The effective conductivity of dilute suspensions

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The effective thermal conductivity of a dilute suspension of particles in a Newtonian fluid is considered. In general, the effective conductivity may be shown to depend not only on the physical properties and concentration of the particles, but also critically on both the type and strength of any bulk flow which may be present. Two distinct types of flow effects are distinguished; the flow dependence of the suspension microstructure (i.e., the statistical distributions of orientation and shape for the particles), and the flow dependence of the local temperature distributions at the microscale of the individual particles. Specific calculations are reported for suspensions of spherical drops, slightly deformed drops, and rigid prolate spheroidal particles all in the limit of small particle Peclet number. In the latter case, we include the influence of Brownian rotations.

W niniejszej pracy rozważamy wpływ małych kropel zawieszonych w cieczy i sztywnych cząsteczek na efektywne przewodnictwo cieplne rozcieńczonej zawiesiny, gdy materiał jako całość poddany jest ruchowi prostego, liniowego ścinania. Rozważane są trzy odrębne przypadki: cząstki kuliste, lekko zdeformowane krople i sztywne elipsoidy obrotowe. W każdym z tych przypadków na teorię składa się szczegółowa analiza zaburzenia pola temperatury w otoczeniu typowej cząstki, po czym następuje proces statycznego uśrednienia, pozwalający na otrzymanie interesujących wielkości makroskopowych przy wykorzystaniu pola lokalnej prędkości i pola temperatury. W rezultacie otrzymuje się jawne wyrażenie na efektywne przewodnictwo ciepła materiału uważanego jako kontinuum złożone. Głównymi uproszczeniami w tej analizie, oprócz słabej koncentracji cząstki, są założenia o niskich (w tej mikroskali) liczbach Reynoldsa i Pecleta.

В настоящей работе рассмотрено влияние малых, взвешенных в жидкости, капель и жестких частиц на эффективную теплопроводность разбавленных взвесей, когда материал, как целое, подвергается движению простого, линейного сдвига. Рассматриваются три отдельных случая: сферические частицы, легко деформируемые капли и жесткие эллипсоиды вращения. В каждом из этих случаев теория состоит из подборного анализа возмущения поля температуры в окрестности типичной частицы, затем наступает процесс статистического усреднения, позволяющий получить интересующие нас макроскопические величины, используя поле локальной скорости и поле температуры. В результате получается явное выражение для эффективной теплопроводности материала, рассматриваемого как сложное континуум. Главными упрощениями в этом анализе являются, кроме слабой концентрации частиц, предположения о малых, в этом микромасштабе, числах Рейнольдса и Пекле.

1. Introduction

A GREAT deal of effort has been expended in an attempt to understand and correctly model the mechanical or rheological properties of suspensions, macromolecular solutions and other non-Newtonian fluids. Equally important in many industrial processing applications, however, is the constitutive behavior of such materials for heat or material transport in the presence of bulk gradients of temperature or concentration of molecular species. Unfortunately, by comparison with the enormous number of rheological investigations, almost no research (either experimental or theoretical) has been reported on this important topic. Indeed, about all that can be said with certainty is that the simple

linear and instantaneous equations of Fourier and Fick relating heat or molecular species flux to the gradients of temperature or concentration do not hold in many non-Newtonian materials.

The present paper is concerned with composite (or suspension-like) materials. In this case, typical dimensions of the suspended particles are large relative to the intermolecular length scales of the suspending material which is thus modelled as a continuum. When the suspension as a whole undergoes a non-uniform bulk flow, a local disturbance flow is induced in the vicinity of each particle and a local convective flux of heat or molecular species will result which can contribute to the effective transport rate for the composite material. As a consequence, the apparent conductivity or diffusivity of the composite may be quite different from that of either the discrete or continuous phase materials; dependent not only on the material properties of the two phases, but also on the type and strength of any bulk flow which is present.

If we consider the material as a whole, there are two distinct approaches available for the development of constitutive relationships. By far the most widely used is the continuum or phenomenological approach in which an initial hypothesis is made of the appropriate form of the constitute relationship for a particular material or class of materials. The vast majority of resulting models are characterized by excessive generality. Less widely applied is the structural model approach in which one attempts to deductively obtain the appropriate constitutive law starting from a description of the material on the scale of the individual suspended particles. This latter approach is difficult, but has the advantage of being a predictive theory in the sense of exposing relationships between microstructure or dynamics and macroscopically observable material properties. Thus, although the requirements of mathematical tractability demand reasonably simple microstructure, the predictions for the resulting material are at least qualitatively correct.

In the present paper, we use the structural model approach to determine the effective thermal conductivity for a dilute suspension of either rigid prolate spheroids, or slightly deformed drops. The suspension is assumed to be undergoing a steady, simple shear flow, with heat transfer occurring as a result of a constant temperature gradient in the cross-flow direction. In order to facilitate rigorous analysis of the problem, it is further assumed that the local Peclet number based on the length scale of the particles is small. Finally, in the case of the rigid spheroids, we allow for a significant degree of rotational Brownian motion but neglect other randomizing disturbance mechanisms. Although admittedly somewhat restricted, these model calculations do at least allow a useful first view of the effect of flow-induced particle orientation and deformation on the bulk thermal transport characteristics of a dilute suspension.

2. Basic formulation and general properties of composite materials

We consider a suspension of neutrally buoyant particles in the presence of a bulk shear flow and a bulk temperature field. The objective of the present theory is the development of a constitutive equation which describes the effective thermal conductivity for the suspension considered as an equivalent homogeneous material.

The point of view adopted is the conventional one in the field of suspension rheology. We assume that particles are sufficiently large that the suspending fluid may be treated as a homogeneous continuum; thus we assume that the minimum dimension l of the particles is large compared to the intermolecular scale α of the suspending medium. The latter is then modeled as an incompressible Newtonian fluid in which a simple Fourier heat conduction law is applicable.

