A FLUID MECHANICAL DESCRIPTION OF FLUIDIZED BEDS

Equations of Motion

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Starting from the equations of motion of a single solid particle and the Navier-Stokes equations for fluid motion, a set of equations of motion is developed for a system of fluidized particles, and the result is compared with previously proposed equations.

IN PRINCIPLE the motion of a system of particles suspended in a fluid is completely determined by the Navier-Stokes equations, to be satisfied at each point of the fluid, and the Newtonian equations of motion, to be satisfied by each particle. However, when the system of interest comprises a large number of closely spaced particles, as in a fluidized bed, the problem is far too complicated to permit direct solution when stated in these terms, and for practical purposes it is necessary to seek some method of simplifying it so that it can be described by a relatively small number of partial differential equations. At present the literature contains several attempts (Hinze, 1962; Jackson, 1963; Murray, 1965; Pigford and Baron, 1965; Soo, 1963; Van Deemter and van der Laan, 1961) to simplify the problem in this way, all of which replace the point mechanical and fluid mechanical variables, which vary rapidly on a scale comparable with the particle spacing, by smoothed variables obtained by averaging over regions large compared with the particle spacing but small compared with the complete system. The resulting equations, therefore, describe the motion of the fluid and particles as though they were interpenetrating continua.

A complete solution of the basic mechanical and fluid mechanical equations would determine quantities such as the fluid-particle interaction forces and the resistance of the assembly to shear, but in the necessarily more superficial view provided by the continuum models quantities such as these appear as formal terms in the equations, and the form of these terms must be determined empirically. Thus there is ample scope for differences of opinion about the final form of the equations describing the continuum model, and indeed no two sets of equations so far proposed are in complete agreement with each other.

If these differences arose solely from different guesses at the forms of the undetermined terms, they might be expected eventually to be resolved as further experimental evidence becomes available. However, the differences run deeper than this and result from radically different forms of the basic momentum balance. It therefore seems desirable to replace the mainly intuitive considerations from which existing continuum equations were constructed by something rather more formal, which will correctly translate the mechanical basis of the detailed point equations of motion into the continuum model and, at the same time, clearly isolate those terms whose form remains to be determined empirically.

There is certainly more than one way of approaching this problem. Murray (1966), for instance, has applied Boltzmann's equation, treating the fluid molecules and the solid particles on essentially the same basis despite the disparity in their sizes. Here we adopt a rather different approach, and use a formal mathematical definition of local mean variables to translate the point Navier-Stokes equations for the fluid and the Newtonian equations of motion for the particles directly into continuum equations representing momentum balances for the fluid and solid phases. This translation throws up a number of terms whose forms are undetermined. However, the choice of terms is essentially empirical, and there is considerable scope for differing opinions on this aspect of the work until a great deal more experimental information becomes available.

Local Averages

The basis of the present work is the replacement of point mechanical variables such as the fluid velocity, the fluid pressure, or the velocity of solid matter at a specified point within a particle, by local mean variables obtained by averaging the point variables over regions which contain many particles but are still small compared with the scale of "macroscopic" variations from point to point in the system. The idea of local mean variables is, of course, familiar in other fields of physical theory. In gas kinetic theory, for example, one of the most elementary concepts is the number of particles per unit volume in the neighborhood of a specified point, and this is obtained by counting the particles in a region which contains many particles, but is still small in a macroscopic sense. Clearly the concept of local mean variables as just outlined is not very precise, but for many purposes what is meant is physically obvious, and a more careful formulation is scarcely necessary. In our case, however, we propose to make extensive use of local averaging as a mathematical manipulation, so a rather more formal definition will be needed.

Let $g(r)$ be a function, defined for $r > 0$, with the following properties:

$g(r) > 0$ for all r, and g decreases monotonically with increasing r.

$g(r)$ possesses derivatives $g^{(n)}(r)$ of all orders for each value of r.

$\int_{V_\infty} g^{(n)}(r)dV$ exists for all values of n, where r denotes distance from a point in three-dimensional space and the suffix V_∞ on the integral indicates that it is taken over the whole of this space. Thus

$$\int_{V_\infty} g(r)dV = 4\pi \int_0^\infty g(r)r^2 dr$$

$g(r)$ is normalized so that

$$\int_{V_\infty} g(r)dV = 1$$

Then $g(r)$ may be used as a weighting function in forming local averages of system point variables, such as the velocities of fluid or solid matter. Although the point variables themselves vary rapidly in both space and time, because of the complicated paths of individual particles and the distortion of fluid streamlines to pass around and between the particles, local averaging through a region containing many particles would be expected to smooth out these fluctuations, so that the local mean variables appearing in the continuum equations are comparatively well behaved. Indeed, the properties of the weighting function specified above ensure that the ordinary operations of vector calculus can be performed on the local mean variables, and also that certain other mathematical manipulations, such as differentiating under an integral sign, can validly be used.

We speak of the radius, r_o, of the weighting function, defined by

$$4\pi \int_0^{r_o} g(r)r^2dr = 4\pi \int_{r_o}^{\infty} g(r)r^2dr = {}^1/_2 \tag{1}$$

and use g to define local averages in the following way.

Local Mean Voidage and Particle Density. The local mean voidage, ϵ, at a point $\mathbf{x}$ and time t will be defined by

$$\epsilon(\mathbf{x},t) = \int_{V_{f\infty}(t)} g(\mathbf{x} - \mathbf{y})\, dV_y \tag{2}$$

where the suffix $V_{f\infty}(t)$ on the integral indicates that it is taken over all points $\mathbf{y}$ occupied by fluid at time t, and dV_y indicates an element of volume in the neighborhood of point $\mathbf{y}$. The notation $g(\mathbf{x} - \mathbf{y})$ has also been used to indicate what should, more correctly, be written $g(|\mathbf{x} - \mathbf{y}|)$, and this practice will be followed throughout in the interest of simplicity.

If ν is the volume of one particle, all particles being assumed identical, the local mean particle density or number of particles per unit volume, $n(\mathbf{x},t)$, is defined by

$$\nu n(\mathbf{x},t) = 1 - \epsilon(\mathbf{x},t) = 1 - \int_{V_{f\infty}(t)} g(\mathbf{x} - \mathbf{y})\, dV_y = \int_{V_\infty} g(\mathbf{x} - \mathbf{y})dV_y - \int_{V_{f\infty}(t)} g(\mathbf{x} - \mathbf{y})dV_y$$

But denoting by $V_{s\infty}(t)$ the set of all points occupied by solid matter at time t—i.e., all points lying in particles,

$$V_{s\infty}(t) = V_\infty - V_{f\infty}(t)$$

so

$$\nu n(\mathbf{x},t) = \int_{V_{s\infty}(t)} g(\mathbf{x} - \mathbf{y})dV_y \tag{3}$$

Local Mean Values of Point Properties of Fluid and Solid Phases. Let $a'(\mathbf{x},t)$ denote any point property of the fluid phase—for example, pressure p', one of the three components u_i' of the velocity, or one of the nine components E_{ij}' of the stress tensor. In the same way let $b'(\mathbf{x},t)$ denote any point property of the solid phase. Then the local mean values a and b of a' and b' are defined by

$$\epsilon(\mathbf{x},t)a(\mathbf{x},t) = \int_{V_{f\infty}(t)} a'(\mathbf{y},t)g(\mathbf{x} - \mathbf{y})dV_y \tag{4}$$

and

$$[1 - \epsilon(\mathbf{x},t)]b(\mathbf{x},t) = \nu n(\mathbf{x},t)b(\mathbf{x},t) = \int_{V_{s\infty}(t)} b'(\mathbf{y},t)g(\mathbf{x} - \mathbf{y})dV_y \tag{5}$$

respectively.

In general, these definitions do not uniquely determine local mean variables, even for a given configuration of the system, since they depend on the form chosen for the weighting function, g, and, in particular, on its radius, since this determines the size of the region over which averaging effectively takes place. However, if the spatial variations in point properties can be regarded as the sum of two contributions, one on a scale comparable with the particle spacing and the other on a very much larger scale comparable with the dimensions of the complete system, the local mean values will be insensitive to the radius or detailed form of the weighting function, provided the radius is large compared with the particle spacing and small compared with the scale of "macroscopic" variations from point to point in the system. It is only in this case that local mean variables would be expected to have an unambiguous physical significance, and hence to be useful in constructing equations of motion.

A limitation of this type on the range of validity of a continuum description of particulate motion is, of course, familiar in the fluid mechanical description of the motion of a gas composed of molecules, which breaks down when the mean free path becomes comparable with the dimensions of the containing apparatus. In the same way, the continuum description would be expected to be appropriate for a dense assembly of small particles, such as a fluidized bed, and we shall therefore assume, from now on, that fluctuations in point variables can be analyzed into contributions of two very different scales, as described above, and the radius of the weighting function is intermediate between these.

Two simple but important mathematical consequences follow from restricting attention to systems with this property. First, g varies little over the interior of a single particle, so we can write

$$\int_{v_p} b'(\mathbf{y},t)g(\mathbf{x} - \mathbf{y})\, dV_y \approx g(\mathbf{x} - \mathbf{x}_p) \int_{v_p} b'(\mathbf{y},t)dV_y = \nu b_p(t)g(\mathbf{x} - \mathbf{x}_p) \tag{6}$$

where the suffix v_p indicates that the integral is taken over the interior of particle p at time t, $\mathbf{x}_p$ is the position of the center of mass of particle p at time t, and $b_p(t)$ is the average value of b', taken over the interior of particle p. This equation will be used to develop alternative expressions for local mean variables relating to the solid phase.

Secondly, the spatial variation of local mean variables is slow compared with that of g, so their values change little over distances comparable with the radius of the weighting function, and we can write

$$\int_{V_{f\infty}(t)} a(\mathbf{y},t)g(\mathbf{x} - \mathbf{y})dV_y \approx a(\mathbf{x},t) \int_{V_{f\infty}(t)} g(\mathbf{x} - \mathbf{y})dV_y = \epsilon(\mathbf{x},t)a(\mathbf{x},t) \tag{7}$$

while similarly

$$\int_{V_{s\infty}(t)} b(\mathbf{y},t)g(\mathbf{x} - \mathbf{y})dV_y \approx [1 - \epsilon(\mathbf{x},t)]b(\mathbf{x},t) \tag{8}$$

When Equations 7 and 8 are compared with 4 and 5, they show that the local mean value of a local mean variable is approximately equal to itself. However, this desirable property is a feature of the local averaging procedure only when point variations can be decomposed into two contributions, of scales much smaller and much larger than the radius of the weighting function. In future equality signs will frequently be used for approximate equalities of the above type, as well as mathematically exact equations.

