## RHEOLOGICAL BEHAVIOR OF A HOMOGENEOUS SUSPENSION IN NONSTATIONARY

FLOW

## Yu. A. Buevich and V. G. Markov

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Forces and moments are calculated for the interaction between phases in the nonstationary flow of a moderately concentrated suspension of small spheres; the equations of motion are derived in closed form.

The behavior of suspended particles in a nonstationary flow of a viscous liquid is substantially different from that in a steady-state flow; there are various inertial forces related to the interaction between the particles and the medium (see [1, 2] for a discussion of these forces), and these forces have a marked influence on two-phase turbulent flows [3]. The forces are also important in many applications, particularly ones requiring acceleration of exchange processes. One example is that of a fluidized bed subject to pressure fluctuations or changes in flow rate of the fluidizing agent, while another is a granular bed fluidized by vibration, such as is used in chemical technology, power engineering, and so on. There are also nonstationary effects arising in the passage of acoustic and shock waves, which can accelerate coagulation in polydisperse media, which is important not only in accelerating the elimination of small gas bubbles from liquids or causing suspensions to sediment, but also in other artificial systems such as rocket-motor nozzles [4] and natural systems such as thunderclouds [5].

The nonstationary flow of a suspension is here considered by methods briefly presented elsewhere [6], which have been applied to stationary flows in [7]. For definiteness we assume that the suspension meets all the requirements stated in the latter; namely, the Reynolds number for an individual spherical particle of the dispersed phase is small, while external moments are absent, and the mean suspension is constant in space and time and is not too high; thus, we can neglect effects arising from particle overlap within the framework of the method of [6, 7]. This is quite reasonable [7] for moderately concentrated suspensions ( $\rho < 0.20-0.25$ ).

Also, it is found [8] that there is a frequency dependence of the effective viscosity and of the other rheological parameters, i.e., the values shown by the parameters in a harmonic flow are dependent on frequency. Consequently, the various dynamic quantities for a nonstationary flow will be functionals of the kinematic variables. This means that memory effects occur, i.e., the history of the motion affects the situation at a given moment, which is commonplace for many homogeneous non-Newtonian media [9]. The effects are important even for single particles, because they give the Basset component, namely, forces acting on a particle from the medium [10]. Analogous phenomena occur in nonstationary heat- and masstransfer processes in dispersed media [11].

We restrict consideration initially to monodisperse suspensions. The convective coordinate system  $\mathbf{x}$  is related to the center of the test particle, and the conservation equations for mass and momentum of the suspension as a whole and for the disperse phase take the following form subject to the assumptions of [6, 7]:

$$\nabla \mathbf{v} = 0, \quad d_0 \varepsilon \frac{\partial \mathbf{v}_0}{\partial t} + d_1 \rho \frac{\partial \mathbf{v}_1}{\partial t} = \nabla \sigma - d\nabla (\Phi + \Psi),$$

$$\nabla \mathbf{v}_1 = 0, \quad d_1 \rho \frac{\partial \mathbf{v}_1}{\partial t} = \mathbf{f} - d_1 \rho \nabla (\Phi + \Psi)$$
(1)

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(these readily yield the corresponding equations for the continuous phase). Here the mean density and speed of the suspension are

$$d = \varepsilon d_0 + \rho d_i, \quad \mathbf{v} = \varepsilon \mathbf{v}_0 + \rho \mathbf{v}_i; \tag{2}$$

the potential of the inertial forces in this coordinate system is

$$\Psi = \mathbf{x} \left\{ \frac{d^{(1)} \mathbf{c}_i}{dt} \right\}_{\mathbf{x}=0} \equiv \mathbf{x} \left\{ \left[ \frac{\partial}{\partial t} + (\mathbf{c}_i \nabla) \right] \mathbf{c}_i \right\}_{\mathbf{x}=0}, \qquad (3)$$