At any arbitrary fixed point in the suspension, the local variables such as velocity, temperature, enthalpy or conductive heat flux are random functions of time whose values at any instant depend upon the proximity of suspended particles. In this sense, the description of bulk or macroscopic quantities for the suspension is a problem of statistics. At the fundamental level, the most appropriate definition of the bulk variables is as an ensemble average of the corresponding microscale quantities for a large number of realizations of the system. Instantaneous local values of the velocity, temperature, enthalpy and conductive heat flux may then be expressed as a sum of the ensemble averaged quantity, and an additional microscale or fluctuating component, i.e.,

$$u_i = \langle u_i \rangle + u_i', \quad T = \langle T \rangle + T',$$
(2.1)
$$h = \langle h \rangle + h', \quad q_i = \langle q_i \rangle + q_i',$$

where, by definition,

$$\text{(2.2)} \qquad \langle u_i' \rangle = \langle T' \rangle = \langle h' \rangle = \langle q_i' \rangle = 0.$$

It is desired to define a bulk conductive heat flux Q_i such that the thermal energy balance for the suspension, viewed as an equivalent homogeneous medium, takes the usual form

$$\text{(2.3)} \qquad \frac{\partial \langle h \rangle}{\partial t} + \langle u_i \rangle \frac{\partial \langle h \rangle}{\partial x_i} + \frac{\partial Q_i}{\partial x_i} = 0.$$

A convenient, if heuristic, method of determining the proper definition of Q_i for this purpose, is to simply apply the same ensemble averaging used in (2.1), to the exact, instantaneous thermal energy balance which is applicable for each realization of the system

$$\text{(2.4)} \qquad \frac{\partial h}{\partial t} + u_i \frac{\partial h}{\partial x_i} + \frac{\partial q_i}{\partial x_i} = 0.$$

Taking account of (2.1) and (2.2), the result is

$$\text{(2.5)} \qquad \frac{\partial \langle h \rangle}{\partial t} + \langle u_i \rangle \frac{\partial \langle h \rangle}{\partial x_i} + \frac{\partial}{\partial x_i} \langle q_i + u_i' h' \rangle = 0.$$

Comparing (2.3) and (2.5), it follows that

$$\text{(2.6)}_1 \qquad Q_i = \langle q_i \rangle + \langle u_i' h' \rangle.$$

It may be seen that the bulk conductive heat flux consists of an ensemble average of the instantaneous, microscale conductive heat flux, plus an additional "convection" term which accounts for the transport of heat by means of the local fluctuating velocity and enthalpy fields.

To proceed further from a theoretical point of view, it is necessary to replace the ensemble averages in (2.6) with more easily calculable spatial (volume) averages. For this purpose, it is assumed that there exists in the suspension a volume V, containing a statistically significant number of particles, whose linear dimensions of $O(V^{1/3})$ are therefore much larger than the particle length scale l, but smaller than the length scale L over which significant variations occur in the bulk velocity or temperature gradients, or in the concentration of particles. The volume averaged variables, such as

$$\langle\langle q_i \rangle\rangle \equiv \frac{1}{V} \int_V q_i dV$$

are then equal to the ensemble averaged variables and are thus independent of the size and shape of V. These volume averaged quantities clearly vary only on the scale of $O(L)$. They are thus point quantities with respect to the overall macroscopic description of the material. The local, fluctuating variables, e.g. q_i', vary at random over distances of $O(l)$ as a result of the assumed random location of the particles in the suspension.

With the conditions of local homogeneity on the scale of $O(L)$ satisfied, the ensemble averages of $(2.6)_1$ can be replaced exactly with volume averages to give

$(2.6)_2$ $$Q_i = \langle\langle q_i \rangle\rangle + \langle\langle u_i' h' \rangle\rangle.$$

Expressing the averaging symbols in terms of the appropriate volume integrals, and assuming that the suspending fluid and particles both satisfy a simple Fourier law for heat conduction, this latter expression can be rewritten as

(2.7) $$Q_i = -\frac{1}{V} \int_{V-\Sigma V_0} k_1 \nabla T dV - \frac{1}{V} \int_{\Sigma V_0} k_2 \nabla T dV$$
$$+ \frac{1}{V} \varrho C_p^{(1)} \int_{V-\Sigma V_0} u_i' T' dV + \frac{1}{V} \varrho C_p^{(2)} \int_{\Sigma V_0} u_i' T' dV.$$

Here, k_1 and k_2 are the thermal conductivities of the suspending fluid and particles, respectively, and $C_p^{(1)}$ and $C_p^{(2)}$ are their heat capacities. The volume of a typical particle in the averaging volume V is denoted as V_0, and ΣV_0 therefore represents the total volume of particles in V. Finally, utilizing the definition of the bulk temperature gradient

$$\langle\langle \nabla T \rangle\rangle = \frac{1}{V} \int_V \nabla T dV = \frac{1}{V} \int_{V-\Sigma V_0} \nabla T dV + \frac{1}{V} \int_{\Sigma V_0} \nabla T dV,$$

the expression (2.7) becomes

(2.8) $$Q_i = -k_1 \langle\langle \nabla T \rangle\rangle + \frac{(k_1 - k_2)}{V} \int_{\Sigma V_0} \nabla T dV + \frac{(C_p^{(2)} - C_p^{(1)})}{V} \int_{\Sigma V_0} u_i' T_1' dV + \varrho C_p^{(1)} \langle\langle u_i' T' \rangle\rangle.$$

The first term on the right-hand side is just the conductive heat flux which would exist in the absence of the particles if the same temperature gradient were maintained. The remainder of the terms thus represent the additional contributions to the bulk conductive heat flux due to the presence of the particles.

The expression for Q_i may first be considered for a composite material which is *not* flowing. In this case, the last two terms in (2.8) are identically zero, and the bulk con-

ductive heat flux differs from that of the suspending fluid only when $k_1 \neq k_2$, as expected on simple physical grounds. When $k_1 \neq k_2$, however, the microscale temperature field, and thus Q_i, depend not only on the particle conductivity, but also on their geometry, concentration and orientation distribution. Detailed results have been obtained by many investigators for this case of conduction through a motionless, composite material, apparently beginning with MAXWELL (1873) and ending, most recently, with ROCHA and ACRIVOS (1973), who considered dilute composites with particles of arbitrary shape, and JEFFREY (1973), who considered non-dilute concentration effects in a composite of spherical inclusions. It may be noted that for $k_2/k_1 = 0(1)$, the purely conducting composite will respond to changes in the imposed temperature field on a time scale which is the same magnitude as would occur for the suspending fluid alone. Furthermore, for a given thermodynamic state, the effective thermal conductivity of the composite is a unique property of the material.

In the presence of a bulk flow, the situation becomes much more complicated. Not only are the geometry, concentration and orientation of the particles important, but the bulk conductive heat flux, Q_i, depends critically on both the type and strength of the flow. Indeed, even when the thermal properties of the dispersed and continuous phases are identical, the presence of the last term in (2.8) strongly suggests that the existence of bulk flow may alter the bulk conductive flux Q_i from that of the motionless composite; that is, the bulk conductive flux Q_i is a function not only of the material, but also of the flow.

In the general case, the presence of a flow has two conceptually distinct, though coupled, effects. *The first* is the direct effect of convection on the local temperature distribution, for a given microstructural configuration (i.e., for particular distributions of particle shape, orientation, and position). As the local temperature distributions are altered by changes in the bulk flow strength, the flow type, or the bulk temperature profile, so too are the contributions from all the integral terms in the expression (2.8) for Q_i. It may be noted that these changes in Q_i due to changes in the local temperature fields will generally show a transient (or memorylike) character. The local disturbance velocity field is established instantaneously for a given microstructure (the microdynamics is assumed to be inertialess and natural convection is neglected). However, the local temperature field only changes on a finite time scale which depends on the thermal properties of the two phases, as well as the nature of the flow.

