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Modeling of Concentrated Suspensions

B. H. A. A. van den Brule[1] and R. J. J. Jongschaap[2]

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The constitutive equation of a concentrated suspension of spherical particles in a Newtonian medium is derived. To this end the method of local volume averaging is employed. To calculate the contribution of the particles to the stress tensor it is assumed that the stress generated in the interstitial holes between the particles is negligible compared to the stress generated in the narrow gaps separating the particles. The use of the resulting expression is demonstrated with two examples on a cubical arrangement of particles: pure shear and simple shear. Furthermore, the validity of the lubrication approximation employed in this work is checked against the results derived by Nunan and Keller for periodic suspensions.

KEY WORDS: Concentrated suspension; volume averaging; viscosity; stress tensor; lubrication approximation.

1. INTRODUCTION

Many models and empirical expressions have been developed to describe the relationship between the particle concentration and the macroscopic mechanical properties of suspensions. The most successful theories are limited to dilute suspensions of particles in a Newtonian medium.[1] In this concentration regime we do not have to take the hydrodynamic interactions between the particles into account. At higher concentrations multiple-particle interactions become important. This problem is very hard to solve. At even higher concentrations, where the particles nearly touch each other, we still have multiple-particle interactions, but the hydrodynamic screening will be effective enough to allow us to assume that the interactions are pairwise additive. (The interaction of a reference particle with one of its

[1] Philips Research Laboratories, P.O. Box 80000, 5600JA, Eindhoven, The Netherlands.
[2] Department of Applied Physics, Twente University, P.O. Box 217, 7500 AE, Enschede, The Netherlands.

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neighbors is not disturbed by the interactions of the reference particle with the other neighboring particles.) A successful theory in the high-concentration regime is that developed by Frankel and Acrivos.[2] The basic idea of this theory is to equate the ratio between the energy dissipation in the suspension and in the homogeneous fluid to the relative viscosity of the suspension. To calculate the energy dissipation in the suspension, the packing geometry must be specified. Furthermore, in this theory it is assumed that the viscous dissipation of energy occurs primarily in the narrow gaps between the particles.

In this paper instead of considering the energy dissipation, an expression for the stress tensor of a concentrated suspension will be derived. In Section 3 this expression will be used to calculate the same situation as the one considered by Frankel and Acrivos; this is the case of a pure shearing deformation of a cubic configuration of particles. The advantage of using the stress tensor instead of the energy dissipation is that it provides more information. This will be demonstrated in Section 4, where the situation of simple shear flow will be considered. In Section 5 the lubrication approximation, which is used throughout this paper to calculate the hydrodynamic forces, will be checked against the theoretical results obtained by Nunan and Keller[3] for periodic suspensions.

2. THE VOLUME-AVERAGED STRESS TENSOR

The macroscopic properties of a suspension are dependent only on the microscopic structure of the system in a statistical sense. If a length scale L is present which is small compared to the macroscopic dimensions of the system, but at the same time is large with respect to the characteristic length scale of the microstructure, it is possible to use the method of local volume averaging[4] to obtain the macroscopic properties of a suspension. The volume-averaged value of a locally defined quantity $Q(\mathbf{r}, t)$, indicated by an overbar, is defined as

$$\bar{Q}(\mathbf{x}, t) = \frac{1}{V} \int_{V(\mathbf{x})} Q(\mathbf{r}, t)\, d^3\mathbf{r} \tag{1}$$

In this expression V represents a reference volume with a typical dimension L. The volume V has a fixed position and orientation with respect to the position $\mathbf{x}$. Throughout this paper it is assumed that the volume-averaged value of a certain property Q equals the macroscopically observable value of Q.

The bulk stress tensor $\bar{\mathbf{T}}$ of a suspension of rigid particles in a Newtonian medium with viscosity η is given by Batchelor,[5]

$$\bar{\mathbf{T}} = -\bar{p}\mathbf{1} + 2\eta\bar{\mathbf{D}} + \frac{1}{V}\sum_i \int_{\partial V_{p,i}} \mathbf{T}\cdot\mathbf{n}(\mathbf{r}-\mathbf{r}_i)\, d^2\mathbf{r} \tag{2}$$

where $\mathbf{T}$ is the local stress tensor. In the expression above, the summation is performed over all particles i contained in the reference volume V. The surface of the ith particle is denoted by $\partial V_{p,i}$. The vector $\mathbf{r}_i$ indicates the center of the ith particle and $\mathbf{n}$ denotes the normal vector. The bulk rate of strain tensor, which is observed macroscopically, is denoted by $\bar{\mathbf{D}}$.