and the mean force f due to the interaction between the phases and the divergence of the tensor  $\sigma$  for the mean stresses take the following form [6, 7]:

$$\mathbf{f} = n \int_{\mathbf{x}=a} (\mathbf{n} \mathbf{\Sigma}) \, d\mathbf{x}, \ \nabla \boldsymbol{\sigma} = - \varepsilon \nabla p + \mu_0 \Delta \mathbf{v} + n \nabla \int_{\mathbf{x}=a} \mathbf{x} * (\mathbf{n} \mathbf{\Sigma}) \, d\mathbf{x} , \qquad (4)$$

where an asterisk denotes dyad multiplication of the vectors. Equations (1) and (3) become those of [7] for steady-state flows; the forms of (2) and (4) remain as before.

Note that the velocity and acceleration of the disperse phase at x = 0 are zero by definition. The orders of the spatial derivatives of the velocities for the two phases are identical and equal to  $|\mathbf{v}_0|/L$ , where L is the spatial scale of the motion. Therefore, at distances of the order of a from the test particle we have

$$|\mathbf{v}_1| \sim (a/L) |\mathbf{v}_0| \ll |\mathbf{v}_0| \,. \tag{5}$$

A priori we expect frequency dispersion in the rheological parameters, so it is convenient to use a Fourier transform applied to the variables, as in [11], which is equivalent to considering an oscillating harmonic flow with a fixed frequency  $\lambda$  and  $\partial/\partial t$  replaced by  $i\lambda$ . For simplicity, no distinction is drawn between the Fourier transforms of the various quantities and the originals.

The general method [6] gives us the Fourier transforms as

$$\mathbf{f} = k_1 (\mathbf{v}_0 - \mathbf{v}_1) + k_2 \Delta \mathbf{v}_0 + k_3 \nabla (\Phi + \Psi), \ \nabla \boldsymbol{\sigma} = -\nabla \boldsymbol{\rho} + k \Delta \mathbf{v}_0, \tag{6}$$

where k and k<sub>1</sub> are certain coefficients not known a priori, which may also be dependent on  $\lambda$ ; the form of (6), in general, is not known in advance and must be derived from very general arguments, in which one incorporates the linearity of the equations of motion and also the dependence on quantities differing in tensor dimensions that, in principle, may control **f** and  $\nabla \sigma$ . The exact form is verified and if necessary corrected subsequently by means of self-consistency conditions, which constitute specifications for the identity of (4) and (5).

On substituting (6) into (1), we see that the motion of the suspension can be simulated as that of a fictitious incompressible homogeneous liquid whose velocity is that of the continuous phase, while the viscosity (in this harmonic flow) is k. Then the inertial density is approximately equal to  $d_0\varepsilon$  by virtue of (5), while the gravitational density is d. The equation for the conservation of momentum of this liquid derived from (1) and (6) implies that

$$\lambda d_0 \sim |k|/L^2 \sim \mu/L^2. \tag{7}$$

Parameters k and  $k_1$  are derived from the self-consistency conditions, and explicit expressions are required for the stresses  $\Sigma$  appearing in the integrands of the surface integrals in (4). The latter can be derived from the solution for the flow around a test particle [6, 7]. Let  $\mathbf{v}_0^*$ ,  $\mathbf{v}_1^*$ , and  $\mathbf{p}^*$  be the distributions of the velocity and pressure as perturbed by the test particle, for which we have equations of the same type as (1) and (6); (5) still applies. We also use (7) and the fact that the spatial scale of the perturbed fields is  $\alpha$  to obtain

$$\lambda d_0 | \mathbf{v}_0^* | \sim \mu | \mathbf{v}_0^* | / L^2 \ll \mu | \Delta \mathbf{v}_0^* |, \ \lambda d_0 a^2 / \mu \ll 1, \tag{8}$$