The *second* major flow effect on the bulk conductive heat flux Q_i is due to the flow dependence of the microstructural configurations. We have noted previously that the bulk conductivity of the material is sensitive to both the shape and orientation distribution of the suspended particles. For rigid nonspherical particles, there is a flow-induced rotation which causes the orientation distribution to change from its form in the motionless composite. For deformable particles, there is the additional effect of hydrodynamically induced particle deformation. In any real suspension these flow-induced changes in the microstructure are resisted by one or more "restoring" mechanisms, like rotational Brownian motion or particle elasticity, which tend to maintain the equilibrium or rest configuration which would exist in the absence of flow. Thus the microstructural state, and Q_i, depend on both the flow *type* and the flow *strength* relative to the strength of the

restoring mechanism. In addition, the hydrodynamically induced changes in microstruc-
ture associated with such effects as the rotation and deformation of the particles are all
deterministic (initial-value) processes, and thus provide a second mechanism by which
the bulk conductive heat flux will exhibit a "memory" for past microstructural states of
the material. The fact that the microstructure always tends to return to its rest state on
a finite time scale λ^{-1} insures that this memory-like contribution to Q_i is "fading" in the
sense that the dependence on recent microstructural states is stronger than the dependence
on earlier states.

In summary, these general considerations indicate that the constitutive behavior for
conductive heat flux of a composite, suspension-like material, in the presence of flow,
will exhibit dependence both on the instantaneous flow *type* and *strength*, as well as the
histories of the bulk deformation, and temperature distributions. Indeed, from a broader
viewpoint, it may be suggested that many other fluid-like materials which are rheologically
complex (polymer solutions, for example), will also exhibit similar flow-induced com-
plexities in the constitutive relations for the conductive heat or diffusive mass flux!

3. Calculations of the bulk conductive heat flux

In order to provide more detailed results, it is necessary to evaluate the expression
(2.8) for specific imposed bulk flows and specific types of particles. Unlike the theories
for motionless composite materials, however, relatively few such calculations have yet
been completed. The main difficulty is that one must determine the detailed local tempera-
ture distribution at the scale of the suspended particles. In general, this would require
solutions of the full microscale thermal energy and momentum balances, both inside
and outside the particles, an impossibly complicated task.

We consider here only the much simplified case of dilute suspensions in which both
thermal and hydrodynamic particle-particle interactions are neglected. Furthermore, we
assume that the microscale disturbance flow is creeping (i.e., the Reynolds number,
$l^2\gamma\varrho/\mu$, is identically zero), so that the local velocity field near a single particle can be
determined exactly. Finally, all of the particles will be assumed to be identical; under
these circumstances the temperature distribution around any single particle is identical
with that around any other particle and the sums over particle volume in (2.8) can be re-
placed by a single integral over one particle multiplied by the volume fraction of sus-
pended particles. In this case, (2.8) becomes

$$(3.1) \quad Q_i = -k_1\langle\langle\nabla T\rangle\rangle$$
$$+ \frac{(k_1-k_2)\Phi}{V_0} \int_{S^*} (n_i T)dS + \varrho\frac{(C_{p_2}-C_{p_1})}{V_0}\Phi \int_{V^*} u_i'T'dV + \varrho\frac{C_{p_1}}{V_0}\Phi \int_{V} u_i'T'dV,$$

where n_i is the outer unit normal to the particle surface, Φ the volume fraction of sus-
pended particles, and V_0 the volume of a single particle. The limits S^*, V^* and V on the
integrals indicate integration over the surface (S^*), the volume (V^*), and the complete
disturbance velocity and temperature domain (V) of a typical particle.

In order to proceed further, it is necessary to consider specific forms for the imposed bulk velocity and temperature fields, and thus calculate the *local* temperature field in the vicinity of a suspended particle. The only case which has so far been considered is a steady bulk shear flow

(3.2) $$\langle\langle u\rangle\rangle = \gamma y, \quad \langle\langle v\rangle\rangle = \langle\langle w\rangle\rangle = 0$$

and a steady linear bulk temperature profile

(3.3) $$\langle\langle T\rangle\rangle = \bar\alpha y.$$

The governing equation for the local temperature field in the two phases [Eq. (2.4) appropriately nondimensionalized, has the general form

(3.4) $$\frac{1}{Pe_i}\nabla^2 T = \mathbf{u}\cdot\nabla T + \frac{\partial T}{\partial\tau}$$

both inside and outside the particle. Here, $\mathbf{u}$ represents the appropriate creeping flow solution of the Navier-Stokes equations and Pe_i is the Peclet number for the particular phase (i)

$$Pe_i \equiv \frac{l^2\delta\gamma\varrho C_{p_i}}{k_i}.$$

The boundary conditions are that the temperature and heat flux inside and outside the particle match at its surface, and that $T \to \alpha y$ as $|r| \to \infty$. Analytic solutions of (3.4), subject to these conditions, can be obtained either in the asymptotic limit $Pe \to 0$, corresponding to conduction dominated heat transfer or in the limit $Pe \to \infty$, in which convection effects dominate.

The problem of interest is to determine the influence of the bulk flow on the bulk conductive heat flux, Q_i. It may be noted that the limit $Pe_i \to 0$ necessarily restricts the convective contributions to Q_i to small magnitude corrections of the results for a motionless composite. NIR and ACRIVOS (1975) have therefore attempted analytical solutions of (3.4) for $Pe_i \gg 1$, where the flow contributions to Q_i may be large, of $0(Pe_i^m)$ with $0 < m \leqslant 1$. Unfortunately, even for rigid, spherical particles the solution is difficult and not yet fully complete. For nonspherical or deformable particles where the flow-induced particle rotation causes the microscale temperature problem to be fully time-dependent and three-dimensional, the large Pe case appears completely intractable, even for steady imposed bulk velocity and temperature distributions. Thus, the detailed calculations which we shall describe in the remainder of this paper deal exclusively with the regime

$$Pe_i \ll 1.$$

Three specific cases are considered: a dilute suspension of spherical drops; a dilute suspension of drops which are slightly deformed by the shear flow; and a dilute suspension of rigid prolate spheroids in the presence of rotational Brownian motion.

3.1. A dilute suspension of spherical drops (Pe ≪ 1)

In the first case we consider perfectly spherical neutrally buoyant drops of radius a which are composed of a Newtonian fluid with viscosity μ_2, thermal conductivity k_2 and heat capacity C_{p_2}. The suspending fluid is also Newtonian with viscosity, thermal conductivity and heat capacity μ_1, k_1 and C_{p_1}, respectively. The analysis of the microscale temperature distribution in this case involves a straightforward application of the methods of matched asymptotic expansions, using the velocity fields calculated by "TAYLOR (1932) for the creeping motion of a spherical drop in a simple shear flow of the suspending fluid. The details of this calculation are reported by LEAL (1973), and will not be repeated here.