To be able to calculate the integral, the stress distribution in the fluid surrounding the particles must be known. In a very dilute suspension the flow field and the stress distribution around a spherical particle can be calculated exactly. In the case of a concentrated suspension this is not possible. One way to overcome this difficulty is to assume that for a very concentrated suspension in which the particles nearly touch each other, the stress generated in the fluid in the interstitial holes between the particles is negligible compared to the stress generated in the narrow gaps separating the particles. The particle contribution to the stress tensor $\bar{\mathbf{T}}_p$ thus becomes

$$\bar{\mathbf{T}}_p = \frac{1}{V}\sum_i \sum_j \int_{A_{ij}} \mathbf{T}\cdot\mathbf{n}(\mathbf{r}-\mathbf{r}_i)\, d^2\mathbf{r} \tag{3}$$

where A_{ij} is the surface of the "gap zone" between the ith and the jth particle. For nearly touching particles the vector $\mathbf{r}-\mathbf{r}_i$, indicating a point in the gap A_{ij}, may be approximated by $\frac{1}{2}(\mathbf{r}_j-\mathbf{r}_i)=\frac{1}{2}\mathbf{q}_{ij}$. The hydrodynamic force exerted by the fluid on the surface A_{ij} is given by

$$\mathbf{f}_{ij} = \int_{A_{ij}} \mathbf{T}\cdot\mathbf{n}\, d^2\mathbf{r} \tag{4}$$

With these approximations the expression for $\bar{\mathbf{T}}_p$ becomes

$$\bar{\mathbf{T}}_p = \frac{1}{2V}\sum_i \sum_j \mathbf{f}_{ij}\mathbf{q}_{ij} \tag{5}$$

A similar expression has been derived by Goddard.[6] Combining (2) and (5) yields the expression for the stress tensor of a concentrated suspension of particles in a Newtonian fluid

$$\bar{\mathbf{T}} = -\bar{p}\mathbf{1} + 2\eta\bar{\mathbf{D}} + \frac{1}{2V}\sum_i \sum_j \mathbf{f}_{ij}\mathbf{q}_{ij} \tag{6}$$

This result can also be expressed in a different way. Since V contains many particles, the result of the summation in (6) is equal to N times the average contribution of a typical particle,

$$\bar{\mathbf{T}}_p = \frac{1}{2V} \sum_i \sum_j \mathbf{f}_{ij} \mathbf{q}_{ij} = \frac{N}{2V} \left\langle \sum_j \mathbf{f}_{0j} \mathbf{q}_{0j} \right\rangle \tag{7}$$

where the index 0 refers to the test particle, and N equals the number of particles contained in V. The angular brackets denote an averaging over all possible realizations of the surrounding particles relative to the test particle. If the forces can be considered as a function of the configuration $Q = \{\mathbf{q}_{01}, \mathbf{q}_{02}, \ldots, \mathbf{q}_{0j}, \ldots\}$ only, then (7) can be written as

$$\bar{\mathbf{T}}_p = \tfrac{1}{2} n \int_Q \sum_j \mathbf{f}_{0j} \mathbf{q}_{0j} \, P(Q) \, dQ \tag{8}$$

where n is the number density of particles and $P(Q)\, d(Q)$ is the probability of finding the surrounding particles in a configuration Q. Only the particles which are relatively close to the test particle contribute to the summation in (8). To restrict the calculation to these particles, a volume V_0 surrounding the test particle is introduced. Furthermore, a vector field $\mathbf{f}$ with the following property is introduced:

$$\mathbf{f}_{0j} = \int \delta(\mathbf{q}_{0j} - \mathbf{q}) \mathbf{f} \, d^3\mathbf{q} \tag{9}$$

Thus (8) becomes

$$\bar{\mathbf{T}}_p = \tfrac{1}{2} n \int_Q P(Q) \int_{V_0} \sum_j \delta(\mathbf{q}_{0j} - \mathbf{q}) \mathbf{f}\mathbf{q} \, d^3\mathbf{q} \, d(Q) \tag{10}$$