which shows that the nonstationary term on the left-hand side of the momentum-conservation equation for the fictitious medium is a small quantity of higher order in the parameter  $\alpha/L$ . We restrict consideration to small quantities of second order in this parameter and use (5) to obtain the result for the flow around the test particle as

$$\nabla \mathbf{v}_0^* = 0, \quad i\lambda d_0 \varepsilon \mathbf{v}_0^* = -\nabla p^* + k\Delta \mathbf{v}_0^* - d\nabla (\Phi + \Psi),$$
  

$$\mathbf{v}_0^* = \mathbf{\omega} \times \mathbf{x} \, (x = a), \quad \mathbf{v}_0^*, \, p^* \to \mathbf{v}_0, \, p \, (x \to \infty),$$
(9)

where the unperturbed fields  $\mathbf{v}_0$  and p satisfy equations following from (1) and (6). The problem of (9) is a particular case of that examined in [8]. We determine the stresses in the flow of the fictitious medium, which is defined by (9), in standard form for a viscous liquid [10] and then calculate the integrals in (4) via the solution of [8] to obtain

$$\mathbf{f} = \rho d\nabla (\Phi + \Psi) + \frac{9}{2} \frac{\rho}{a^2} k \left( 1 + \beta + \frac{1}{3} \beta^2 \right) (\mathbf{v}_0 - \mathbf{v}_1) + \frac{9}{2} \frac{\rho}{\beta^2} k \left[ e^\beta - \left( 1 + \beta + \frac{1}{3} \beta^2 \right) \right] \Delta \mathbf{v}_0 , \qquad (10)$$

$$\nabla \boldsymbol{\sigma} = -\nabla p + \mu_0 \Delta \mathbf{v} + \frac{5\rho k}{2(1+\beta)} \left( 1 + \beta + \frac{2}{5} \beta^2 + \frac{1}{15} \beta^3 \right) \Delta \mathbf{v}_0 + \frac{1}{2} \left( 1 + \beta + \frac{1}{3} \beta^2 \right) \Delta \mathbf{v}_0 + \frac{1}{3} \left( 1 + \beta + \frac{2}{3} \beta^2 + \frac{1}{3} \beta^3 \right) \Delta \mathbf{v}_0 + \frac{1}{3} \left( 1 + \beta + \frac{1}{3} \beta^2 \right) \left( 1 + \beta + \frac{1}{3} \beta^2 \right) \Delta \mathbf{v}_0 + \frac{1}{3} \left( 1 + \beta + \frac{1}{3} \beta^2 \right) \left( 1 + \beta + \frac{1}{3} \beta^2 \right) \Delta \mathbf{v}_0 + \frac{1}{3} \left( 1 + \beta + \frac{1}{3} \beta^2 \right) \right) \left( 1 + \beta + \frac{1}{3} \beta^2 \right) \right) \left( 1 + \beta + \frac{1}{3} \beta^2 \right) \right) \left( 1 + \beta + \frac{1}{3} \beta^2 \right) \left( 1 + \beta + \frac{1}{3} \beta^2 \right) \left( 1 + \beta + \frac{1}{3} \beta^2 \right) \right) \left( 1 + \beta + \frac{1}{3} \beta^2 \right) \left( 1 + \beta + \frac{1}{3} \beta^2 \right) \right) \left( 1 + \beta + \frac{1}{3} \beta^2 \right) \right) \left( 1 + \beta + \frac{1}{3} \beta^2 \right) \right) \left( 1 + \beta + \frac{1}{3} \beta^2 \right) \left( 1 + \beta + \frac{1}{3} \beta^2 \right) \left( 1 + \beta + \frac{1}{3} \beta^2 \right) \right) \left( 1 + \beta + \frac{1}{3} \beta^2 \right) \right) \left( 1 + \beta + \frac{1}{3} \beta^2 \right) \left( 1 + \beta + \frac{1}{3} \beta^2 \right) \left( 1 + \beta + \frac{1}{3} \beta^2 \right) \left( 1 + \beta + \frac{1}{3}$$