When the integrals in (9) are evaluated using the calculated microscale temperature distributions, an explicit expression for the bulk conductive heat flux is obtained,

$$(3.5) \quad Q_y = -k_1\bar{\alpha} - k_1\bar{\alpha}\Phi\left[(k_2-k_1)\frac{3}{k_2+2k_1} + \left(1.176\frac{(k_2-k_1)^2}{(k_2+2k_1)^2}\right.\right.$$

$$\left.\left. + \left(\frac{2\mu_1+5\mu_2}{\mu_1+\mu_2}\right)\left(0.12\frac{2\mu_1+5\mu_2}{\mu_1+\mu_2} - 0.028\frac{k_2-k_1}{k_2+2k_1}\right)\right)\text{Pe}_1^{3/2} + 0(\text{Pe}_1^2)\right].$$

We restrict attention to the component of Q_i in the cross stream direction of the bulk temperature gradient. The effective conductivity k^*, corresponding to (3.5), is simply

$$(3.6) \qquad\qquad\qquad k^* = -\frac{Q_y}{\alpha}.$$

Of particular interest is the case $k_1 = k_2$ where

$$(3.7) \qquad\qquad k^* = k_1\left[1 + 0.12\Phi\left(\frac{2\mu_1+5\mu_2}{\mu_1+\mu_2}\right)^2\text{Pe}_1^{3/2}\right].$$

As suggested in the previous section, the effective conductivity is enhanced by the existence of the shear flow, even when the thermal conductivities of the two phases are identical. Indeed, as the difference between k_1 and k_2 is increased, the flow contribution to k^* increases rapidly, regardless of whether $k_2 > k_1$ or $k_2 < k_1$. The pure conduction contribution to (3.6), on the other hand, either increases or decreases as k_2 is increased or decreased relative to k_1. Furthermore, as shown both by (3.5) and (3.7), the degree of flow enhancement depends not only on the physical properties of the two fluids, but also on the shear rate, through the dependence of k^* on Pe_1. Finally, an interesting feature of the analysis leading to (3.5) is the fact that the first flow-induced contribution to k^* is $0(\text{Pe}_1^{3/2})$ in spite of the fact that there are nontrivial modifications of the local temperature field at $0(\text{Pe}_1)$. Since this result would appear to be due to the special symmetry induced in the temperature field by the assumed sphericity of the drop, one may ask whether any fundamental change would occur in the predicted flow rate dependence of k^* when account is taken of deformations of the drop to non-spherical geometry. Specifically, we may ask whether even very small deformations of shape might not lead to significant modifications of k^* when compared to the $0(\text{Pe}_1^{3/2})$ terms for the perfect sphere. From a more applied point of view, the problem is to determine the circumstances in which

(3.5) provides an adequate first estimate of the flow enhancement of k^* for real emulsions, where small deviations from the spherical shape are inevitable even for large values of the surface tension or droplet viscosity.

3.2. A dilute suspension of slightly deformed drops (Pe ≪ 1)

In order to investigate the influence of particle deformation on the conductivity of a dilute suspension, we now consider the case of neutrally buoyant drops which are slightly (order ε) deformed by the ambient shear flow. Following the earlier work of TAYLOR (1932), the analysis is restricted to the two limiting cases of dominant interfacial tension forces ($\varepsilon \sim a\gamma\mu_1/\sigma \ll 1$), and dominant internal (drop) viscosity effects ($\varepsilon \sim$ $\sim 1/\lambda \ll 1$). Here, λ is the ratio μ_2/μ_1, σ the interfacial tension and a the undeformed drop radius. The local temperature field is obtained as a double expansion in Pe_1 and ε. The details are reported by McMILLEN and LEAL (1975). At $0(1)$ in ε, the solutions are those obtained by LEAL (1973). The solutions at $0(\varepsilon)$ were carried out to $0(\varepsilon Pe_1)$ for the case of deformation dominated by interfacial tension forces and to $0(\varepsilon Pe^{3/2})$ in the viscosity dominated limit.

Using the microscale temperature solutions, the effective conductivity, k^*, can again be evaluated from (3.1) and (3.6). For the case of surface tension controlling deformation, we obtain

$$(3.8) \quad \frac{k^*}{k_1} = 1 + \Phi\left\{\frac{3(m-1)}{m+2} + \left(\frac{1.176(m-1)^2}{(m+2)^2} + \frac{5\lambda+2}{\lambda+1}\left[0.12\left|\frac{5\lambda+2}{\lambda+1}\right|\right.\right.\right.$$
$$\left.\left.\left. -0.028\left|\frac{m-1}{m+2}\right|\right]\right)Pe_1^{3/2} + I(m, \lambda, \tau)\varepsilon Pe_1 + 0(\varepsilon^2) + 0(Pe_1^2) + 0(\varepsilon Pe_1^{3/2}) + ...\right\},$$

where m and τ are the ratios k_2/k_1 and C_{p_2}/C_{p_1}, respectively. $I(m, \lambda, \tau)$ is a rather complex function of the three physical property ratios m, λ, τ, which is given explicitly by McMILLEN and LEAL (1975). It may be noted that $I(m, \lambda, \tau) = 0$ for equal values of the conductivities, $m = 1$[1]. For the case of internal viscous forces controlling deformation we find

$$(3.9) \quad \frac{k^*}{k_1} = 1 + \Phi\left\{\frac{3(m-1)}{m+2} + \left(3.00 - 0.14\frac{(m-1)}{(m+2)} + 1.176\frac{(m-1)^2}{(m+2)^2}\right)Pe_1^{3/2} - 3.6\frac{(m-1)^2}{(m+2)^2}\varepsilon\right.$$
$$\left. + \left(1.411\frac{m(m-1)^2}{(m+2)^3}\left|\left(\frac{\tau}{m}\right)^{3/2} - 1\right| - 0.168\frac{(m-1)^2}{(m+2)^2}\right)\varepsilon Pe_1^{3/2} + 0(\varepsilon^2) + 0(Pe_1^2) + 0(\varepsilon Pe_1^2) + ...\right\}.$$

In this case, the terms which arise due to drop deformation are of order ε and $\varepsilon Pe_1^{3/2}$, and are thus never more than small corrections relative to the contributions of $0(1)$ and $0(Pe_1^{3/2})$ for the case of a perfect sphere, Eq. (3.5). In contrast, however, the deformation-induced convective contribution to k^* in the interfacial tension dominant limit is of $0(\varepsilon Pe_1)$. In this case the relative magnitudes of the flow-induced contribution for a sphere which is $0(Pe_1^{3/2})$, and for a slightly deformed sphere where there is an additional term

[1] It may be shown, by consideration of the structure of the general expression (2.8) and the asymptotic solutions of (3.5) for $Pe_1 \ll 1$, that the $0(Pe_1)$ contribution must vanish in all cases when $k_2 = k_1$.

of $0(\varepsilon Pe_1)$, depend critically on ε and Pe_1 (for each fixed $m \neq 1$). Clearly, however, the perfect sphere result can be significantly modified or even dominated completely in some circumstances by the deformation-induced flow contribution. Let us now examine the results, (3.8) and (3.9), in more detail.