Changing the order of integration in (10) yields

$$\bar{\mathbf{T}}_p = \tfrac{1}{2} n \int_{V_0} \mathbf{f}\mathbf{q} \int_Q \sum_j \delta(\mathbf{q}_{0j} - \mathbf{q}) \, P(Q) \, dQ \, d^3\mathbf{q} \tag{11}$$

The integral over Q equals the pair distribution function $g(\mathbf{q})$, which expresses the probability density of finding a particle at a position $\mathbf{q}$ relative to the test particle. Substitution of (11) in the expression for the stress tensor yields

$$\bar{\mathbf{T}} = -\bar{p}\mathbf{1} + 2\eta \bar{\mathbf{D}} + \tfrac{1}{2} n \int_{V_0} g(\mathbf{q}) \mathbf{f}\mathbf{q} \, d^3\mathbf{q} \tag{12}$$

To be able to derive an explicit expression for the stress tensor, some additional assumptions have to be made with respect to the interaction forces and to the pair distribution function. For the special case that the instantaneous positions of the neighboring particles are known exactly, (12) reduces to

$$\bar{\mathbf{T}} = -\bar{p}\mathbf{1} + 2\eta\bar{\mathbf{D}} + \tfrac{1}{2}n\sum \mathbf{fq} \tag{13}$$

where the summation is performed over the nearest neighbors.

3. COMPARISON WITH THE THEORY OF FRANKEL AND ACRIVOS

Frankel and Acrivos calculated the viscosity of a concentrated suspension of inert spheres in a Newtonian medium using a cell model. In their derivation they made the following assumptions:

1. The force between two neighboring particles may be determined using the lubrication approximation. Furthermore, it is assumed that the force caused by the relative shearing motion between the particles is negligible compared to the force caused by the relative squeezing motion. This force is given by

$$\mathbf{f} = \frac{3\pi\eta a^2}{2h}\mathbf{u}_{||} \tag{14}$$

where $\mathbf{u}_{||}$ is the projection of the relative velocity $\mathbf{u}$ of the particles upon the line of centers, a is the radius of the particles, and h is the face-to-face distance between the particles.

2. The particles move affinely. In this case the relative velocity between two particles is equal to

$$\mathbf{u} = q\bar{\mathbf{L}}\cdot\mathbf{e} \tag{15}$$

where $\bar{\mathbf{L}}$ is the macroscopic velocity gradient and $\mathbf{e}$ is the unit vector in the direction of $\mathbf{q}$. The projection upon the line of centers thus becomes

$$\mathbf{u}_{||} = (\mathbf{u}\cdot\mathbf{e})\mathbf{e} = q(\bar{\mathbf{L}}:\mathbf{ee})\mathbf{e} = q(\bar{\mathbf{D}}:\mathbf{ee})\mathbf{e} \tag{16}$$

3. The particles are arranged in a simple cubical packing. The orientation of the packing is aligned with the principal axes of the rate of strain tensor. This effectively means that a pure shearing deformation of a cubical arrangement of particles is considered. See Fig. 1.

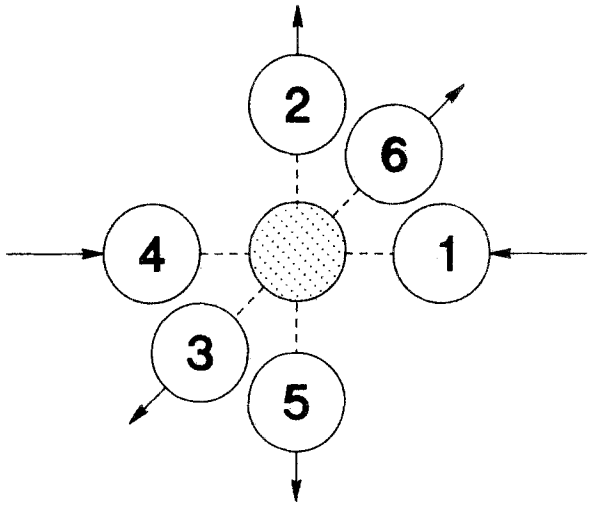

Fig. 1. A simple cubical arrangement of particles aligned with the principal axes of the rate-of-strain tensor. Particles 1 and 4 approach the reference particle, whereas the other particles move away from the reference particle.