The point value of a variable may be written as the sum of its local mean value and a fluctuation about this value:

$$\left.\begin{aligned} a'(\mathbf{x},t) &= a(\mathbf{x},t) + a''(\mathbf{x},t) \\ b'(\mathbf{x},t) &= b(\mathbf{x},t) + b''(\mathbf{x},t) \end{aligned}\right\} \tag{9}$$

Then, although a'' and b'' are not necessarily small, Equations 7 and 8 show that their local mean values are very much smaller than a and b, and can usually be neglected in consequence.

As remarked above, Equation 6 permits the local mean values of properties of the solid phase to be written in an alternative form, since

$$\begin{aligned} \nu n(\mathbf{x},t)b(\mathbf{x},t) &= \int_{V_{s\infty}(t)} b'(\mathbf{y},t)g(\mathbf{x}-\mathbf{y})dV_y \\ &= \sum_{p\infty} \int_{v_p(t)} b'(\mathbf{y},t)g(\mathbf{x}-\mathbf{y})dV_y \\ &\approx \sum_{p\infty} \nu b_p(t)g(\mathbf{x}-\mathbf{x}_p) \end{aligned}$$

Thus

$$n(\mathbf{x},t)b(\mathbf{x},t) = \sum_{p\infty} b_p(t)g(\mathbf{x}-\mathbf{x}_p) \tag{10}$$

where $\sum_{p\infty}$ denotes summation over all the particles of the system. In particular, putting $b' \equiv 1$,

$$n(\mathbf{x},t) = \sum_{p\infty} g(\mathbf{x}-\mathbf{x}_p) \tag{11}$$

If $b_p''(t)$ is used to denote the average value of $b''(\mathbf{y},t)$ taken over the interior of particle p, in the same way as b_p denotes the corresponding average value of $b'(\mathbf{y},t)$, it is also easy to show that

$$b_p(t) = b(\mathbf{x}_p,t) + b_p''(t) \tag{12}$$

and

$$\sum_{p\infty} b_p''(t)\, g(\mathbf{x}-\mathbf{x}_p) \approx 0 \tag{13}$$

Equation 13 being interpreted as meaning that the left-hand side is very small compared with $b(\mathbf{x}_p,t)$.

Finally, local mean variables are smooth functions of position defined at all points of space, even though the corresponding point variables are defined only at points instantaneously occupied by one or other of the phases. Thus local mean variables may be manipulated and differentiated as scalar, vector, or tensor fields defined over the whole of the region occupied by the system.

Derivatives of Local Mean Variables

In order to construct equations of motion it is necessary to know the relation between space and time derivatives of local mean variables and the corresponding derivatives of their associated point variables.

Let a' be a point property of the fluid and a the corresponding local mean variable, as before. Then from the definition (4)

$$\frac{\partial}{\partial x_k}[\epsilon(\mathbf{x},t)a(\mathbf{x},t)] = \int_{V_{f\infty}(t)} g(\mathbf{x}-\mathbf{y})\frac{\partial a'(\mathbf{y},t)}{\partial y_k}\,dV_y - \int_{S_f(t)} n_k a'(\mathbf{y},t)g(\mathbf{x}-\mathbf{y})\,dS_y \tag{14}$$

where n_k denotes the outward normal to the fluid at points on its boundary, and we have made use of the divergence theorem and the fact that

$$\frac{\partial}{\partial x_k} g(\mathbf{x}-\mathbf{y}) = -\frac{\partial}{\partial y_k} g(\mathbf{x}-\mathbf{y}) \tag{15}$$

(The suffix notation with the usual summation convention for repeated suffixes is used throughout for vectors and tensors.)

$S_f(t)$ indicates the surface bounding the fluid phase at time t, and this consists of a number of disjoint parts—namely, the surface $S_{f\infty}$ bounding the whole system, and the surfaces $s_p(t)$ of the separate solid particles at time t—so the second term on the right-hand side of Equation 14 may be written

$$\int_{S_f(t)} n_k a'(\mathbf{y},t)g(\mathbf{x}-\mathbf{y})dS_y = \int_{S_{f\infty}} n_k a'(\mathbf{y},t)g(\mathbf{x}-\mathbf{y})dS_y - \sum_{p\infty}\int_{s_p(t)} n_k a'(\mathbf{y},t)g(\mathbf{x}-\mathbf{y})dS_y \tag{16}$$

In the second term on the right-hand side of this the direction of n_k has been reversed, so that here it refers to the unit normal at the surface of a particle, drawn outward from the particle. Provided the shortest distance from point $\mathbf{x}$ to the surface $S_{f\infty}$ is considerably larger than r_o, the first integral on the right-hand side of Equation 16 is negligibly small compared with the other contributions, and neglecting it Equations 14 and 16 combine to give

$$\int_{V_{f\infty}(t)} g(\mathbf{x}-\mathbf{y})\frac{\partial a'(\mathbf{y},t)}{\partial y_k}\,dV_y = \frac{\partial}{\partial x_k}[\epsilon(\mathbf{x},t)a(\mathbf{x},t)] - \sum_{p\infty}\int_{s_p(t)} n_k a'(\mathbf{y},t)g(\mathbf{x}-\mathbf{y})dS_y \tag{17}$$

which is the desired equation relating space derivatives of point and local mean variables.

A similar relation between time derivatives of point and local mean variables can also be obtained. The argument needs a little more care, since the region of integration $V_{f\infty}(t)$ is itself time-dependent because of the particle motion, but the final result is

$$\int_{V_{f\infty}(t)} g(\mathbf{x}-\mathbf{y})\frac{\partial a'(\mathbf{y},t)}{\partial t}\,dV_y = \frac{\partial}{\partial t}[\epsilon(\mathbf{x},t)a(\mathbf{x},t)] + \sum_{p\infty}\int_{s_p(t)} a'(\mathbf{y},t)n_k v_k'(\mathbf{y},t)g(\mathbf{x}-\mathbf{y})dS_y \tag{18}$$

where $v_k'(y,t)$ denotes the velocity of solid matter at a point $\mathbf{y}$ lying in particle p at time t.

Equations analogous to 17 and 18 can also be obtained relating derivatives of point and local mean values of variables associated with the solid phase. These are

$$\sum_{p\infty}\int_{v_p(t)} g(\mathbf{x}-\mathbf{y})\frac{\partial b'(\mathbf{y},t)}{\partial y_k}\,dV_y = \frac{\partial}{\partial x_k}[(1-\epsilon(\mathbf{x},t))b(\mathbf{x},t)] + \sum_{p\infty}\int_{s_p(t)} n_k b'(\mathbf{y},t)g(\mathbf{x}-\mathbf{y})dS_y \tag{19}$$

and

$$\sum_{p\infty}\int_{v_p(t)} g(\mathbf{x}-\mathbf{y})\frac{\partial b'(\mathbf{y},t)}{\partial t}\,dV_y = \frac{\partial}{\partial t}[(1-\epsilon(\mathbf{x},t))b(\mathbf{x},t)] - \sum_{p\infty}\int_{s_p(t)} b'(\mathbf{y},t)n_k v_k'(\mathbf{y},t)g(\mathbf{x}-\mathbf{y})dS_y \tag{20}$$

Equations of Continuity

If the fluid phase is incompressible, the point velocity satisfies the continuity equation

$$\frac{\partial u_k'}{\partial y_k} = 0$$

at all points currently occupied by fluid. Multiplying by $g(\mathbf{x} - \mathbf{y})$ and integrating over all such points

$$\int_{V_{f\infty}} g(\mathbf{x} - \mathbf{y}) \frac{\partial u_k'(\mathbf{y})}{\partial y_k} dV_y = 0$$

then using Equation 17 this may be written

$$\frac{\partial}{\partial x_k} [\epsilon(\mathbf{x}) u_k(\mathbf{x})] = \sum_{p\infty} \int_{s_p} n_k u_k'(\mathbf{y}) g(\mathbf{x} - \mathbf{y}) dS_y \qquad (21)$$

where u_k is the local mean fluid velocity. [In future independent variables, on which velocity fields, etc., depend, will not always be indicated explicitly. For instance, $V_{f\infty}$ is written rather than $V_{f\infty}(t)$, and $u_k'(\mathbf{y})$ rather than $u_k'(\mathbf{y},t)$ when there is no possible ambiguity.]

But setting $a' \equiv 1$ in Equation 18 (from which it follows that $a \equiv 1$ also), we obtain

$$\frac{\partial}{\partial t} \epsilon(\mathbf{x}) = - \sum_{p\infty} \int_{s_p} n_k v_k'(\mathbf{y}) g(\mathbf{x} - \mathbf{y}) dS_y \qquad (22)$$

At all points lying on the surface of a particle, however, the velocities of fluid and solid matter are equal, so $u_k' = v_k'$, and the right-hand sides of Equations 21 and 22 are equal in magnitude and opposite in sign. Thus, by addition

$$\frac{\partial \epsilon}{\partial t} + \frac{\partial}{\partial x_k} (\epsilon u_k) = 0 \qquad (23)$$

which is the desired equation of continuity for the fluid, expressed in terms of local mean variables.

The corresponding equation of continuity for the particles can be derived in a completely analogous way from Equations 19 and 20. If v_k represents the local mean particle velocity, this is

$$\frac{\partial}{\partial t} (1 - \epsilon) + \frac{\partial}{\partial x_k} [(1 - \epsilon) v_k] = 0 \qquad (24)$$

which may also be written in the alternative form

$$\frac{\partial n}{\partial t} + \frac{\partial}{\partial x_k} (n v_k) = 0 \qquad (24')$$

Equations 23 and 24 are, of course, just what would be suggested by intuitive ideas of local average variables.

Equations of Motion for the Fluid

We assume that the fluid is incompressible and must everywhere satisfy the point Navier-Stokes equations

$$\rho_f \left[\frac{\partial u_i'}{\partial t} + \frac{\partial}{\partial y_k} (u_i' u_k') \right] = \frac{\partial E_{ik}'}{\partial y_k} + \rho_f g_i \qquad (25)$$

where E_{ik}' is the point value of the fluid stress tensor and g_i is a vector pointing vertically downward and equal in magnitude to the gravitational force per unit mass.

Now local averages of both sides of Equation 25 are formed, multiplying through by $g(\mathbf{x} - \mathbf{y})$ and integrating with respect to $\mathbf{y}$ over the whole region $V_{f\infty}(t)$ currently occupied by fluid

$$\rho_f \int_{V_{f\infty}} g(\mathbf{x} - \mathbf{y}) \left[\frac{\partial u_i'}{\partial t} + \frac{\partial}{\partial y_k} (u_i' u_k') \right] dV_y =$$

$$\int_{V_{f\infty}} g(\mathbf{x} - \mathbf{y}) \frac{\partial E_{ik}'}{\partial y_k} dV_y + \rho_f g_i \int_{V_{f\infty}} g(\mathbf{x} - \mathbf{y}) dV_y \qquad (26)$$

it being understood that all expressions in the integrands are evaluated at the current point of integration, $\mathbf{y}$.