For the case of deformation dominated by viscous effects, we may note first that the pure conduction term induced by the change from spherical shape is always negative (except for $m = 1$ where it is zero). This is easily understood by noting that the drop deforms with its principle axis of contraction in the cross-flow (y) direction and of extension in the flow (x) direction. Thus, in cases where $k_2 > k_1$ (i.e., $m > 1$), the pure conduction enhancement of k^* is decreased as the dimension of the drop in the direction of heat transfer is decreased. On the other hand, when $k_2 < k_1$ so that the pure conduction effect of the drops is to decrease k^*, the change becomes smaller as the drop deforms. The flow-induced contribution to k^* is always strictly positive in the case of a perfect sphere. However, the deformation-induced correction, $0(\varepsilon Pe_1^{3/2})$, may be either positive, negative, or zero, as shown in Fig. 1, depending on the magnitude of the thermal con-
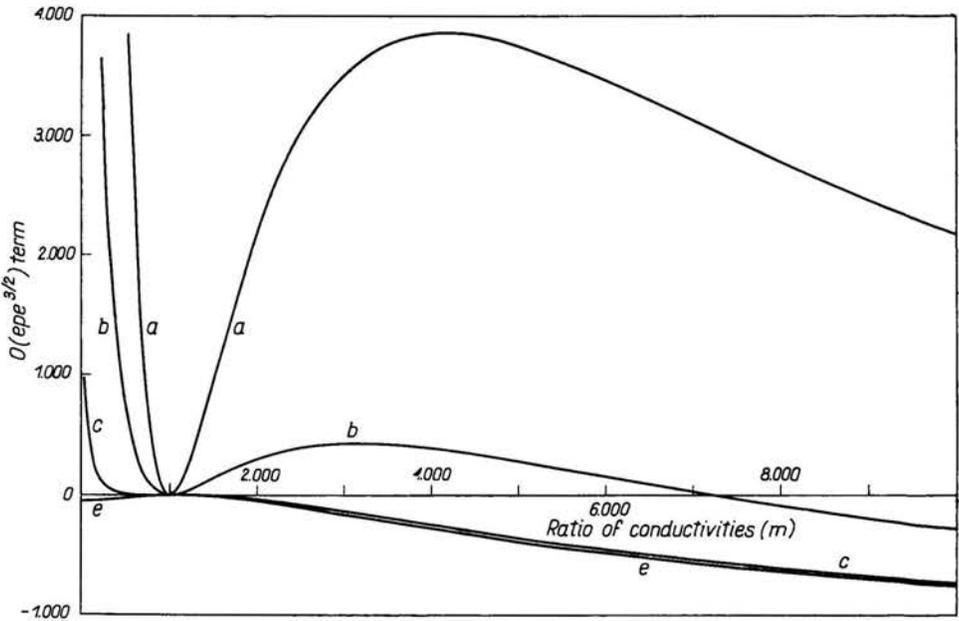
FIG. 1. The $0(\varepsilon Pe_1^{3/2})$ term in the effective conductivity when viscous forces control deformation, as a function of the conductivity ratio, $m = k_2/k_1$, for several values of the heat capacity ratio, $\tau = C_{P_2}/C_{P_1}$, a: $\tau = 3$, b: $\tau = 2$, c: $\tau = 1$, d: $\tau = 0.5$, e: $\tau = 0$.

ductivity ratio, m. In any case, the flow contribution associated with small deformations is always dominated by the contribution for the sphere and the complex results inherent in Fig. 1 are no more than qualitatively suggestive of the more important changes which may occur when larger deformations of shape occur, as will often happen in the real system.

For the case of deformation controlled by surface tension forces, the drops deform along the principle axis of strain of the undisturbed shear flow, thus elongating along an axis 45° from the flow (x) direction. As noted previously, the flow-induced contribution for a sphere is always positive (i.e., k^* is enhanced by the existence of flow). However, the dominant deformation-induced contribution to k^* is the flow term, of order εPe_1, which can be either positive or negative depending on the values of m, λ, and τ. The general characteristics are demonstrated in Fig. 2, where $I(m, \lambda, \tau)$ is plotted as a function of m and λ for three values of τ. We again note that $I(m, \lambda, \tau) \equiv 0$ for $m = 1$. The limiting case, $m \to 0$, is of special interest and can also be evaluated analytically provided care is taken to hold Pe_2 fixed (and small) so that the constraints Pe_1, $Pe_2 \ll 1$ are not violated. In this case,

$$(3.10) \quad \frac{k^*}{k_1} \sim 1 + \Phi \left\{ -\frac{3}{2} + \left[(-9.382\lambda^2 - 17.640\lambda - 24.543) + \frac{1}{\tau}(1.341\lambda^2 \right. \right.$$
$$\left. \left. + 7.096\lambda + 2.940) \right] \frac{\varepsilon Pe_2}{32(\lambda+1)^2} + 0(m^{3/2} Pe_2^{3/2}) + \dots \right\}, \quad (m \to 0).$$

It is particularly noteworthy that the deformation-induced contribution in this limit completely dominates the largest flow contribution which occurs for a spherical drop. Thus, as suggested in the preceding discussion, the presence of even a small deformation of shape can cause a fundamental change in the dependence of k^* on the flow and material parameters. It would thus appear that care must be taken in attempting to correlate experimental data for any suspension in which the particles are not *exactly* spherical with theoretical results for a suspension of spheres.

3.3. A dilute suspension of rigid prolate spheroidal particles with rotational Brownian motion (Pe ≪ 1)

As a final example, we consider the case of a dilute suspension of rigid spheroidal particles which are subjected to significant rotational Brownian motion. Unlike the previous example, the degree of departure from spherical shape is not restricted. Indeed, we assume that the ratio of major to minor semi-axis length, which we denote as Γ_p, can vary from 1 (sphere) $\leqslant \Gamma_p < \infty$ (a highly elongated rod).