With these assumptions the stress tensor given by (12) can be calculated. The size of the volume V_0 is such that only the interactions with the nearest neighbors are taken into account. Within the volume V_0 the pair distribution function is zero everywhere except at the six positions on the axes of the rate of strain tensor where the probability of finding a particle is equal to one. The contribution of a particle at a position $\mathbf{q}$ is found from (14) and (16),

$$\mathbf{fq} = \frac{3\pi\eta a^2 q^2}{2h}(\bar{\mathbf{D}}:\mathbf{ee})\mathbf{ee} \tag{17}$$

For particle "1" in Fig.1 this expression reduces to

$$\begin{aligned}\mathbf{f}_1\mathbf{q}_1 &= \frac{3\pi\eta a^2 q^2}{2h}(\bar{D}_{ij}\boldsymbol{\delta}_i\boldsymbol{\delta}_j:\mathbf{e}_1\mathbf{e}_1)\mathbf{e}_1\mathbf{e}_1 \\ &= \frac{3\pi\eta a^2 q^2}{2h}\bar{D}_{ij}(\boldsymbol{\delta}_i\cdot\mathbf{e}_1)(\boldsymbol{\delta}_j\cdot\mathbf{e}_1)\mathbf{e}_1\mathbf{e}_1 \\ &= \frac{3\pi\eta a^2 q^2}{2h}\bar{D}_{11}\mathbf{e}_1\mathbf{e}_1\end{aligned} \tag{18}$$

In the expression above the vectors $\boldsymbol{\delta}_i$ $(i = 1, 2, 3)$ denote the base vectors of the coordinate system. In an analogous way the contributions of the

other neighboring particles are obtained. Adding the contributions of the six nearest neighbors and substitution of the result into (13) yields the following expression for the stress tensor:

$$\bar{\mathbf{T}} = -\bar{p}\mathbf{1} + 2\eta\left(1 + \frac{3\pi n a^2 q^2}{4h}\right)\bar{\mathbf{D}} \tag{19}$$

It has to be noted here that the fact that a Newtonian fluid is obtained is a consequence of the symmetry of the particle packing with respect to the principal directions of the rate of strain tensor. The value of the viscosity depends on the type of symmetry assumed. In the limiting case of nearly touching particles, (19) with $n = 1/q^3$ reduces to

$$\bar{\mathbf{T}} = -\bar{p}\mathbf{1} + 2\eta\left(1 + \frac{3\pi a}{8h}\right)\bar{\mathbf{D}} \tag{20}$$

It is convenient to express the quantity a/h in terms of the concentration of the suspension

$$\frac{a}{h} = \frac{1}{2}\left(\frac{1-\varepsilon}{\varepsilon}\right), \qquad \varepsilon = 1 - \left(\frac{\phi}{\phi_m}\right)^{1/3} \tag{21}$$

where ϕ_m is the maximum obtainable concentration. From (20) and (21) it can be seen that the relative viscosity of the suspension is given by

$$\eta_R = 1 + \frac{3\pi}{16}\frac{1-\varepsilon}{\varepsilon} \tag{22}$$

Except for the first term, which is negligible at high concentrations, this result is also obtained in the Frankel and Acrivos theory if one chooses a cubical instead of a spherical cell. Frankel and Acrivos chose a spherical cell since this increases the value of the constant from $3\pi/16$ to $9/8$, which brings the theoretical predictions in closer agreement with experiments. However, the choice of a spherical cell is debatable, since in this case the energy dissipation in the homogeneous fluid is only partially taken into account. Moreover, there is the problem that the experimental data are fitted with a value of $\phi_m = 0.625$, whereas the maximum obtainable concentration for a cubical packing is only $\pi/6$.