First we express the left-hand side of this equation in terms of local mean variables, using Equation 18 with $a' \equiv u_i'$, Equation 17 with $a' \equiv u_i' u_k'$, and taking into account the fact that $u_k' = v_k'$ at all points on the surface of a particle. This gives immediately

$$\rho_f \int_{V_{f\infty}} g(\mathbf{x} - \mathbf{y}) \left[\frac{\partial u_i'}{\partial t} + \frac{\partial}{\partial y_k} (u_i' u_k') \right] dV_y =$$

$$\rho_f \frac{\partial}{\partial t} \left[\epsilon(\mathbf{x}) u_i(\mathbf{x}) \right] + \rho_f \frac{\partial}{\partial x_k} \int_{V_{f\infty}} u_i'(\mathbf{y})\, u_k'(\mathbf{y}) g(\mathbf{x} - \mathbf{y}) dV_y \qquad (27)$$

In the integral on the right-hand side of Equation 27 write $u_i' = u_i + u_i''$, giving

$$\int_{V_{f\infty}} u_i'(\mathbf{y}) u_k'(\mathbf{y}) g(\mathbf{x} - \mathbf{y}) dV_y =$$

$$\int_{V_{f\infty}} u_i u_k(\mathbf{y}) g(\mathbf{x} - \mathbf{y}) dV_y + \int_{V_{f\infty}} (u_i'' u_k + u_i u_k'')\ (\mathbf{y}) \times$$

$$g(\mathbf{x} - \mathbf{y}) dV_y + \int_{V_{f\infty}} u_i'' u_k''(\mathbf{y}) g(\mathbf{x} - \mathbf{y}) dV_y \qquad (28)$$

Taking $a \equiv u_i u_k$ in Equation 7, the first term on the right-hand side of this is seen to reduce to

$$\epsilon(\mathbf{x}) u_i(\mathbf{x}) u_k(\mathbf{x})$$

Again, u_k and u_i vary little over the radius of g, so $u_k(\mathbf{x})$ and $u_i(\mathbf{x})$, respectively, can be taken outside the integrals in the second term on the right-hand side. The integrals then reduce to the local mean values of fluctuations u_i'' and u_k'', which are negligibly small. Thus the second term on the right-hand side of Equation 28 can be neglected. Using these results in the second term on the right-hand side of Equation 27, this equation may be written

$$\rho_f \int_{V_{f\infty}} g(\mathbf{x} - \mathbf{y}) \left[\frac{\partial u_i'}{\partial t} + \frac{\partial}{\partial y_k} (u_i' u_k') \right] dV_y =$$

$$\rho_f \left[\frac{\partial}{\partial t} \{\epsilon(\mathbf{x}) u_i(\mathbf{x})\} + \frac{\partial}{\partial x_k} \{\epsilon(\mathbf{x}) u_i(\mathbf{x}) u_k(\mathbf{x})\} \right] + \frac{\partial R_{ik}}{\partial x_k} \qquad (29)$$

where

$$R_{ik}(\mathbf{x}) = \rho_f \int_{V_{f\infty}} u_i''(\mathbf{y}) u_k''(\mathbf{y}) g(\mathbf{x} - \mathbf{y})\, dV_y \qquad (30)$$

Turning now to the right-hand side of Equation 26, Equation 2 shows immediately that the last term is simply $\epsilon(\mathbf{x}) \rho_f g_i$. The first term may be evaluated by setting $a' \equiv E_{ik}'$ in Equation 17, giving

$$\int_{V_{f\infty}} g(\mathbf{x} - \mathbf{y}) \frac{\partial E_{ik}'}{\partial y_k} dV_y = \frac{\partial}{\partial x_k} [\epsilon(\mathbf{x}) E_{ik}(\mathbf{x})] -$$

$$\sum_{p\infty} \int_{s_p} n_k\, E_{ik}'(\mathbf{y})\, g(\mathbf{x} - \mathbf{y}) dS_y = \frac{\partial}{\partial x_k} [\epsilon(\mathbf{x}) E_{ik}(\mathbf{x})] -$$

$$\sum_{p\infty} \int_{s_p} n_k E_{ik}(\mathbf{y}) g(\mathbf{x} - \mathbf{y}) dS_y - \sum_{p\infty} \int_{s_p} n_k E_{ik}''(\mathbf{y}) g(\mathbf{x} - \mathbf{y}) dS_y \qquad (31)$$

Since E_{ik} is a function of position defined at all points of the system, whether occupied by fluid or solid matter, the second term on the right-hand side of this may be transformed by Gauss's theorem, giving

$$\sum_{p\infty} \int_{s_p} n_k E_{ik}(\mathbf{y}) g(\mathbf{x} - \mathbf{y}) dS_y = \sum_{p\infty} \int_{v_p} \frac{\partial}{\partial y_k} \times [E_{ik}(\mathbf{y}) g(\mathbf{x} - \mathbf{y})] dV_y = \int_{V_{s\infty}} \frac{\partial}{\partial y_k} [E_{ik}(\mathbf{y}) g(\mathbf{x} - \mathbf{y})] dV_y = \int_{V_{s\infty}} g(\mathbf{x} - \mathbf{y}) \frac{\partial E_{ik}}{\partial y_k} dV_y - \frac{\partial}{\partial x_k} \int_{V_{s\infty}} E_{ik}(\mathbf{y}) g(\mathbf{x} - \mathbf{y}) dV_y \tag{32}$$

where we have used the fact that

$$\frac{\partial}{\partial y_k} g(\mathbf{x} - \mathbf{y}) = - \frac{\partial}{\partial x_k} g(\mathbf{x} - \mathbf{y})$$

However, since both E_{ik} and $\partial E_{ik}/\partial y_k$ vary little over distances comparable with the radius of g, they may be evaluated at $\mathbf{x}$ rather than $\mathbf{y}$ and taken outside the integrals, giving

$$\sum_{\infty} \int_{s_p} n_k E_{ik}(\mathbf{y}) g(\mathbf{x} - \mathbf{y}) dS_y = \frac{\partial E_{ik}(\mathbf{x})}{\partial x_k} \times \int_{V_{s\infty}} g(\mathbf{x} - \mathbf{y}) dV_y - \frac{\partial}{\partial x_k} \left[E_{ik}(\mathbf{x}) \int_{V_{s\infty}} g(\mathbf{x} - \mathbf{y}) dV_y \right] = \{1 - \epsilon(\mathbf{x})\} \frac{\partial E_{ik}(\mathbf{x})}{\partial x_k} - \frac{\partial}{\partial x_k} [\{1 - \epsilon(\mathbf{x})\} E_{ik}(\mathbf{x})] = E_{ik}(\mathbf{x}) \frac{\partial \epsilon(\mathbf{x})}{\partial x_k} \tag{33}$$

Using this result Equation 31 becomes

$$\int_{V_{f\infty}} g(\mathbf{x} - \mathbf{y}) \frac{\partial E_{ik}'}{\partial y_k} dV_y = \epsilon(\mathbf{x}) \frac{\partial E_{ik}(\mathbf{x})}{\partial x_k} - \sum_{p\infty} \int_{s_p} n_k E_{ik}''(\mathbf{y}) g(\mathbf{x} - \mathbf{y}) dS_y \tag{34}$$

We know that $g(\mathbf{x} - \mathbf{y})$ varies little over distances comparable with the particle diameter and it is tempting, therefore, to simplify an integral such as that which appears in the last term of Equation 34 by replacing $g(\mathbf{x} - \mathbf{y})$ by $g(\mathbf{x} - \mathbf{x}_p)$, where $\mathbf{x}_p$ is the center of mass of particle p, then taking this function outside the integral. But if this method of simplification was generally valid, Equation 31 could have been simplified without expressing E_{ik}' as $E_{ik} + E_{ik}''$, and hence all the subsequent work could have been eliminated.

That this procedure cannot, in fact, be justified in general for integrals of the form

$$\int_{s_p} f(\mathbf{y}) n_k(\mathbf{y}) g(\mathbf{x} - \mathbf{y}) dS_y \tag{35}$$

may be seen by considering the simple case $f(\mathbf{y}) \equiv A = \text{constant}$, when the above reduces to

$$A \int_{s_p} n_k(\mathbf{y}) g(\mathbf{x} - \mathbf{y}) dS_y \tag{36}$$

If $g(\mathbf{x} - \mathbf{y})$ is replaced by $g(\mathbf{x} - \mathbf{x}_p)$, on the grounds that it is a slowly varying function, this becomes

$$A\, g(\mathbf{x} - \mathbf{x}_p) \int_{s_p} n_k(\mathbf{y})\, dS_y$$

and the integral vanishes, leading to the conclusion that (36) should vanish. However, this conclusion is false in general, since the varying component of g leads to a finite contribution to the integral, and indeed (36) may be very large if A is sufficiently large. Thus, although the variations in g are small compared with g itself, the integral would vanish if these variations were neglected and their contribution is actually more important than that of the constant component of g in evaluating the integral. It is for this reason that the integral involving E_{ik}' in Equation 31 could not have been simplified directly by this procedure, instead of expressing E_{ik}' as the sum of E_{ik} and E_{ik}'' and dealing with the resulting integrals separately.

However, while an integral of the form of (35) cannot in general be reduced by neglecting the variation of g over s_p, it is not difficult to show that this does lead to a good approximation when:

A. The mean value of $f(\mathbf{y})$, taken over s_p, is small compared with the variations about this mean.
B. The variation of $f(\mathbf{y})$ over s_p is reasonably smooth.
C. The shape of the surface s_p does not depart too much from a sphere.