In the absence of Brownian motion, the rotational motion of the particles is periodic and described by the familiar orbit equations of JEFFREY (1923). JEFFREY also calculated the quasi-steady Stoke's velocity field for an arbitrary instantaneous orientation and rate of particle rotation. Unfortunately, however, Jeffrey's solution was obtained using a Cartesian axis system fixed in the particle, and is extremely inconvenient for use in (3.5). Thus, in the present work, it was necessary to re-solve the problem using prolate spheroidal coordinates to obtain a more convenient form. The microscale heat transfer problem is, of course, time-dependent as a result of the particle rotation, however the limiting case Pe ≪ 1 can still be solved using a straightforward perturbation scheme provided care is taken to include the rotation-induced time-dependent terms at each order in Pe. The final result for Q_y is of the general form

$$(3.11) \quad Q_y(\theta_1, \phi_1) = Q_y(\theta_1, \phi_1; \Gamma_p, m, \tau),$$

a

λ

m

$I_{(m,\lambda,\Upsilon)}$

-17

b

m

$I_{(m,\lambda,\Upsilon)}$

-17
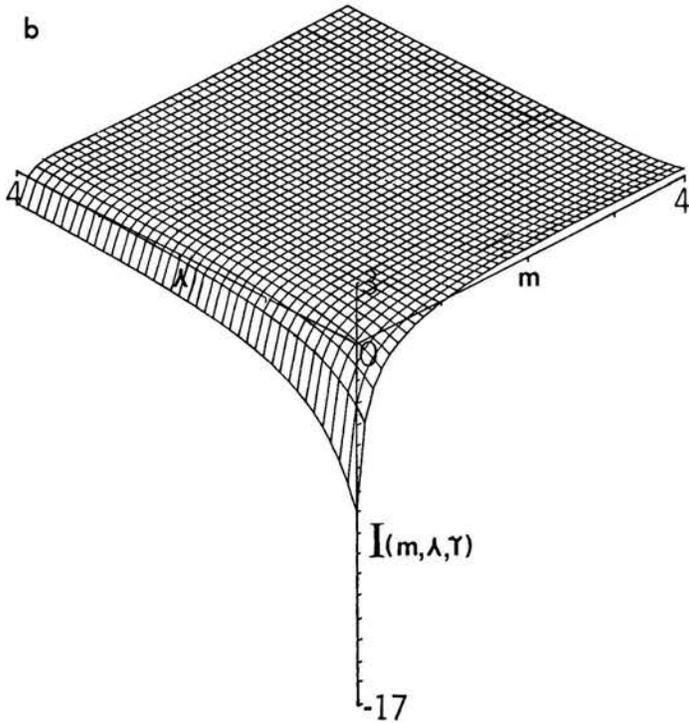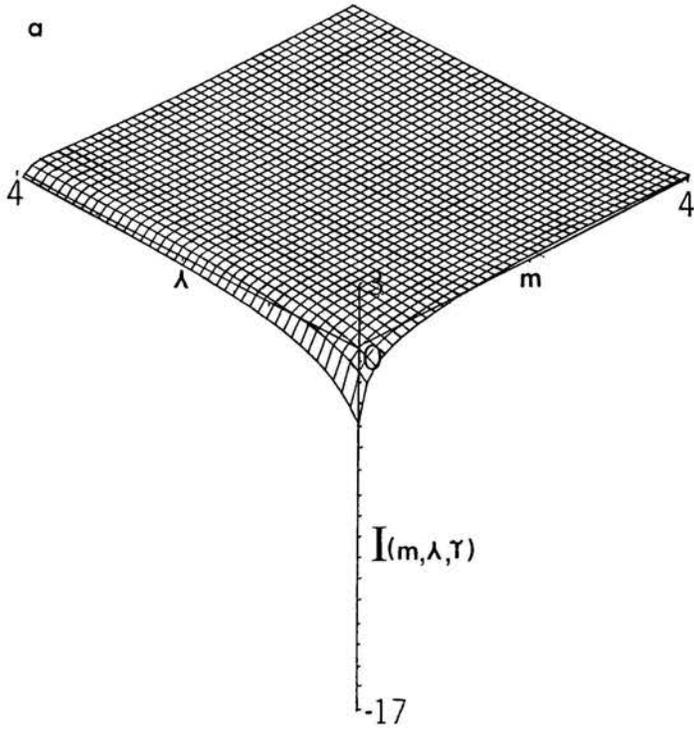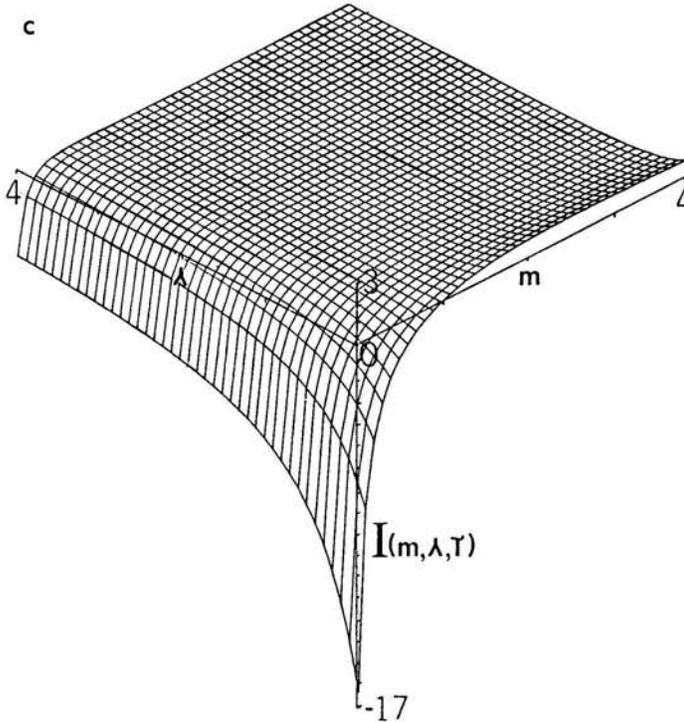
FIG. 2. The $0(\varepsilon Pe_1)$ term, $I(m, \lambda, \tau)$, in the effective conductivity when interfacial tension forces control deformation, as a function of the conductivity ratio, $m = k_2/k_1$, and the viscosity ratio, $\lambda = \mu_2/\mu_1$, for several values of the heat capacity ratio, $\tau = C_{p_2}/C_{p_1}$. Detail A: $\tau = 0.5$; Detail B: $\tau = 1$; Detail C: $\tau = 2$.

where θ_1 and ϕ_1 are the polar angles defined in Fig. 3. In order to proceed further, this orientation dependent expression must be averaged over all possible orientations taking account of the orientation distribution of the particles. If the orientation distribution function is denoted by $N(\theta_1, \phi_1)$, the quantity of interest is

$$(3.12) \qquad \bar{Q}_y \equiv \int_0^\pi d\theta_1 \int_0^{2\pi} \sin\theta_1 \, \partial\phi_1 [N(\theta_1, \phi_1) Q_y(\theta_1, \phi_1)].$$

In the absence of Brownian motion or other disturbance effects, the orientation distribution is determined in a dilute suspension by the motion about Jeffrey orbits. The action of rotary Brownian motion is a randomizing influence on the particle orientation. Thus, with Brownian motion present, the distribution of orientations represents a compromise between the hydrodynamic-induced distribution associated with the undisturbed Jeffrey orbits and the uniform distribution which results from unopposed Brownian rotation. The probability distribution function for particle orientation is governed by a modified Fokker-Plank equation. Solutions of this equation, including a partial list of references to earlier work may be found in Leal and HINCH (1971) and HINCH and LEAL (1972). We consider here only the limit of strong Brownian motion, $D/\gamma \gg 1$, where D is the