If instead of a cubical arrangement the particles are assumed to be in a hexagonal arrangement, then the value of the relative viscosity, calculated from (13), is equal to

$$\eta_R = \frac{3\sqrt{2}\,\pi}{16}\frac{1-\varepsilon}{\varepsilon} \tag{23}$$

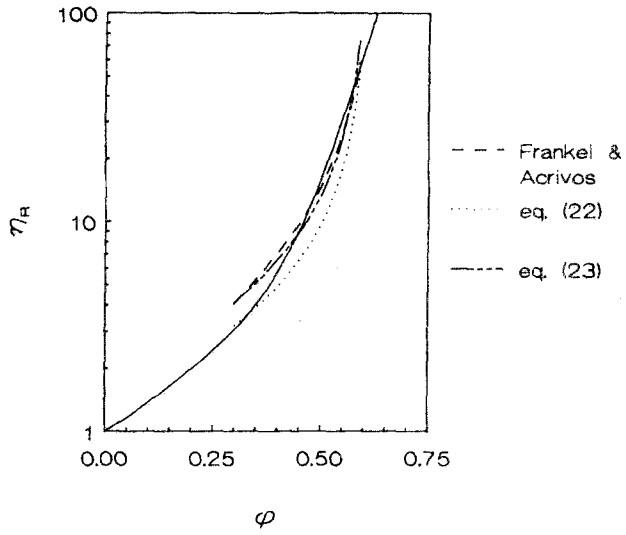

Fig. 2. Comparison of the theoretical predictions with experiments. The solid line represents the empirical curve of Thomas. The Frankel and Acrivos formula is evaluated with $\phi_m = 0.625$, whereas Eqs. (20) and (21) are evaluated with a value $\phi_m = 0.61$.

In this case the experiments can be fitted with a value $\phi_m = 0.61$, which is indeed lower than the maximum obtainable concentration in a hexagonal packing. In Fig. 2 the theoretical predictions are compared with an experimental curve fit of Thomas.[7] It can be seen that the theoretical predictions are in good agreement with the experiments.

4. LAYERED FLOW OF A CONCENTRATED SUSPENSION

The advantage of using the expression for the stress tensor instead of calculating the energy dissipation becomes especially clear when the case of layered flow of a suspension is considered. The requirement for the stress tensor to be symmetrical provides the extra information needed to calculate the angular velocity of the particles. A layered structure conform the observations of Hoffman[9] has been analyzed in a previous paper.[8] In the present paper a layered flow of a cubical arrangement of particles will be considered. See Fig. 3. Of course, the cubical arrangement of the particles will be distorted by the flow. Therefore, the value of the viscosity which will be derived here will be valid only at the moment that the packing is cubical. The instantaneous value of the viscosity will be a periodic function of time. In principle, the effective value of the viscosity can be obtained by averaging the viscosity over all subsequent configurations through which the

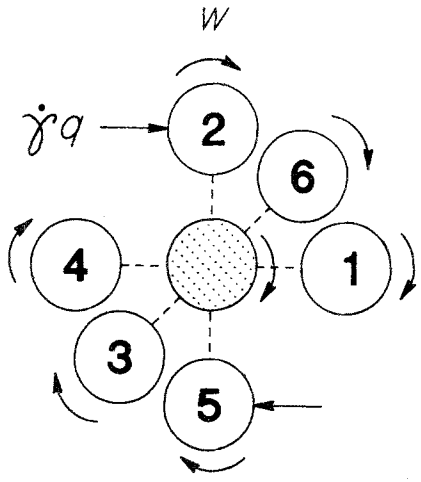

Fig. 3. A simple cubical arrangement of particles in simple shear flow. Note that there is no relative velocity between particles 3 and 6 and the reference particle.

lattice passes. (This has been done in the analysis of the layered structure as suggested by Hoffman.)

Since the squeezing motion of the particles just vanishes at the instant that the lattice becomes cubical it is no longer permissible to neglect the forces which are related to the relative shearing motion of the particles and due to particle rotation. Using the lubrication approximation, the force due to the shearing motion between two particles can be found to be[2]

$$\mathbf{f}_{\perp} = \frac{\pi n}{2}\left(-2a + q \ln \frac{q}{h}\right)\mathbf{u}_{\perp}, \qquad \frac{h}{a} \to 0 \tag{24}$$

where $\mathbf{u}_{\perp}$ is the component of the relative velocity perpendicular to the line joining the centers of the particles. To calculate the force caused by the rotation of the particles $\mathbf{f}_{w}$, only the component of the angular velocity $\mathbf{w}$ which is perpendicular to the line joining the centers has to be taken into account. This component is given by