Now conditions A and B are satisfied by E_{ik}'' (though not by E_{ik}'), since it represents the variation of E_{ik}' about its local mean value. The mean value of E_{ik}'' will therefore be small compared with its variations, and these variations will be smooth when considered as a function of position on the surface of a single particle, because they result principally from the distortion of the fluid streamlines to pass around the particle. Thus, if the particles are of a reasonable convex shape not too different from spheres, so that condition C is also satisfied, it is justifiable to neglect the variation of g over s_p in the last term on the right-hand side of Equation 34, which therefore becomes

$$\int_{V_{f\infty}} g(\mathbf{x} - \mathbf{y}) \frac{\partial E_{ik}'}{\partial y_k} dV_y = \epsilon(\mathbf{x}) \frac{\partial E_{ik}(\mathbf{x})}{\partial x_k} - \sum_{p\infty} g(\mathbf{x} - \mathbf{x}_p) \int_{s_p} E_{ik}'' n_k dS \tag{37}$$

Using Equations 29, 30, and 37 and the fluid continuity Equation 23, the locally averaged equation of motion (Equation 26) may be written

$$\rho_f \epsilon \left[\frac{\partial u_i}{\partial t} + u_k \frac{\partial u_i}{\partial x_k} \right] = \epsilon \frac{\partial E_{ik}}{\partial x_k} - \frac{\partial R_{ik}}{\partial x_k} + \epsilon \rho_f g_i - \sum_{p\infty} g(\mathbf{x} - \mathbf{x}_p) \int_{s_p} E_{ik}'' n_k dS \tag{38}$$

Now the force, f_{pi}, exerted on a single particle p by the surrounding fluid is given by

$$f_{pi} = \int_{s_p} E_{ik}' n_k dS = \int_{s_p} (E_{ik} + E_{ik}'') n_k dS = \int_{v_p} \frac{\partial E_{ik}}{\partial x_k} dv + \int_{s_p} E_{ik}'' n_k dS \approx v \left(\frac{\partial E_{ik}}{\partial x_k} \right)_{\mathbf{x}_p} + \int_{s_p} E_{ik}'' n_k dS$$

where we have used the divergence theorem and the fact that $\partial E_{ik}/\partial x_k$ varies little over the interior of a single particle. Multiplying both sides of this equation by $g(\mathbf{x} - \mathbf{x}_p)$ and summing over all particles, it is seen from Equation 10 that the left-hand side becomes $n(\mathbf{x}) f_i(\mathbf{x})$, where $f_i(\mathbf{x})$ is the local mean value of the force exerted on a particle by the surrounding fluid. Since $\partial E_{ik}/\partial x_k$ varies little over the radius of g, $(\partial E_{ik}/\partial x_k)_{\mathbf{x}_p}$ may be replaced by $(\partial E_{ik}/\partial x_k)_{\mathbf{x}}$ and taken outside the summa-

tion in the first term on the right-hand side, when it follows from Equation 11 that this term reduces to $[1 - \epsilon(\mathbf{x})](\partial E_{ik}/\partial x_k)_{\mathbf{x}}$.

Thus

$$\sum_{p\infty} g(\mathbf{x} - \mathbf{x}_p) \int_{s_p} E_{ik}'' n_k dS = n(\mathbf{x}) f_i(\mathbf{x}) - [1 - \epsilon(\mathbf{x})] \left(\frac{\partial E_{ik}}{\partial x_k}\right)_{\mathbf{x}} \quad (39)$$

and using this in the last term on the right-hand side of Equation 38 finally gives

$$\rho_f \epsilon \left[\frac{\partial u_i}{\partial t} + u_k \frac{\partial u_i}{\partial x_k}\right] = \frac{\partial \mathcal{E}_{ik}}{\partial x_k} - n f_i + \epsilon \rho_f g_i \quad (40)$$

where we have written

$$\mathcal{E}_{ik} = E_{ik} - R_{ik} \quad (41)$$

Equation 40 is the required momentum balance in terms of local mean variables, and it should be compared with the point equations of motion (Equation 25), which may also be written

$$\rho_f \left[\frac{\partial u_i'}{\partial t} + u_k' \frac{\partial u_i'}{\partial x_k}\right] = \frac{\partial E_{ik}'}{\partial x_k} + \rho_f g_i \quad (25')$$

The most obvious difference is the introduction of the fluid-particle interaction force, nf_i, as an explicit term in the equations, though it influences the point equations of motion only through their boundary values at the particle surfaces. The coupling between the momentum equations for the fluid and the particles is therefore explicit when the equations are written in terms of local mean variables. The local mean stress tensor, E_{ik}, is also augmented by the tensor, R_{ik}, which is analogous to the Reynolds stress in the averaged descriptions of turbulent flow. This has the dimensions of stress and enters the equations in the form expected for the divergence of a stress tensor, but it appears only because the left-hand side of Equation 40 is not strictly equal to the locally averaged rate of change of momentum of the fluid, as seen from Equation 29.

Equations of Motion for the Particles

The required continuum equations for the particles are obtained in an analogous way by locally averaging the equations of motion for a single particle. The equations of motion for the center of mass of a single particle are

$$\nu \rho_s \frac{dv_{pi}}{dt} = f_{pi} + \nu \rho_s g_i + \phi_{pi} \quad (42)$$

where v_{pi} is the ith component of the velocity of the center of mass, f_{pi}, as before, the force exerted by the surrounding fluid on the particle, and ϕ_{pi} the force experienced by the particle as a result of direct contact with other particles. (For a complete description of the motion of a particle other equations describing rotational motion are, of course, necessary.) The local average of each side of Equation 42 may be formed using the process defined by Equation 10, giving

$$\nu \rho_s \sum_{p\infty} g(\mathbf{x} - \mathbf{x}_p) \frac{dv_{pi}}{dt} = n(\mathbf{x}) f_i(\mathbf{x}) + [1 - \epsilon(\mathbf{x})] \rho_s g_i + n(\mathbf{x}) \phi_i(\mathbf{x}) \quad (43)$$

where we have introduced f_i, the local mean fluid-particle interaction force already defined, and also a corresponding local mean particle-particle interaction force ϕ_i, defined by

$$n(\mathbf{x}) \phi_i(\mathbf{x}) = \sum_{p\infty} g(\mathbf{x} - \mathbf{x}_p) \phi_{pi} \quad (44)$$

The second term on the right-hand side of Equation 43 has also been simplified by using Equation 11 and the fact that $n\nu = 1 - \epsilon$.

Now

$$\frac{\partial}{\partial t}[n(\mathbf{x}) v_i(\mathbf{x})] = \frac{\partial}{\partial t} \sum_{p\infty} v_{pi} g(\mathbf{x} - \mathbf{x}_p) = \sum_{p\infty} \left\{\frac{dv_{pi}}{dt} g(\mathbf{x} - \mathbf{x}_p) - v_{pi} v_{pk} \frac{\partial g}{\partial x_k}\right\}$$

whence

$$\nu \rho_s \sum_{p\infty} \frac{dv_{pi}}{dt} g(\mathbf{x} - \mathbf{x}_p) = \nu \rho_s \frac{\partial}{\partial t}[n(\mathbf{x}) v_i(\mathbf{x})] + \nu \rho_s \frac{\partial}{\partial x_k} \sum_{p\infty} v_{pi} v_{pk} g(\mathbf{x} - \mathbf{x}_p) \quad (45)$$

where v_i is the local mean particle velocity.

But v_{pi} may be expressed as the sum of its local mean value and a fluctuation

$$v_{pi} = v_i(\mathbf{x}_p) + v_{pi}''$$

whence

$$\sum_{p\infty} v_{pi} v_{pk} g(\mathbf{x} - \mathbf{x}_p) = \sum_{p\infty} v_i(\mathbf{x}_p) v_k(\mathbf{x}_p) g(\mathbf{x} - \mathbf{x}_p) + \sum_{p\infty} [v_i(\mathbf{x}_p) v_{pk}'' + v_k(\mathbf{x}_p) v_{pi}''] g(\mathbf{x} - \mathbf{x}_p) + \sum_{p\infty} v_{pi}'' v_{pk}'' g(\mathbf{x} - \mathbf{x}_p)$$

and by reasoning analogous to that following Equation 28, this may be reduced to

$$\nu \rho_s \sum_{p\infty} v_{pi} v_{pk} g(\mathbf{x} - \mathbf{x}_p) = \nu \rho_s n(\mathbf{x}) v_i(\mathbf{x}) v_k(\mathbf{x}) + S_{ik}(\mathbf{x}) \quad (46)$$

where

$$S_{ik}(\mathbf{x}) = \nu \rho_s \sum_{p\infty} v_{pi}'' v_{pk}'' g(\mathbf{x} - \mathbf{x}_p) \quad (47)$$

Using Equation 46 in the second term on the right-hand side of Equation 45, and using 45 to evaluate the left-hand side of Equation 43, we obtain

$$\nu \rho_s \left[\frac{\partial}{\partial t}(n v_i) + \frac{\partial}{\partial x_k}(n v_i v_k)\right] = n \phi_i - \frac{\partial S_{ik}}{\partial x_k} + n f_i + (1 - \epsilon) \rho_s g_i$$

or, in view of the particle continuity Equation 24',

$$\rho_s (1 - \epsilon) \left[\frac{\partial v_i}{\partial t} + v_k \frac{\partial v_i}{\partial x_k}\right] = n \phi_i - \frac{\partial S_{ik}}{\partial x_k} + n f_i + (1 - \epsilon) \rho_s g_i \quad (48)$$

which is the required equation of motion for the particles in terms of local mean variables. Comparing Equations 40 and 48, the fluid particle interaction force is seen to appear in the same form but with opposite signs, thus ensuring that Newton's third law is satisfied.

Evaluation of Undetermined Terms in Equations of Motion

The equations of motion have now been expressed in the form

$$\rho_f \epsilon \left[\frac{\partial u_i}{\partial t} + u_k \frac{\partial u_i}{\partial x_k}\right] = \frac{\partial \mathcal{E}_{ik}}{\partial x_k} - n f_i + \epsilon \rho_f g_i \quad (40)$$

and

$$\rho_s (1 - \epsilon) \left[\frac{\partial v_i}{\partial t} + v_k \frac{\partial v_i}{\partial x_k}\right] = n \phi_i - \frac{\partial S_{ik}}{\partial x_k} + n f_i + (1 - \epsilon) \rho_s g_i \quad (48)$$

obtained by pure manipulation from the point equation of motion of the fluid and the equation of motion for the center of mass of a single particle, using only the assumption that the scale of "macroscopic" variation is much larger than the particle spacing. These equations, together with the equations of continuity, do not, of course, determine the state of motion of the system unless the dependence of $\mathcal{E}_{ik}$, ϕ_i, S_{ik}, and f_i on the voidage, the local mean velocities, and the pressure is known. Thus the effect of the preceding manipulations is not to eliminate the empiricism of the theory, but merely to concentrate it in these particular terms. However, this represents a genuine advance, since we believe it is possible to make guesses at the form of these terms which are more rationally based than attempts to guess the complete form of the equations without preliminaries.

The empiricism required to complete the above equations differs little, in principle, from that required to obtain the Navier-Stokes equations themselves from the momentum balance

$$\rho_f \left[\frac{\partial u_i'}{\partial t} + u_k' \frac{\partial u_i'}{\partial x_k} \right] = \frac{\partial E_{ik}'}{\partial x_k} + \rho_f g_i$$

for a single fluid. This requires the postulation of a form for the constitutive equations relating E_{ik}' to the fluid pressure and velocity and, in the same way, we now need constitutive equations relating $\mathcal{E}_{ik}$, ϕ_i, S_{ik}, and f_i to the voidage, pressure, and local mean velocity fields.