 $+\frac{5\rho a^{2}k}{\beta^{2}(1+\beta)}\left[e^{\beta}-\left(1+\beta+\frac{2}{5}\beta^{2}+\frac{1}{15}\beta^{3}\right)\right]\Delta^{2}\mathbf{v}_{0},$ (11)

where the parameter is

$$\beta^2 = i\lambda d_0 \epsilon a^2/k. \tag{12}$$

The equation for the conservation of momentum of the disperse phase gives us from (1) by means of (5) and the expression for f in (6) that

$$\mathbf{v}_{i} = \mathbf{v}_{0} + \frac{k_{2}}{k_{i}} \Delta \mathbf{v}_{0} + \frac{k_{3} - d_{i}\rho}{k_{i}} \nabla (\Phi + \Psi).$$
(13)

Since  $\Delta(\nabla \Psi) = 0$  by virtue of the definition of  $\Psi$  in (3), we assume for simplicity that  $\Delta(\nabla \Phi) = 0$  (although all the results are readily generalized to the case where this is not so), and we use  $\Delta[-\nabla p - d\nabla(\Phi + \Psi)] = 0$ , which follows directly from (1) and (6), to get from (2) and (13) that

$$\Delta^2 \mathbf{v}_0 = \frac{\beta^2}{a^2} \Delta \mathbf{v}_0, \ \Delta \mathbf{v} = \left(1 + \rho \frac{\beta^2}{a^2} \frac{k_2}{k_1}\right) \Delta \mathbf{v}_0. \tag{14}$$

Up to terms of the second order in  $\alpha/L$  inclusive [or up to terms of the first order in  $|\beta|^2$ , which is the same by virtue of (7) and (8)], we have that  $k_2/k_1$  in (14) should be expressed up to terms of zero order in this parameter, which can be done by means of the results of [7] (see also below). We have  $k_2/k_1 \approx \alpha^2/6$ , so the expressions for  $\forall \sigma$  in (6) and (11) give us with (14) that

$$k\left(1-\frac{5}{2}\rho\frac{e^{\beta}}{1+\beta}\right)=\mu_0\left(1+\frac{1}{6}\rho\beta^2\right).$$
(15)

We use (12) to obtain with the required accuracy that

$$k = \mu + i\lambda T, \quad \mu = \frac{\mu_0}{1 - 2.5\rho}, \quad T = \rho \varepsilon \left[\frac{1}{6} + \frac{5}{4(1 - 2.5\rho)}\right] a^2 d_0, \tag{16}$$

where  $\mu$  is the effective viscosity of the suspension in a state of steady flow, which has been calculated [7], while the term containing T denotes the frequency dispersion of the viscosity.

We now compare the expressions for f in (6) and (10) and use (12) and (16) to obtain the following representations for the ki:

$$k_{1} = \frac{9}{2} \rho K_{1}(\rho) \frac{\mu_{0}}{a^{2}} + \frac{9}{2} \rho K_{2}(\rho) \sqrt{i\lambda d_{0}a^{2}\mu_{0}} + \frac{3}{2} \rho K_{3}(\rho)i\lambda d_{0}, \quad k_{2} = \frac{3}{4} \rho K_{1}(\rho) \mu_{0}, \quad k_{3} = \rho d, \quad (17)$$

$$K_{1}(\rho) = \frac{1}{1 - 2.5\rho}, \quad K_{2}(\rho) = \left(\frac{1 - \rho}{1 - 2.5\rho}\right)^{1/2}, \quad K_{3}(\rho) = (1 - \rho) \left[1 + \rho \left(\frac{1}{2} + \frac{15}{4(1 - 2.5\rho)}\right)\right]. \quad (18)$$

The functions  $K_i(\rho)$  describe the effects of the hindered flow on the various components of the force on the particles, and they all become unity as  $\rho \rightarrow 0$ ; also, (17) coincide with the formulas of [7] as  $\lambda \rightarrow 0$ .