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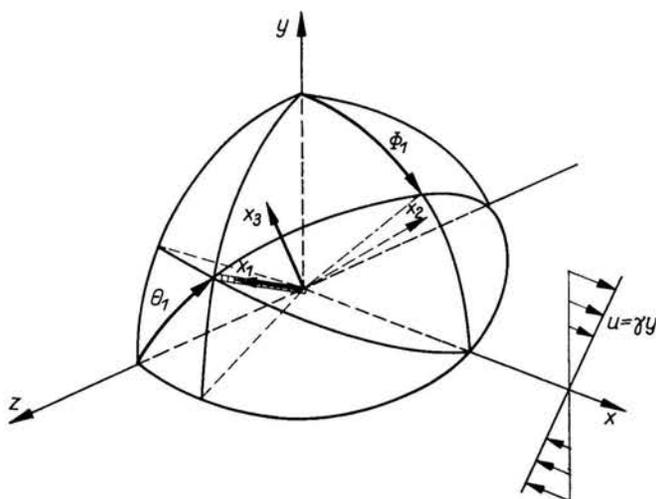
FIG. 3. The orientation angles θ_1 and ϕ_1 for the rotating particle.

rotational Stokes-Einstein diffusion coefficient for the particles. In this case, the orientation distribution function takes the general form

$$(3.13) \qquad N(\theta_1, \phi_1) = \frac{1}{4\pi}\left\{1 + \left(\frac{D}{\gamma}\right)N_1(\theta_1, \phi_1) + \left(\frac{D}{\gamma}\right)^2 N_2(\theta_1, \phi_1) + \dots\right\}.$$

Substituting (3.13) into (3.12) one obtains

$$(3.14) \qquad \bar{Q}_y = -k_1\alpha - k_1\alpha\Phi\left\{\bar{Q}_0^0(m, \Gamma_p) + \bar{Q}_2^0(m, \Gamma_p)\left(\frac{\gamma}{D}\right)^2 + 0\left(\frac{\gamma}{D}\right)^4 \right.$$

$$\left. + \mathrm{Pe}_1\left(\frac{\gamma}{D}\right)\bar{Q}_1^1(m, \tau, \Gamma_p) + 0\left(\mathrm{Pe}_1\left(\frac{\gamma}{D}\right)^2\right) + 0(\mathrm{Pe}_1^{3/2})\right\}.$$

The first term, $\bar{Q}_0^0$, is the pure conduction contribution with a random orientation distribution. The second term, $\bar{Q}_2^0$, represents the first effect of non-random orientation on the pure conduction contribution[2]. Finally, the first flow-induced term, $\bar{Q}_1^0$, occurs at $0\left(\mathrm{Pe}_1\dfrac{\gamma}{D}\right)$. It is significant that the flow-induced contribution to $\bar{Q}_y$ is identically zero

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[2] It may be noted that the $0\left(\dfrac{\gamma}{D}\right)$ departure from a random orientation distribution produces no pure conduction contribution to $\bar{Q}_y$.

in the limit of a purely random orientation distribution. Detailed expressions for the various coefficients $\bar{Q}_i^j$ in (3.14) may be found in McMillen's Ph. D. thesis (1976).

In addition to its effect on the orientation distribution function, rotary Brownian motion also contributes to the bulk conductive heat flux in a more direct way. Physically, this direct effect is due to the random particle rotations acting on the mean temperature field in the presence of an orientation distribution function $N(\theta_1, \phi_1)$ which is not uniform. In the low Pe limit considered here, we model this direct contribution by including an effective angular velocity of the particles which is associated with the diffusion process across statistical population gradients in the orientation probability space. A similar direct effect has also been included in most (though not all) calculations of the bulk stress in suspensions (cf. HINCH and LEAL, 1972, or the recent review by BRENNER, 1972). It should be noted, however, that the existence of a direct Brownian motion contribution has never been conclusively justified to the best of our knowledge. We are presently attempting to provide such a justification for the thermal conductivity problem. For present purposes we shall follow the preponderance of *prior* workers and include the direct contribution without rigorous justification. Following the above discussion, we thus calculate the contribution to Q_y resulting from an effective particle angular velocity, $-D\nabla(\log N)$. To $0(\text{Pe})$, the microscale temperature distribution induced by this motion is uncoupled from that which arises due to the hydrodynamically induced particle rotation in the absence of Brownian motion effects. The resulting expression for Q_y, when averaged in the manner of equation (3.12), has the general form

$$(3.15) \qquad \bar{\bar{Q}}_y = -k_1 \alpha \Phi \left\{ \text{Pe}_1 \left(\frac{\gamma}{D} \right) \bar{\bar{Q}}_1^0(m, \tau, \Gamma_p) + 0 \left(\text{Pe}_1 \left(\frac{\gamma}{D} \right)^2 \right) \right\}.$$