Consider first the direct interaction force, $n\phi_i$, between the particles. When they rest in permanent contact, as in a packed bed, the assembly can sustain both isotropic compressive stresses and, to a limited extent, shear stresses without motion. When fluidized, however, the particles do not rest in permanent contact with each other, and although they can support a compressive stress by momentum exchange through collisions without continued displacement, any shear stress generates a motion which continues as long as the stress is maintained, as in a fluid. In the fluidized state we might therefore surmise that the particle-particle interaction will be expressible as the divergence of a stress tensor

$$n\phi_i = \partial E_{ik}^s / \partial x_k$$

where E_{ik}^s contains an isotropic term depending on the voidage, and representing the elastic resistance of the particle assembly to compression, together with terms related to the rate of strain tensor and representing the fluid-like behavior of the assembly. The simplest physically reasonable procedure is to lump the "Reynolds stress" term $\partial S_{ik}/\partial x_k$ with $n\phi_i$, writing

$$n\phi_i - \frac{\partial S_{ik}}{\partial x_k} = \frac{\partial E_{ik}^s}{\partial x_k} - \frac{\partial S_{ik}}{\partial x_k} = \frac{\partial \mathcal{E}_{ik}^s}{\partial x_k} \tag{49}$$

and to assume that the form of $\mathcal{E}_{ik}^s$ is analogous to the stress tensor of a Newtonian fluid—namely,

$$\mathcal{E}_{ik}^s = -p^s(\epsilon)\delta_{ik} + \lambda^s(\epsilon) \frac{\partial v_m}{\partial x_m} \delta_{ik} + \mu^s(\epsilon) \left[\frac{\partial v_i}{\partial x_k} + \frac{\partial v_k}{\partial x_i} - \frac{2}{3} \delta_{ik} \frac{\partial v_m}{\partial x_m} \right] \tag{50}$$

where δ_{ik} is the Kronecker delta. $p^s(\epsilon)$ then represents an interparticle "pressure," while λ^s and μ^s represent effective bulk and shear viscosities for the particle assembly, all three being assumed to be local variables depending on the local mean voidage, ϵ. Although the behavior of the particle assembly in shear is almost certainly inadequately represented by Equation 50 at very low rates of shear, there is some evidence (Anderson and Bryden, 1965) that the behavior is approximately Newtonian at higher rates of shear. Using Equation 49 the particle equation of motion (Equation 48) can be written in a form symmetric with the fluid equation

$$\rho_s(1-\epsilon) \left[\frac{\partial v_i}{\partial t} + v_k \frac{\partial v_i}{\partial x_k} \right] = \frac{\partial \mathcal{E}_{ik}^s}{\partial x_k} + nf_i + (1-\epsilon)\rho_s g_i \tag{51}$$

The fluid equation of motion (Equation 40) also contains the divergence of a stress tensor, E_{ik}, together with the divergence of the "Reynolds stress" tensor, R_{ik}, the two being combined in the term $\partial \mathcal{E}_{ik}/\partial x_k$. As in the case of the particles, the simplest physically reasonable assumption is that $\mathcal{E}_{ik}$ is given in terms of the local mean fluid variables by an expression analogous to that for the stress tensor in a Newtonian fluid—namely

$$\mathcal{E}_{ik} = -p\delta_{ik} + \lambda(\epsilon) \frac{\partial u_m}{\partial x_m} \delta_{ik} + \mu(\epsilon) \left[\frac{\partial u_i}{\partial x_k} + \frac{\partial u_k}{\partial x_i} - \frac{2}{3} \delta_{ik} \frac{\partial u_m}{\partial x_m} \right] \tag{52}$$

where p is the local mean fluid pressure and λ and μ are effective bulk and shear viscosities which take account of the stresses of Reynolds type as well as the true mechanical stresses. Both λ and μ would be expected to depend on the local voidage, as indicated.

It remains to evaluate f_i, the local mean value of the force per particle resulting from interaction with the fluid, and it is in the estimation of this term that differences principally arise in existing publications. The form of f_i is suggested by the solution of another much simpler problem which nevertheless has features in common with the present one—namely, the evaluation of the interaction force between a single spherical particle and an infinite fluid for a slow rectilinear motion of the particle starting from rest at $t = 0$. With the assumptions that the fluid is also at rest at $t = 0$ and that there is no gravitational field, it was shown in the classical work of Basset (1888) that the interaction is such as to impede the motion, and its magnitude is given by

$$f_p = \alpha^0 v_p(t) + C^0 \rho_f \frac{dv_p}{dt} - \frac{9\nu}{d_p} \sqrt{\frac{\rho_f \mu}{\pi}} \int_0^t \frac{dv_p(t-\tau)}{d\tau} \cdot \frac{d\tau}{\sqrt{\tau}} \tag{53}$$

where $v_p(t)$ is the velocity of the particle as a function of time, μ is the viscosity of the fluid, and d_p is the diameter of the particle. α^0 and C^0 are referred to as the drag and virtual mass coefficients, respectively, and for Basset's problem they are given by

$$\alpha^0 = \frac{18\nu\mu}{d_p^2}; \quad C^0 = 1/2$$

The first term on the right-hand side of Equation 53 represents a drag proportional to the particle velocity, and takes the well known Stokesian form. The second term represents an interaction force proportional to the acceleration, and is therefore known as a virtual mass term. The final term of Equation 53, sometimes called the "Basset force," must be included because the interaction force depends, not only on the instantaneous motion of the particle, but also on the instantaneous fluid velocity field in which it moves, which in turn depends on the complete history of the particle's motion.

The situation of a particle in a fluidized bed is much more complicated, but regarding the difference between the local mean particle and fluid velocities as equivalent to $v_p(t)$ above,

we might expect to find drag and virtual mass forces depending, respectively, on the relative (local mean) velocity and acceleration of the particles and the fluid. It seems less reasonable, however, to include a term analogous to the Basset force, since the presence of a large assembly of particles dispersed throughout the fluid is likely to erase any historical effect of the motion of a given particle on the fluid flow in its own neighborhood. Thus it appears not unreasonable to postulate a local mean interaction force of the form

$$f_i = \alpha(\epsilon)(u_i - v_i)\, h(|u_i - v_i|) + C(\epsilon)\nu\rho_f \frac{d}{dt}(u_i - v_i) \quad (54)$$

where the first term represents a drag force in the direction of the relative velocity $(u_i - v_i)$ but not necessarily proportional to it in magnitude, and the second term a virtual mass force proportional to the mass of fluid displaced by a particle. Both the drag coefficient, α, and the virtual mass coefficient, C, would be expected to depend on the voidage, as indicated, and there is good experimental evidence that the drag coefficient increases monotonically with decreasing ϵ, the rate of increasing becoming large as ϵ approaches the voidage at minimum fluidization.

The virtual mass term calls for further comment, as the relative acceleration is not a well defined quantity. For example,

$$\frac{d}{dt}(u_i - v_i) \equiv \left(\frac{\partial u_i}{\partial t} + u_k \frac{\partial u_i}{\partial x_k}\right) - \left(\frac{\partial v_i}{\partial t} + v_k \frac{\partial v_i}{\partial x_k}\right) \quad (55)$$

and

$$\frac{d}{dt}(u_i - v_i) \equiv \frac{\partial}{\partial t}(u_i - v_i) + v_k \frac{\partial}{\partial x_k}(u_i - v_i) \quad (56)$$

both reduce to the particle acceleration term $-(\partial v_i/\partial t + v_k \partial v_i/\partial x_k)$ when $u_i \equiv 0$ and to zero when $u_i \equiv v_i$, and both are therefore acceptable as definitions of the relative acceleration. The second may be slightly more appealing on rational grounds, since it evaluates the change in relative velocity following the particle motion and reduces to zero when the fluid motion is steady and the particles are at rest, but there seems to be no sound *a priori* way of choosing the form of the relative acceleration term. This difficulty has been pointed out by Murray (1965).

If the expression (Equation 54) for the interaction force is substituted into the particle equation of motion (Equation 51), we obtain

$$\rho_s(1 - \epsilon)\left[\frac{\partial v_i}{\partial t} + v_k \frac{\partial v_i}{\partial x_k}\right] = \frac{\partial \mathcal{E}_{ik}^s}{\partial x_k} + n\alpha(u_i - v_i)\, h(|u_i - v_i|) + nC\rho_f \frac{d}{dt}(u_i - v_i) + (1 - \epsilon)\rho_s g_i \quad (57)$$

which is clearly unsatisfactory in at least one respect since, although it takes account of the gravitational body force on the particles, it does not include any buoyancy force offsetting this term. Physically the buoyancy force is a result of the action on the particles of one component of the fluid stress—namely, the vertical pressure gradient induced by the gravitational body force. It should therefore enter the equations through the fluid-particle interaction force, and one way of introducing it would be to augment Equation 54 with a term $-\nu\rho_f g_i$, giving

$$f_i = \alpha(\epsilon)(u_i - v_i)\, h(u_i - v_i) + C(\epsilon)\nu\rho_f \frac{d}{dt}(u_i - v_i) - \nu\rho_f g_i \quad (54')$$

However, we believe a more radical modification of the interaction force is necessary to obtain satisfactory equations of motion. Perhaps this is best introduced by considering, once again, a single particle in interaction with a fluid.

Figure 1 (left) shows a vertical cylindrical stream tube of a fluid whose velocity is everywhere u in the upward vertical direction. If a particle is immersed at rest in this fluid, as indicated, it will experience a drag force $f_d(u)$ in the direction of fluid flow, so that the total force on the particle in the upward vertical direction is

$$f = f_d(u) - \nu(\rho_s - \rho_f)g \quad (58)$$

where the second term represents gravitational and buoyancy forces.

Now suppose the undisturbed fluid has slowly diverging streamlines so that a stream tube has the form indicated in Figure 1 (right). There will now be an additional pressure gradient

$$\frac{dp_1}{dx} = -\rho_f u \frac{du}{dx} \quad (59)$$

due to the fluid deceleration. If the particle is immersed in this flow field at a point on the vertical streamline where the velocity has the same value u as before, the drag force will not differ appreciably from $f_d(u)$, provided the divergence of the streamlines is not too sharp. However, there will now be an additional component of force,

$$-\nu \frac{dp_1}{dx} = \rho_f \nu u \frac{du}{dx} \quad (60)$$

due to the reaction of the additional pressure gradient (Equation 59) on the particle. Thus Equation 58 is replaced by

$$f = f_d(u) - \nu(\rho_s - \rho_f)g + \rho_f \nu u \frac{du}{dx} \quad (61)$$

Equations 58 and 61 can be brought into a common form by writing

$$f = f_d(u) - \nu\rho_s g - \nu \frac{dp}{dx} \quad (62)$$

since the pressure gradient dp/dx in the undisturbed fluid, without the particle, is given by

$$\frac{dp}{dx} = -\rho_f g$$

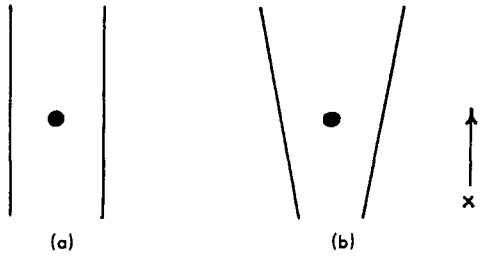

Figure 1. Single particle suspended in a fluid stream

for flow with parallel streamlines, and by

$$\frac{dp}{dx} = -\rho_f g - \rho_f u \frac{du}{dx}$$

for flow with diverging streamlines.