We convert from the Fourier transforms in (17) to the originals by means of the techniques of [10] and use the laboratory coordinate system instead of the convective one to obtain the following representation for the phase-interaction forces referred to the particles in unit volume:

$$\mathbf{f} = \frac{9}{2} \rho K_1(\rho) \frac{\mu_0}{a^2} (\mathbf{c}_0 - \mathbf{c}_1) + \frac{9}{2} \rho K_2(\rho) \left(\frac{\mu_0 d_0}{\pi a^2}\right)^{1/2} \int_{-\infty}^{\infty} \left(\frac{\partial}{\partial \tau} + \mathbf{c}_1 \nabla\right) (\mathbf{c}_0 - \mathbf{c}_1) \frac{d\tau}{\sqrt{t - \tau}} + \frac{3}{2} \rho K_3(\rho) d_0 \left(\frac{\partial}{\partial t} + \mathbf{c}_1 \nabla\right) (\mathbf{c}_0 - \mathbf{c}_1) + \frac{3}{4} \rho K_1(\rho) \mu_0 \Delta \mathbf{c}_0 + \rho d \nabla \left[\Phi + \mathbf{r} \left(\frac{\partial}{\partial t} + \mathbf{c}_1 \nabla\right) \mathbf{c}_1\right].$$
(19)

The first term here clearly describes the effective viscous force, while the second corresponds to the Basset force in hindered flow, which is dependent on the previous history; the third represents the effective force arising from the acceleration of the interphase slip and, in particular, it is related to the adjoint mass; the fourth term corresponds to the Faxen force, and the fifth describes the effective buoyancy, which is due in part to the external mass forces and in part to the inertial forces of (3). Finally, (19) becomes a standard expression long known for dilute suspensions for single particles as  $\rho \neq 0$ .

It is of interest to compare (19) with results [10, 12] for a single particle in the nonviscous approximation in the absence of external mass forces; the force  $\mathbf{F} = \mathbf{f}/\mathbf{n}$  acting on a single particle is found from (19) with  $\rho = 0$ ,  $\mu_0 = 0$ , and  $\Phi = 0$  as given by the equation

$$M \frac{d\mathbf{u}}{dt} = -\frac{1}{2} M_0 \left( \frac{d\mathbf{u}}{dt} - \frac{d\mathbf{c}}{dt} \right) + M_0 \frac{d\mathbf{c}}{dt} , \qquad (20)$$

where  $\mathbf{u}$  and M are the velocity and mass of a particle, while  $\mathbf{c}$  and M<sub>o</sub> are the velocity mass of the liquid in the particle volume, which is precisely the equation given in [10, 12].

We now write the equation of motion in the laboratory coordinate system  $\mathbf{r}$ ; (7) shows that allowance for the frequency dispersion of the viscosity is unnecessary, because this would represent exceeding the available accuracy. Therefore, simple transformation of (1) gives us the equation for conservation of the momentum of the suspension as

 $d_{0}\varepsilon\left(\frac{\partial}{\partial t}+\mathbf{c}_{0}\nabla\right)\mathbf{c}_{0}+d_{1}\rho\left(\frac{\partial}{\partial t}+\mathbf{c}_{1}\nabla\right)\mathbf{c}_{1}=-\nabla p+\mu\Delta\mathbf{c}_{0}-d\nabla\Phi.$ (21)

The equation for the conservation of mass that supplements (21) is of the form  $\nabla c_0 = 0$  for a macroscopically homogeneous suspension whose concentration is not dependent on time; however, it is readily shown that the above results, in particular, (21), are correct also in the case where  $\rho$  is dependent on time if the equation for the conservation of mass for the continuous phase takes the form

$$\frac{\partial \varepsilon}{\partial t} + \varepsilon_{\nabla} \mathbf{c}_{\mathbf{0}} = 0. \tag{22}$$