$$\mathbf{w}_{\perp} = \mathbf{w} \cdot (\mathbf{1} - \mathbf{ee}) \tag{25}$$

The force $\mathbf{f}_{w}$ is calculated in an analogous way to $\mathbf{f}_{\perp}$,

$$\mathbf{f}_{w} = \pi\eta\left(-2a + q \ln \frac{q}{h}\right) w_{\perp} a \frac{\mathbf{q} \times \mathbf{w}_{\perp}}{|\mathbf{q} \times \mathbf{w}_{\perp}|} \tag{26}$$

The contribution of particle 2 in Fig. 3 to the stress tensor is calculated as follows:

$$\text{shearing motion:} \quad (\mathbf{f}_1\,\mathbf{q})_2 = \frac{\pi\eta}{2}\left(-2a + q\ln\frac{q}{h}\right)\dot{\gamma}q^2\boldsymbol{\delta}_1\boldsymbol{\delta}_2$$
$$\text{rotation:} \quad (\mathbf{f}_w\,\mathbf{q})_2 = -\pi\eta\left(-2a + q\ln\frac{q}{h}\right)waq\boldsymbol{\delta}_1\boldsymbol{\delta}_2 \tag{27}$$

In a similar way the contribution of the other particles can be found. Adding the results of all particles finally yields

$$\sum \mathbf{fq} = \pi\eta\left(-2a + q\ln\frac{q}{h}\right)\begin{pmatrix} 0 & \dot{\gamma}q^2 - 2waq & 0 \\ 2waq & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix} \tag{28}$$

The angular velocity of the particles is obtained by equating the off-diagonal components of (28), which leads to the required symmetry of the stress tensor

$$w = \dot{\gamma}q/4a \cong \dot{\gamma}/2 \qquad (h/a \to 0) \tag{29}$$

From (28) and (29) we obtain

$$\sum \mathbf{fq} = \pi\eta q^2\left(-2a + q\ln\frac{q}{h}\right)\bar{\mathbf{D}} \tag{30}$$

Substitution of (30) into (12) with $n = 1/q^3$ yields for the stress tensor

$$\bar{\mathbf{T}} = -\bar{p}\mathbf{1} + 2\eta\left\{1 + \frac{\pi}{4q}\left(-2a + q\ln\frac{q}{h}\right)\right\}\bar{\mathbf{D}} \tag{31}$$

From (31) the instantaneous value of the relative viscosity η_R is found to be

$$\eta_R = 1 + \frac{\pi}{4q}\left(-2a + q\ln\frac{q}{h}\right) \tag{32}$$

Using (21), this expression can be rewritten in terms of ε,

$$\eta_R = 1 + \frac{\pi}{4}\left(\varepsilon - 1 + \ln\frac{1}{\varepsilon}\right) \tag{33}$$

5. A CHECK OF THE LUBRICATION APPROXIMATION

Nunan and Keller analyzed the effective viscosity of a periodic suspension. Their results provide a check for the validity of the lubrication approximation. A simple cubical arrangement of the particles will be analyzed. First, the case of a pure shearing motion will be considered. (This is the case analyzed in Section 3.) According to Nunan and Keller, for this