In Equation 62 the terms

$$f_d(u) - \nu \frac{dp}{dx} \tag{63}$$

represent the complete fluid-particle interaction force. This is the sum of two terms, $f_d(u)$ representing the forces of skin friction and form drag arising from small scale distortions of the fluid streamlines in the neighborhood of the particle, and $-\nu dp/dx$ representing the reaction on the particle of large scale pressure gradients in the fluid. The simple addition of a buoyancy force to the drag force, as in Equation 58, takes only partial account of the second term in Equation 63. Physically, the particle cannot distinguish the part arising from gravity from any other contributions to the fluid pressure gradient, so it is not rational to include the buoyancy reaction, as in Equation 58, without including the effects of other contributions such as accelerational pressure gradients in the fluid. However, provided the stress in the fluid is a pure pressure, the form given by Equation 62 is of general validity and all relevant contributions are included when dp/dx is evaluated from the fluid equation of motion—i.e., Bernoulli's equation in the above example.

Turning now to the case of an assembly of particles, the force exerted on a single particle as a result of its interaction with the fluid is

$$f_{pi} = \int_{s_p} E_{ik}' n_k dS$$

and, corresponding to Equation 63, we wish to decompose this into a component due to "macroscopic" variations in the fluid stress tensor on a scale large compared with the particle spacing, together with a component representing the effect of detailed variations of the point stress tensor as the fluid flows around the particle. This second component will include skin friction and form drag contributions, and corresponds to the term $f_d(u)$ in Equation 63. The simplest decomposition of this sort is achieved by writing E_{ik}' in the form

$$E_{ik}' = E_{ik} + E_{ik}''$$

but in the first term we would like to include the "Reynolds stresses" represented by the tensor R_{ik}. Accordingly we modify the above to

$$E_{ik}' = \mathcal{E}_{ik} + \mathcal{D}_{ik}$$

so that

$$f_{pi} = \int_{s_p} \mathcal{E}_{ik}\, n_k dS + \int_{s_p} \mathcal{D}_{ik} n_k dS$$

$$= \int_{v_p} \frac{\partial \mathcal{E}_{ik}}{\partial x_k} dV + \int_{s_p} \mathcal{D}_{ik} n_k dS$$

where Gauss's theorem has been used to transform the first term, since $\mathcal{E}_{ik}$ is defined at all points of space. (E_{ik}' and $\mathcal{D}_{ik}$ are, of course, defined only at points occupied by fluid.) Since $\partial \mathcal{E}_{ik}/\partial x_k$ varies little over the interior of a particle, this becomes

$$f_{pi} = \nu \left(\frac{\partial \mathcal{E}_{ik}}{\partial x_k}\right)_{\mathbf{x}_p} + \tilde{f}_{pi} \tag{64}$$

where

$$\tilde{f}_{pi} = \int_{s_p} \mathcal{D}_{ik} n_k dS$$

Using Equation 64

$$n(\mathbf{x}) f_i(\mathbf{x}) = \sum_{p\infty} f_{pi}\, g(\mathbf{x} - \mathbf{x}_p) = \nu \sum_{p\infty} \left(\frac{\partial \mathcal{E}_{ik}}{\partial x_k}\right)_{\mathbf{x}_p} g(\mathbf{x} - \mathbf{x}_p) + \sum_{p\infty} \tilde{f}_{pi}\, g(\mathbf{x} - \mathbf{x}_p) \approx$$

$$\nu \left(\frac{\partial \mathcal{E}_{ik}}{\partial x_k}\right)_{\mathbf{x}} \sum_{p\infty} g(\mathbf{x} - \mathbf{x}_p) + \sum_{p\infty} \tilde{f}_{pi}\, g(\mathbf{x} - \mathbf{x}_p)$$

since $\partial \mathcal{E}_{ik}/\partial x_k$ varies little over distances comparable with the radius of g. Thus

$$nf_i = n\nu \frac{\partial \mathcal{E}_{ik}}{\partial x_k} + n\tilde{f}_i = (1 - \epsilon) \frac{\partial \mathcal{E}_{ik}}{\partial x_k} + n\tilde{f}_i \tag{65}$$

where

$$n\tilde{f}_i = \sum_{p\infty} \tilde{f}_{pi}\, g(\mathbf{x} - \mathbf{x}_p)$$

Then $n\tilde{f}_i$ represents the part of the total fluid-particle interaction force per unit bed volume arising from the detailed variations in the stress tensor induced by fluctuations in velocity as the fluid passes around individual particles and through the interstices between particles.

Referring to the discussion of the single particle above, f_{pi} is analogous to the total interaction force and $\tilde{f}_{pi}$ to the force $f_d(u)$; indeed if $\mathcal{E}_{ik}$ represents a pure pressure, $\partial \mathcal{E}_{ik}/\partial x_k = -\partial p/\partial x_i$, and the right-hand side of Equation 64 is formally identical with 63. Therefore it is $\tilde{f}_i$, rather than f_i, which would be expected to be expressible in the form of Equation 54, and we should write

$$n\tilde{f}_i = n\alpha(\epsilon)(u_i - v_i)\, h(|u_i - v_i|) + n\nu C(\epsilon)\rho_f \frac{d}{dt}(u_i - v_i) \tag{66}$$

in place of Equation 54.

Using Equation 65, the equations of motion (40 and 51) become

$$\rho_f \epsilon \left[\frac{\partial u_i}{\partial t} + u_k \frac{\partial u_i}{\partial x_k}\right] = \epsilon \frac{\partial \mathcal{E}_{ik}}{\partial x_k} - n\tilde{f}_i + \epsilon \rho_f g_i \tag{67}$$

and

$$\rho_s(1 - \epsilon) \left[\frac{\partial v_i}{\partial t} + v_k \frac{\partial v_i}{\partial x_k}\right] = (1 - \epsilon) \frac{\partial \mathcal{E}_{ik}}{\partial x_k} + n\tilde{f}_i + (1 - \epsilon)\rho_s g_i + \frac{\partial \mathcal{E}_{ik}^s}{\partial x_k} \tag{68}$$

with $\mathcal{E}_{ik}$ given by Equation 52, $\mathcal{E}_{ik}^s$ by Equation 50, and $n\tilde{f}_i$ by Equation 66.

•The six Equations 67 and 68, together with the continuity equations, provide a set of eight equations in the eight variables u_i, v_i, ϵ, and p. They are thus the equations required to provide a mechanical description of the system in terms of local mean variables.

However, although Equations 40 and 48 are directly derivable from the basic equations of fluid mechanics for the system, the subsequent developments leading to Equations 67 and 68 reflect our own opinions of the most appropriate form for the undetermined terms in Equations 40 and 48. At the present time there remains considerable scope for differing opinions on these points.

We have implicitly assumed the temperature to be specified, so that quantities such as ρ_f and ρ_s may be regarded as given. Thus our equations are appropriate only in cases where large rates of heat transfer are not involved. Strictly speaking, the temperature is another dependent variable, and should be determined by a further differential equation representing energy balance. A form for such an energy equation has been postulated, for example, by Murray (1965).

Discussion of Equations and Comparison with Previously Published Work

One of the earliest descriptions of equations of motion for a system of particles dispersed in a fluid is due to van Deemter and van der Laan (1961). Transcribed into the present notation, their equations are

$$\rho_f \epsilon \left[\frac{\partial u_i}{\partial t} + u_k \frac{\partial u_i}{\partial x_k}\right] = \frac{\partial E_{ik}}{\partial x_k} - F_i + \epsilon \rho_f g_i \tag{69}$$

and

$$\rho_s(1-\epsilon)\left[\frac{\partial v_i}{\partial t} + v_k \frac{\partial v_i}{\partial x_k}\right] = \frac{\partial E_{ik}^s}{\partial x_k} + F_i + (1-\epsilon)\rho_s g_i \tag{70}$$

where F_i is the ith component of the total force per unit volume exerted on the particles by the continuous phase. F_i is therefore identical with the quantity we have denoted by nf_i, and the similarity between the above equations and our intermediate forms 40 and 51 is obvious; indeed our equations differ from 69 and 70 only by the inclusion of the "Reynolds stress" terms R_{ik} and S_{ik} thrown up by the process of local averaging. However, van Deemter and van der Laan did not speculate about the form of F_i, so further comparison is not possible.

In a much more recent publication Pigford and Baron (1965) proposed equations of the form

$$\rho_f \epsilon \left[\frac{\partial u_i}{\partial t} + u_k \frac{\partial u_i}{\partial x_k}\right] = -\epsilon \frac{\partial p}{\partial x_i} + X_{fi} - \frac{\mu}{\kappa}(u_i - v_i) \tag{71}$$

$$\rho_s(1-\epsilon)\left[\frac{\partial v_i}{\partial t} + v_k \frac{\partial v_i}{\partial x_k}\right] = -(1-\epsilon)\frac{\partial p}{\partial x_i} + X_{si} + \frac{\mu}{\kappa}(u_i - v_i) \tag{72}$$

where p is the fluid pressure, X_{fi} and X_{si} are the ith components of the body forces per unit bed volume on the fluid and the particles, respectively, and μ/κ is a drag coefficient proportional to the fluid viscosity μ, and inversely proportional to the "permeability," κ, of the assembly of particles. These authors explicitly neglected the direct transmission of momentum between the particles and the transmission of shear stresses by the fluid in deriving the above equations, which should therefore be compared with our Equations 67 and 68 after setting

$$\mathcal{E}_{ik} = -p\delta_{ik} \quad \text{and} \quad \mathcal{E}_{ik}^s = 0$$

Noting that $X_{fi} = \epsilon \rho_f g_i$ and $X_{si} = (1-\epsilon)\rho_s g_i$ when the only body force is gravity, Equations 71 and 72 are of the same form as 67 and 68, provided

$$n\bar{f}_i = \frac{\mu}{\kappa}(u_i - v_i)$$

This has the same form as our Equation 66 if virtual mass effects are neglected, so Pigford and Baron's equations agree with the special case of ours for which

$$\mathcal{E}_{ik} = -p\delta_{ik},\ \mathcal{E}_{ik}^s = 0, \text{ and } C(\epsilon) = 0$$

Some caution must be exercised, however, in relating the factor μ/κ to experimentally determined drag coefficients.

