\|) \right] \\ = \sum_{\mathcal{F}_i} \left[\dot{\xi}_i(t) \psi(\|\mathbf{x} - \mathbf{x}_i\|) \right] + \sum_{\mathcal{F}_i} \left[\xi_i(t) \frac{\partial \psi}{\partial t}(\|\mathbf{x} - \mathbf{x}_i\|) \right] \end{aligned} \quad (113)$$

From the definition of particle phase local average, it is:

$$\sum_{\mathcal{F}_i} \left[\dot{\xi}_i(t) \psi(\|\mathbf{x} - \mathbf{x}_i\|) \right] = n_i(\mathbf{x}, t) \langle \dot{\xi} \rangle_p^i(\mathbf{x}, t) \quad (114)$$

Applying the derivation chain rule yields:

$$\begin{aligned} \sum_{\mathcal{F}_i} \left[\xi_i(t) \frac{\partial \psi}{\partial t} (\|\mathbf{x} - \mathbf{x}_i\|) \right] &= - \sum_{\mathcal{F}_i} \left[\xi_i(t) v_{i,a}(t) \frac{\partial \psi}{\partial x_a} (\|\mathbf{x} - \mathbf{x}_i\|) \right] \\ &= - \frac{\partial}{\partial x_a} \sum_{\mathcal{F}_i} \left[\xi_i(t) v_{i,a}(t) \psi (\|\mathbf{x} - \mathbf{x}_i\|) \right] = - \frac{\partial}{\partial x_a} \sum_{\mathcal{F}_i} \left[n_i(\mathbf{x}, t) \langle \xi v_a \rangle_p^i(\mathbf{x}, t) \right] \quad (115) \end{aligned}$$

having used again the partial derivatives commutative property and the definition of particle phase average. Replacing these last two results in equation (113) yields equation (21).

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