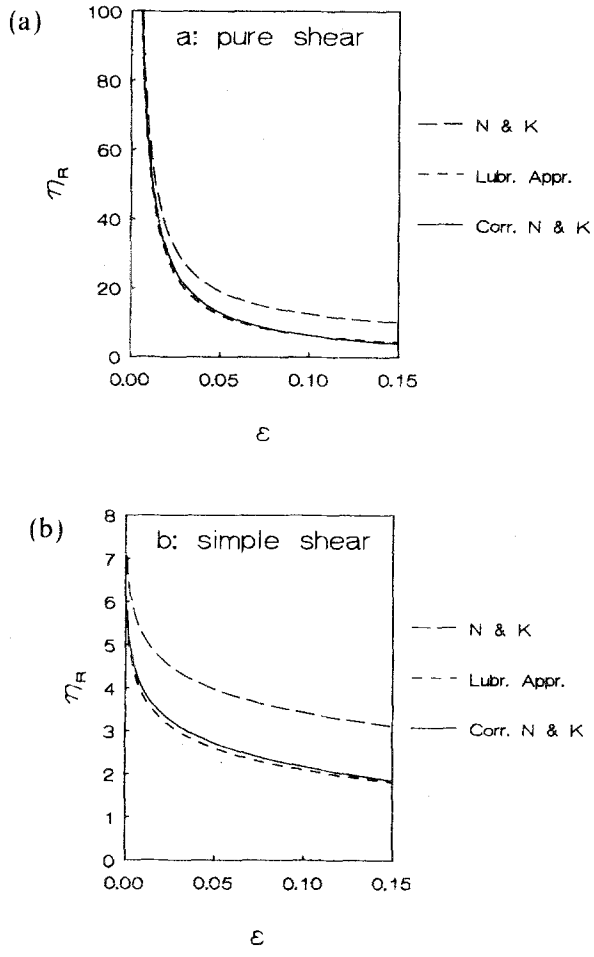

Fig. 4. A comparison between the results of Nunan and Keller and the expressions obtained on the basis of the lubrication approximation. It can be seen that both for pure shear and for simple shear the type of asymptotic behavior is predicted well by the lubrication theory. There is, however, a nearly constant difference between both theories. This difference is negligible if the corrected results of Nunan and Keller (see footnote 3) are compared with our results.

flow field, the high-concentration expansion for the relative viscosity becomes

$$\eta_R = 4.1 + \frac{3\pi}{16}\frac{1}{\varepsilon} + \frac{27\pi}{80}\ln\frac{1}{\varepsilon} + 0.25\varepsilon\ln\frac{1}{\varepsilon} + O(\varepsilon) \tag{34}$$

According to the lubrication theory, the relative viscosity is [cf. (21)]

$$\eta_R = 1 + \frac{3\pi}{16}\frac{1-\varepsilon}{\varepsilon} \tag{35}$$

Next, the case of simple shear will be considered. The instantaneous value of the relative viscosity according to Nunan and Keller is equal to

$$\eta_R = 1.63 + \frac{\pi}{4}\ln\frac{1}{\varepsilon} + O(\varepsilon) \tag{36}$$

Calculation of this situation in the lubrication approximation yields [cf. (22)]

$$\eta_R = 1 + \frac{\pi}{4}\left(\varepsilon - 1 + \ln\frac{1}{\varepsilon}\right) \tag{37}$$

In Figs. 4a and 4b the results of the lubrication approximation are compared with the results of Numan and Keller. It can be seen that the type of asymptotic behavior is predicted well by the lubrication theory. There is, however, a nearly constant difference between the predicted values of the relative viscosity.[3]

6. CONCLUSION

In this paper a systematic derivation of the stress tensor of a concentrated suspension is presented. If the assumptions made by Frankel and Acrivos in their dissipation calculation are adopted in the theory presented here, it can be shown that the same relation between the concentration and the viscosity is obtained if a cubical cell is chosen in the dissipation calculation. The value of 9/8 for the constant, which is

[3] One of the referees was so kind to point out to us that there are typographical mistakes in Nunan and Keller's asymptotic formulas. The constants 4.1 and 1.63 in Eqs. (34) and (36) should be replaced by -2.1 and 0.37, respectively. In Figs. 4a and 4b it can be seen that the differences between the corrected results of Nunan and Keller and the results obtained on the basis of the lubrication approximation are negligibly small.

suggested by Frankel and Acrivos, is the result of an incorrect choice of a spherical cell. The experimental data can be fitted equally well if one assumes a hexagonal instead of a cubical packing.

In calculating the case of simple shear of a cubical arrangement of particles the advantage of using the stress tensor instead of the energy dissipation becomes clear. The angular velocity of the particles is obtained from the requirement that the stress tensor should be symmetrical. Once the angular velocity is known, the value of the relative viscosity can be calculated.

The lubrication approximation is checked against the results obtained by Nunan and Keller. It is shown that both for pure shear and simple shear of a cubical lattice of particles the difference between the lubrication approach and the asymptotic formulas of Nunan and Keller is negligibly small. This indicates that the hydrodynamic interaction between particles in a concentrated suspension may be calculated using the lubrication approximation.

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