The equations for the conservation of momentum and mass for the dispersed phase can be put as follows:

$$\frac{\partial \rho}{\partial t} + \rho_{\nabla} \mathbf{c}_{i} = 0, \ d_{i} \rho \left( \frac{\partial}{\partial t} + \mathbf{c}_{i} \nabla \right) \mathbf{c}_{i} = \mathbf{f} - d_{i} \rho_{\nabla} \Phi, \tag{23}$$

where f is defined by (19); the unknown variables in (21)-(23) are  $\rho$ , p, c<sub>0</sub>, and c<sub>1</sub>.

It is more difficult to generalize the results to inhomogeneous systems on account of the occurrence of the additional vector  $\nabla \rho$ , which characterizes the situation at some point in the flow and which can also occur in (4). However, if we neglect the additional terms proportional to  $\nabla \rho$  in (4), we can still write the equations readily for the case where  $\rho$  is dependent on time and on the coordinates [6].

In principle, it is possible to consider components of f and  $\forall \sigma$  of higher orders in a/L; the main difficulty then is that we cannot use (5), which simplifies the calculations considerably. The result would be equations of motion that contain time derivatives of order higher than the first, which describe various relaxation effects (such equations have been derived for heat and mass transfer [11]). However, the desirability of such a generalization is, at present, very doubtful, not only because of the specific features of the motion in a two-speed disperse medium, but also because of the uncertainty arising in using asymptotically correct equations to describe oscillating flows even for single particles [13].

We now determine the moment resulting from the phase interaction; we use the results of [8] to obtain that

$$\mathbf{m} = 6\rho K_1(\rho) \,\mu_0\left(\frac{1}{2} \operatorname{rot} \mathbf{c}_0 - \boldsymbol{\omega}\right) + 6\rho a^2 d_0 \frac{d^{(1)}}{dt} \left(G_1(\rho) \frac{1}{2} \operatorname{rot} \mathbf{c}_0 - G_2(\rho) \,\boldsymbol{\omega}\right), \tag{24}$$

$$G_{1}(\rho) = (1-\rho) \left[ \frac{1}{2} + \rho \left( \frac{1}{6} + \frac{5}{4(1-2.5\rho)} \right) \right],$$

$$G_{2}(\rho) = (1-\rho) \left[ \frac{1}{3} + \rho \left( \frac{1}{6} + \frac{5}{4(1-2.5\rho)} \right) \right].$$
(25)

This shows that adjoint-mass effects are also important in particle rotation. The adjointmass coefficients are determined in relation to the rotation of the particle by the external flow or in terms of the inherent rotation; the two differ somewhat.

In conclusion, we note that all of these results are readily transferred to polydisperse suspensions. This generalization has been considered in detail elsewhere [7], so it is not repeated here.

## NOTATION

 $\alpha$ , particle radius; c, velocity in laboratory coordinate system r; d, density; f, phaseinteraction force; G<sub>i</sub>, functions defined in (25); K<sub>i</sub>, functions defined in (18); k, k<sub>i</sub>, coefficients in (4), M, M<sub>o</sub>, mass of a particle and mass of liquid in particle volume; m, phase-

interaction moment; n, numerical concentration of particles; n, unit vector of external normal; p, pressure; u, particle velocity; v, velocity in the convective coordinate system x linked to the center of a test particle;  $\beta$ , parameter in (12);  $\varepsilon = 1 - \rho$ ;  $\mu$ , viscosity;  $\rho$ , volume concentration of suspension;  $\sigma$ ,  $\Sigma$ , stress tensors;  $\Phi$ , potential of external mass forces;  $\Psi$ , inertial force potential defined in (3);  $\omega$ , angular velocity of particle rotation. Indices: 0, 1, continuous and dispersed phases, respectively; \*, fields perturbed by a test particle.

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