Several years ago one of the present writers based some predictions of the behavior of fluidized systems on the following equations of motion (Jackson, 1963),

$$\rho_f \left[\frac{\partial u_i}{\partial t} + u_k \frac{\partial u_i}{\partial x_k}\right] = -\frac{\partial p}{\partial x_i} - \beta(\epsilon)(u_i - v_i) + \rho_f g_i \tag{73}$$

and

$$\rho_s(1-\epsilon)\left[\frac{\partial v_i}{\partial t} + v_k \frac{\partial v_i}{\partial x_k}\right] - \rho_f(1-\epsilon)\left[\frac{\partial u_i}{\partial t} + u_k \frac{\partial u_i}{\partial x_k}\right] = (\rho_s - \rho_f)(1-\epsilon)g_i + \beta(\epsilon)(u_i - v_i) \tag{74}$$

which appear, at first sight, to have a structure rather different from those now proposed. However, any two independent linear combinations of Equations 67 and 68 will serve equally well in their place. In particular, we might take Equation 67 together with an equation obtained by multiplying Equation 67 by $(1-\epsilon)/\epsilon$ and subtracting from Equation 68. This gives the following pair of equations,

$$\rho_f \left[\frac{\partial u_i}{\partial t} + u_k \frac{\partial u_i}{\partial x_k}\right] = \frac{\partial \mathcal{E}_{ik}}{\partial x_k} - \frac{n\bar{f}_i}{\epsilon} + \rho_f g_i \tag{67'}$$

and

$$\rho_s(1-\epsilon)\left[\frac{\partial v_i}{\partial t} + v_k \frac{\partial v_i}{\partial x_k}\right] - \rho_f(1-\epsilon)\left[\frac{\partial u_i}{\partial t} + u_k \frac{\partial u_i}{\partial x_k}\right] = \frac{n\bar{f}_i}{\epsilon} + (1-\epsilon)(\rho_s - \rho_f)g_i + \frac{\partial \mathcal{E}_{ik}^s}{\partial x_k} \tag{75}$$

where Equation 67′ is obtained from 67 simply by dividing through by ϵ. If now we make the same approximations as Pigford and Baron, writing $\mathcal{E}_{ik} = -p\delta_{ik}$, $\mathcal{E}_{ik}^s = 0$, and $n\bar{f}_i/\epsilon$, $\propto (u_i - v_i)$, these equations reduce to 73 and 74, when β is the factor of proportionality between $n\bar{f}_i/\epsilon$ and $(u_i - v_i)$. Thus Equations 73 and 74 are equivalent to the equations of Pigford and Baron, and both these sets of equations correspond to those proposed in the present work, with certain radical simplifying assumptions.

Transposing one term, Equation 75 may also be written

$$\rho_s(1-\epsilon)\left[\frac{\partial v_i}{\partial t} + v_k \frac{\partial v_i}{\partial x_k}\right] = \frac{n\bar{f}_i}{\epsilon} + (1-\epsilon)(\rho_s - \rho_f)g_i + \rho_f(1-\epsilon)\left[\frac{\partial u_i}{\partial t} + u_k \frac{\partial u_i}{\partial x_k}\right] + \frac{\partial \mathcal{E}_{ik}^s}{\partial x_k} \tag{75'}$$

when the right-hand side gives the total force (including Reynolds stress effects) experienced by the particles in unit bed volume. Essentially Equation 75′ is obtained from Equation 68 by substituting for $\partial \mathcal{E}_{ik}/\partial x_k$ its value found from Equation 67, and the result is an equation of motion for the particles which does not contain the fluid stress tensor. This elimination of $\mathcal{E}_{ik}$ has introduced a buoyancy force $-\rho_f(1-\epsilon)g_i$ and a term $\rho_f(1-\epsilon)[\partial u_i/\partial t + u_k \partial u_i/\partial x_k]$ representing the reaction on the particles of pressure gradients associated with fluid accelerations. The necessity for both these terms has previously been discussed in relation to a single particle in a fluid stream, where Equation 61 gave the interaction force. Comparison of the right-hand sides of Equations 61 and 75′ shows that precisely analogous terms appear in both, except that a particle-particle interaction term, $\mathcal{E}_{ik}^s$, is clearly inappropriate in the case of a single particle. The term $\rho_f(1-\epsilon)[\partial u_i/\partial t + u_k \partial u_i/\partial x_k]$, like the virtual mass term in Equation 66, introduces the fluid acceleration into the

particle equation of motion. However, not only is the physical origin of these terms distinct, but also their mathematical form, since the virtual mass term depends on the relative acceleration of the fluid and particles, while the above term depends on the acceleration of the fluid relative to the frame of reference in which the equations are written. The difference is not merely a matter of choice of the frame of reference, since it is not possible to find a single frame which moves everywhere with one phase or the other except in the special case where u or v is uniform in space.

We return now to the question, mentioned briefly above when discussing the equations of Pigford and Baron, of relating quantities appearing in the equations of motion to experimentally based drag coefficients. Consider the state of uniform fluidization, in which a uniform upward fluid flow supports a uniform assembly of particles at rest. Then Equation 75′ reduces to

$$0 = \frac{n\bar{f}_i}{\epsilon} + (1 - \epsilon)(\rho_s - \rho_f)g_i + \partial\mathcal{E}_{ik}^s/\partial x_k \tag{76}$$

Furthermore, the relative acceleration of fluid and particles vanishes, so the second term on the right-hand side of Equation 66 vanishes, and

$$n\bar{f}_i = n\alpha(\epsilon)(u_i - v_i)h(|u_i - v_i|)$$

Using this, Equation 76 becomes

$$-(1 - \epsilon)(\rho_s - \rho_f)g_i = \frac{n\alpha(\epsilon)}{\epsilon}(u_i - v_i)h(|u_i - v_i|) + \frac{\partial\mathcal{E}_{ik}^s}{\partial x_k} \tag{77}$$

The left-hand side of this is the buoyant weight of the particles, so the right-hand side represents the force experienced by the particles in unit bed volume which, in this case, must just balance the buoyant weight. The last term represents the mutual support derived from collisions between particles, so the term

$$\frac{n\alpha(\epsilon)}{\epsilon}(u_i - v_i)\,h(|u_i - v_i|) \tag{78}$$

must represent the total drag force exerted by the fluid on the particles.

Now the total drag force per unit volume is a quantity which has been measured directly (Rowe, 1961) for an assembly of particles constrained to remain at rest in a uniform fluid stream, and has also been inferred from measurements of the pressure drop through particle beds (Carman, 1956) and from determinations of the settling velocity of uniform suspensions through stationary fluid (Richardson and Zaki, 1954). The results of these measurements have been correlated by equations of the form

$$\text{Drag per unit volume} = \beta(\epsilon)(u_i - v_i)h(|u_i - v_i|)$$

and comparing this with Equation 78 it is seen that the experimentally determined drag coefficient, β, must be identified with $n\alpha/\epsilon$ rather than $n\alpha$. Equation 66 could therefore be written in the alternative form

$$n\bar{f}_i = \epsilon\beta(\epsilon)(u_i - v_i)h(|u_i - v_i|) + (1 - \epsilon)C(\epsilon)\rho_f\frac{d}{dt}(u_i - v_i) \tag{66′}$$

in terms of the experimental drag coefficient, β. Substitution of this form in Equations 67′ and 75 and comparison of the result with Equations 73 and 74 show that β is identical with the drag coefficient in Jackson's (1963) form of the equations of motion, thus justifying the use of the same symbol for these quantities. However, it would not be correct to identify Pigford and Baron's coefficient μ/κ with the experimental coefficient. Comparison of their Equations 71 and 72 with Equations 66, 67, and 68 shows that μ/κ should be identified with $n\alpha(\epsilon)$, and hence equated to ϵ times the experimental drag coefficient.

The physical reason for these differences is clear when we recall that the term $n\bar{f}_i$, which we wrote as the sum of a drag force and a virtual mass term, represents only part of the fluid-particle interaction force. According to Equation 65 it must be augmented by a term $(1 - \epsilon)\partial\mathcal{E}_{ik}/\partial x_k$ to give the total interaction force, and this term becomes $-(1 - \epsilon)\partial p/\partial x_i$ in the uniform fluidization situation. Thus $n\bar{f}_i$ is augmented by a force proportional to the pressure gradient in the fluid, and the total drag experienced by the particles is greater than $n\bar{f}_i$; indeed, as shown in Equation 77, it is increased to $n\bar{f}_i/\epsilon$. But our coefficient $n\alpha(\epsilon)$ is defined in terms of the partial interaction force, $n\bar{f}_i$, while the experimental drag coefficient is related to the total force, $n\bar{f}_i/\epsilon$, which explains why they differ by a factor ϵ.

Recently Murray (1965) has proposed a set of equations of motion which take account of shear stresses transmitted in both phases, and of virtual mass effects, and are therefore more complete than the equations of Jackson or of Pigford and Baron. In the present notation, Murray's equations are

$$\rho_f\epsilon\left[\frac{\partial u_i}{\partial t} + u_k\frac{\partial u_i}{\partial x_k}\right] = -\frac{\partial p}{\partial x_i} + \epsilon\frac{\partial\sigma_{ik}}{\partial x_k} - A(u_i - v_i) - (1 - \epsilon)C\rho_f\frac{d}{dt}(u_i - v_i) + \rho_f g_i + \rho_f\epsilon F_{fi} \tag{79}$$

and

$$\rho_s(1 - \epsilon)\left[\frac{\partial v_i}{\partial t} + v_k\frac{\partial v_i}{\partial x_k}\right] = (1 - \epsilon)\frac{\partial\sigma_{ik}^s}{\partial x_k} + A(u_i - v_i) + (1 - \epsilon)C\rho_f\frac{d}{dt}(u_i - v_i) + (1 - \epsilon)(\rho_s - \rho_f)g_i + \rho_s(1 - \epsilon)F_{si} \tag{80}$$

where σ_{ik} and σ_{ik}^s are the parts of the fluid and particle stress tensors representing pure shears, so that

$$E_{ik} = -p\delta_{ik} + \sigma_{ik} \tag{81}$$

$$E_{ik}^s = -p^s\delta_{ik} + \sigma_{ik}^s \tag{82}$$

and the contribution to the particle equation from the isotropic stress p^s is assumed to be negligible. F_{fi} and F_{si} represent body forces per unit mass, in addition to gravity, acting on the fluid and particle phases, respectively. Murray includes these to take account, for example, of possible electrical effects if the particles are charged and there is an applied electromagnetic field.