The overall bulk conductive heat flux in the cross-flow direction is thus

$$(3.16) \qquad Q_y = \bar{Q}_y + \bar{\bar{Q}}_y.$$

The behavior of Q_y is best depicted by considering the various terms in (3.14) and (3.15) individually. We begin with $\bar{Q}_0^0/(-k_1 \alpha)$ which is plotted in Fig. 4 as a function of Γ_p for various values of m. Of particular note is the strong dependence of the magnitude of this random orientation, pure conduction contribution to Q_y on the value of m, for moderate values of Γ_p, and the rapid approach, for small to intermediate values of m, to its asymptotic value for $\Gamma_p \to \infty$. In fact, when $m = 10$, the value of $(-\bar{Q}_0^0/(k\alpha)$ for $\Gamma_p = 20$ is already within 4% of its final, large particle aspect ratio value, 4.09. Finally, it may be noted that the contribution $(\bar{Q}_0^0/-k\alpha)$ becomes very large when both m and Γ are large. For m large, but fixed, and $\Gamma_p \to \infty$, the limiting value is $m/3$, independent of Γ_p. On the other hand, for Γ_p large but fixed, and $m \to \infty$, we obtain the limiting value $\frac{4}{3}\Gamma_p^2$, independent of m. We now turn to the term $(-\bar{Q}_0^2/k_1 \alpha)$ which represents the first influence of nonrandom orientation on the pure conduction contribution to the effective conductivity. The most striking feature in this case is the very small magnitudes, and very weak dependence on either m or Γ_p below $m \sim 10$. For $m < 1$, there is a very small negative value which increases to zero for $m = 1$ and then begins again to decrease to
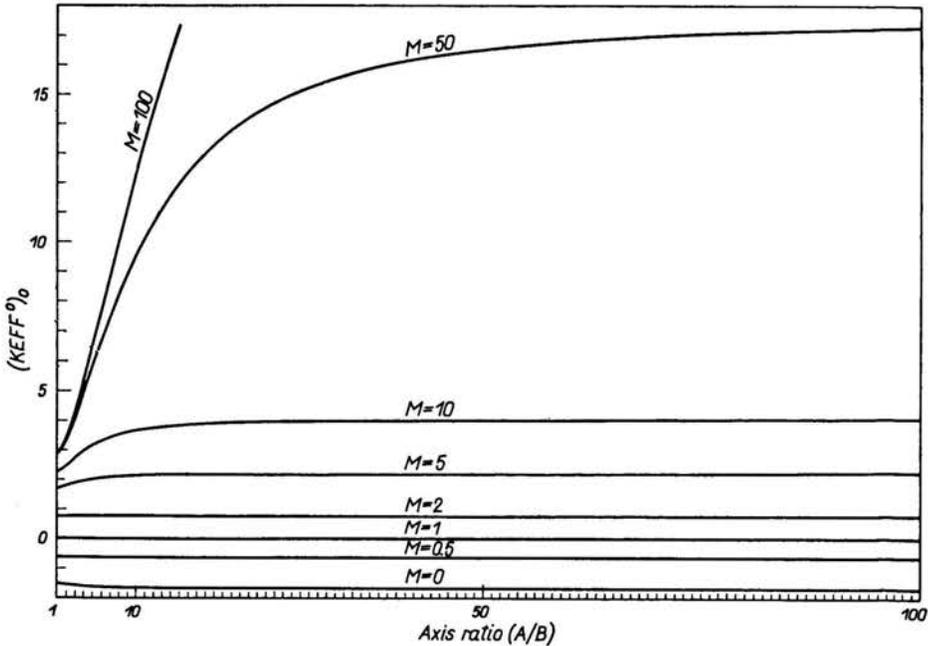
FIG. 4. The pure conduction, random orientation, contribution $\bar{Q}_0^0/(-k\alpha)$ as a function of the particle axis ratio $\Gamma_p = A/B$, for various values of the conductivity ratio $m = k_2/k_1$.

negative values. For $m > 10$, where the change is significant, this behavior simply reflects the decreasing fraction of particles which are aligned in the cross-flow (y) direction as the hydrodynamic induced rotation begins to have a significant influence on the orientation distribution function. As shown earlier by HINCH and LEAL (1973), alignment in the flow direction becomes increasingly significant as Γ_p is increased, all else being equal, thus accounting for the increasing large negative values of $(-\bar{Q}_2^0/\alpha k_1)$ as Γ_p is increased. Finally, we have also obtained expressions for $\bar{Q}_1^1$ and $\bar{\bar{Q}}_1^1$. The general form in both cases

$$(3.17) \quad \bar{Q}_1^1 + \bar{\bar{Q}}_1^1 = \text{Pe}_1 \left(\frac{\gamma}{D}\right)(m-1)\left\{\left(\frac{\alpha_1}{(m+A_1)} + \frac{\alpha_2}{m+A_2} + \frac{\alpha_3}{(m+A_1)(m+A_2)} + \frac{\alpha_4}{(m+A_1)^2}\right.\right.$$

$$\left. + \frac{\alpha_5}{(m+A_2)^2}\right)\frac{\tau}{m} + \left(\frac{\alpha_6}{(m+A_1)} + \frac{\alpha_7}{(m+A_2)} + \frac{\alpha_8}{(m+A_1)(m+A_2)} + \frac{\alpha_9}{(m+A_1)^2}\right.$$

$$\left. + \frac{\alpha_{10}}{(m+A_2)^2}\right)\tau + \frac{\alpha_{11}}{(m+A_1)} + \frac{\alpha_{12}}{(m+A_2)} + \frac{\alpha_{13}(1-m)}{(m+A_1)(m+A_2)} + \frac{\alpha_{14}(1-m)}{(m+A_1)^2} + \left.\frac{\alpha_{15}(1-m)}{(m+A_2)^2}\right\}.$$

Here, A_1, A_2 and the α_i's are shape factors, with A_1 and A_2 always > 0, and the α_i's $= 0$ when $\Gamma_p = 1$. Unfortunately, the shape factors α_i are extremely complicated functions of Γ_p and thus they have not yet been evaluated numerically. However, it can be seen that the complete flow-induced contribution (3.17) vanishes for $m = 1$, as was also true for the 0(Pe) contributions in the case of a spherical drop. Furthermore, in the limit $m \to 0$, a finite contribution of $0\left(\frac{\gamma}{D}\text{Pe}_2\right)$ will be obtained. Finally, it may be noted that
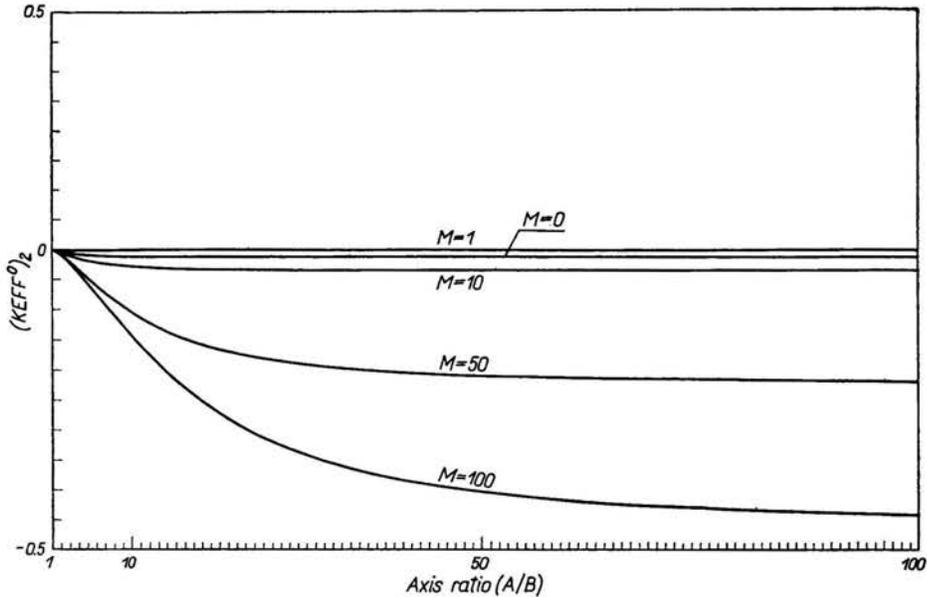
FIG. 5. The pure conduction, non-random orientation, contribution $\bar{Q}_0^2/(-k\alpha)$ as a function of the particle axis ratio $\Gamma_p = A/B$, for various values of the conductivity ratio $m = k_2/k_1$.

the Brownian motion and hydrodynamic contributions to Q_y both require nonuniform orientation distribution to be effective, i.e., the terms are $0(\gamma/D)$ in both cases. More detailed results based on (3.17) will be given by McMILLEN (1976).

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