Before comparing Equations 79 and 80 with the present work, two detailed inconsistencies in the equations must be cleared up. First, Equation 80 is seen to contain a buoyancy force $-(1 - \epsilon)\rho_f g_i$ reacting on the particles. Physically this is a reaction of the pressure gradient in the fluid generated by the gravitational body force. However, the other body forces, F_{fi}, also generate pressure gradients in the fluid in the same way as gravity and, since the particles are unable to distinguish these from the gravitational pressure gradient, they should also give rise to buoyancy effects. Thus extra buoyancy terms should appear in Murray's Equation 80 if the body

force F_{fi} is nonvanishing. For comparison with the present work, however, gravity is the only relevant body force, and henceforth we shall set $F_{fi} = F_{si} = 0$, so that the above is of no importance.

A second, more important inconsistency can be seen by considering the first two terms on the right-hand side of Equation 79. As seen from Equation 81, $\partial p/\partial x_i$ and $\partial \sigma_{ik}/\partial x_k$ are two contributions to the divergence of the fluid stress tensor. But this divergence, $\partial E_{ik}/\partial x_k$, must always appear in the equations as a single entity. It is not possible to multiply the shear contribution $\partial \sigma_{ik}/\partial x_k$ by ϵ and the isotropic contribution $\partial p/\partial x_i$ by unity as in Equation 79; if $\partial E_{ik}/\partial x_k$ is to be multiplied by any factor, this same factor must multiply both $\partial p/\partial x_i$ and $\partial \sigma_{ik}/\partial x_k$. The difficulty could be eliminated either by multiplying $\partial p/\partial x_i$ by ϵ, or by omitting the factor ϵ which multiplies $\partial \sigma_{ik}/\partial x_k$ in Equation 79. If the second of these remedies is chosen, then to be consistent the factor $(1 - \epsilon)$ multiplying $\partial \sigma_{ik}^s/\partial x_k$ in Equation 80 should also be omitted. [We understand that Murray (1966) has himself modified his equations to eliminate these difficulties since his original publication.] Thus we can obtain two alternative forms—namely,

$$\rho_f \epsilon \left[\frac{\partial u_i}{\partial t} + u_k \frac{\partial u_i}{\partial x_k}\right] = \epsilon \frac{\partial E_{ik}}{\partial x_k} - A(u_i - v_i) - (1 - \epsilon) C \rho_f \frac{d}{dt}(u_i - v_i) + \rho_f g_i \quad (79')$$

and

$$\rho_s (1 - \epsilon) \left[\frac{\partial v_i}{\partial t} + v_k \frac{\partial v_i}{\partial x_k}\right] = (1 - \epsilon) \frac{\partial E_{ik}^s}{\partial x_k} + A(u_i - v_i) + (1 - \epsilon) C \rho_f \frac{d}{dt}(u_i - v_i) + (1 - \epsilon)(\rho_s - \rho_f) g_i \quad (80')$$

by introducing a factor ϵ in the term $\partial p/\partial x_i$, or

$$\rho_f \epsilon \left[\frac{\partial u_i}{\partial t} + u_k \frac{\partial u_i}{\partial x_k}\right] = \frac{\partial E_{ik}}{\partial x_k} - A(u_i - v_i) - (1 - \epsilon) C \rho_f \frac{d}{dt}(u_i - v_i) + \rho_f g_i \quad (79'')$$

and

$$\rho_s (1 - \epsilon) \left[\frac{\partial v_i}{\partial t} + v_k \frac{\partial v_i}{\partial x_k}\right] = \frac{\partial E_{ik}^s}{\partial x_k} + A(u_i - v_i) + (1 - \epsilon) C \rho_f \frac{d}{dt}(u_i - v_i) + (1 - \epsilon)(\rho_s - \rho_f) g_i \quad (80'')$$

by the alternative procedure.

For the purpose of comparing these with our own equations we assume that the Reynolds stresses we have discussed are effectively included in Murray's stress tensors, so that E_{ik} and E_{ik}^s may be replaced by $\mathcal{E}_{ik}$ and $\mathcal{E}_{ik}^s$ in Equations 79′, 80′, 79″, and 80″. Then it is not difficult to see that there is no possible choice for the form of the term nf_i, which would reduce our basic equations of motion Equations 40 and 51 to Equations 79′ and 80′; in other words, the latter equations are not consistent with Newton's third law of motion. However, Equations 79″ and 80″ can be obtained from Equations 40 and 51 by taking

$$nf_i = A(u_i - v_i) + (1 - \epsilon) C \rho_f \frac{d}{dt}(u_i - v_i) - (1 - \epsilon)\rho_f g_i \quad (83)$$

so the second "rationalized" form of Murray's equations is mechanically consistent, provided an interaction force of the form of Equation 83 is chosen. But Equation 83 is seen to correspond to the form of Equation 54′ for the interaction force, and we have already explained in some detail why we regard this as less satisfactory than the form given by Equations 65 and 66, which leads to our equations of motion. Nevertheless, the difference between our equations and Equations 79″ and 80″ lies only in the choice of empirical terms, and for the present it must remain a matter of opinion which provides the better model of the system's behavior.

The equations proposed by Hinze (1962) differ more radically from those presented here for two reasons. First, instead of considering separately the resistances to shear in the particle assembly and in the fluid, a single rate of shear is constructed from the average velocity field

$$\mathbf{w} = \epsilon \mathbf{u} + (1 - \epsilon)\mathbf{v}$$

and the shear stress is set proportional to this. Thus a single shear viscosity coefficient characterizes the system. Secondly, a virtual mass term is introduced in the particle equation of motion without any corresponding term in the fluid equation. But the virtual mass effect is part of the fluid-particle interaction force and consequently, as pointed out by Murray (1965), must appear with opposite signs in the fluid and particle equations if Newton's third law is to be satisfied.

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Nomenclature

A = drag coefficient in Murray's form of the equations of motion
C = virtual mass coefficient
C^0 = virtual mass coefficient in Basset's equation
$\mathcal{D}_{ik}'$ = $\mathcal{E}_{ik} - E_{ik}'$
d_p = particle diameter
E_{ik} = fluid phase stress tensor
E_{ik}^s = particle phase stress tensor
$\mathcal{E}_{ik}$ = effective stress tensor for fluid phase
$\mathcal{E}_{ik}^s$ = effective stress tensor for particle phase
F_{fi} = body force per unit mass, other than gravity, for fluid phase
F_{si} = body force per unit mass, other than gravity, for particle phase
f_d = drag force experienced by single particle suspended in a fluid stream
f_i = local mean value of fluid-particle interaction force
f_{pi} = force experienced by particle p as a result of interaction with fluid
$\bar{f}_i$ = local mean value of $\bar{f}_{pi}$
$\bar{f}_{pi}$ = contribution to force experienced by particle p; defined by Equation 75
$g(r)$ = weighting function used in defining local averages
g_i = gravity force vector
n = number of particles per unit volume
$\mathbf{n}$, n_k = unit vector normal to a surface
p = fluid pressure
R_{ik} = tensor representing "Reynolds stresses" for fluid phase
r = radial distance
r_o = "radius" of weighting function
S_{ik} = tensor representing "Reynolds stresses" for particle phase
t = time
$\mathbf{u}$, u_k = velocity of fluid phase
V_∞ = space occupied by complete system
$V_{f\infty}$ = space occupied by fluid matter of system
$V_{s\infty}$ = space occupied by solid matter of system

$\mathbf{v}, v_k$ = velocity of solid phase
$\mathbf{v}_p, v_{pk}$ = velocity of center of mass of particle
X_{fi} = body force per unit mass for fluid phase
X_{si} = body force per unit mass for particle phase
$\mathbf{x}, x_k$ = position vector
$\mathbf{x}_p$ = position vector of a point of particle p
$\mathbf{y}, y_k$ = position vector of current point of integration
α = drag coefficient in equation defining fluid-particle interaction force
α^0 = drag coefficient in Basset's equation
β = drag coefficient measured experimentally for particle assembly
ϵ = voidage
κ = permeability factor in equations of Pigford and Baron
μ = viscosity of the fluid
ν = volume of one particle
ρ_f = density of fluid
ρ_s = density of solid
σ_{ik} = pure shear contribution to fluid phase stress tensor
σ_{ik}^s = pure shear contribution to particle phase stress tensor
ϕ_i = local mean value of particle-particle interaction force
ϕ_{pi} = force experienced by particle p as a result of collisions with other particles

Variables representing point properties of the fluid or solid are distinguished by a single prime.

Variables representing the difference between point properties and their local mean values are distinguished by a double prime.

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DETERMINATION OF RATE CONSTANTS FOR COMPLEX KINETICS MODELS

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A method of determining kinetic rate coefficients for complex reactions by user-oriented iterative methods is described. The differential equations representing the reaction system may be nonlinear in the dependent variable but must be linear in the coefficients. Direct integration of the model for a sequence of time steps permits the model coefficients to be estimated by maximizing the multivariate correlation coefficient, which is the same as minimizing the sum of the squares of the weighted deviations. Three weighting schemes are compared. Simulations studies and tests with actual data indicate that the proposed technique is effective.

RATE coefficients for models of chemical kinetics must be determined prior to the use of empirical data in analysis, simulation, and design. Innumerable ways have been suggested (Lapidus, 1961) to obtain the best estimates of these coefficients from experimental data for any proposed model. Large, high speed digital and hybrid computers have made it possible to utilize techniques which until recently would have been prohibitively tedious. The technique used below, although it must be executed on a digital (or hybrid) computer, is straightforward, is user-oriented, and appears to merit consideration for these types of problems.

It will be assumed that ideal, isothermal perfectly mixed batch (or plug flow) kinetics data are available. Extensions to other more physically complex cases may be possible, but experience with this simple case is first desired. Thus, the data can be represented by sets of ordinary nonlinear differential equations in time, which, however, are linear in the coefficients to be estimated. There are essentially six methods of estimating kinetic coefficients for this type of model from experimental data.

1. Analytical (exact or approximate) integration of the set of differential equations, and subsequent application of iterative nonlinear least-squares regression techniques (Blakemore and Hoerl, 1963; Kittrell *et al.*, 1966; Levenspiel, 1962; Rubin, 1963; Wright, 1964).

2a. Differentiation of the empirical data directly, and subsequent application of linear least-squares regression techniques (Garfinkel *et al.*, 1961; Levenspiel, 1962; Lindsay, 1962; Steiner and Schoenemann, 1965).

2b. Linear regression to fit the empirical data, differentiation of empirical regression equation, followed by linear regression to estimate the coefficients (Bak, 1963).

3a. Numerical integration of the set of differential equations using empirical data directly, followed by iterative nonlinear least-squares (Bak, 1963; Ball and Groenweghe, 1966; Cull and Brenner, 1961; Peterson, 1960, 1962).

3b. Linear regression to fit the empirical data, followed by numerical integration of the set of differential equations.

4. Trial and error search using analog computers to match the empirical data (Katarov and Lutsenko, 1965; Schrodt *et al.*, 1963).

5. Method of differential corrections (Howland and Vaillancourt, 1961).

6. Quasilinearization (Bellman and Kalaba, 1965; Box and Hunter, 1962).