

Modelling of porous media by renormalization of the Stokes equations

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The permeability of a random array of fixed spheres has been calculated over the range of volume fractions from dilute to almost closest packing, by assuming pairwise-additive (low-Reynolds-number) hydrodynamic interactions within an effective medium. Non-convergent pair interactions arising from the long-range decay of the Stokeslet were removed by renormalizing the Stokes equation to determine the permeability of the effective medium, i.e. to include the mean screening effect of the other spheres. Pair interactions in this Brinkman medium were calculated by the method of reflections in the far field and boundary collocation in the near field.

The permeability predicted by the theory asymptotes correctly to established results for dilute arrays, and compares favourably (within 15%) with the Carman correlation for volume fractions between 0.3 and 0.5. The magnitude also falls within the range of exact results for periodic arrays at the higher concentrations, but our model does not reproduce the dependence on structure.

Use of the Brinkman equation with an effective viscosity leads to an apparent slip velocity at the boundary of a porous medium. Our calculation of the bulk stress via volume averaging determines the effective viscosity and hence the slip coefficient unambiguously for dilute porous media. However, at concentrations corresponding to the available experimental results the lengthscale characterizing pressure or velocity gradients becomes comparable to the interparticle spacing, and the averaging technique fails. Indeed the Brinkman equation itself is no longer valid.

1. Introduction

In this work we have calculated the permeability and bulk stress for flow through a random array of fixed spheres over the entire range of concentrations by extending the dilute-limit theory of Hinch (1977) to account for multiparticle hydrodynamic interactions. Our approach treats interactions as pairwise-additive within an effective medium whose properties follow from the dilute theory. The effective medium rigorously incorporates the dominant, long-range many-body hydrodynamic interactions, as do self-consistent field theories. Consequently, the predictions for the permeability compare favourably with the experimental correlation of Carman (1937) in the range of concentrations relevant to real systems. In addition, the truncation at the pair, rather than the single-particle, level preserves the exact dilute limit to second order in the concentration.

Problems involving multiparticle interactions cannot be solved exactly. Consequently a number of approximate techniques have emerged, among which the more

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popular are cell models (reviewed by Happel & Brenner 1965), self-consistent-field models (Hashin 1964) and pairwise-additive interaction models (Glendinning & Russel 1982). We present here an alternative approach that is based on rigorous averaged equations developed by Hinch (1977) for dilute systems. Our modification closes the hierarchy of equations arising from the averaged-equation formalism by using an effective medium at the two-particle fixed level. The method is shown first for the permeability problem and then applied to the bulk stress produced by a velocity gradient.

Brinkman (1947) pioneered the modelling of porous media via fixed arrays, by equating the average drag per volume, $\langle \mathbf{f} \rangle(\mathbf{x})$, to the product of drag per sphere and the number density n :

$$\langle \mathbf{f} \rangle(\mathbf{x}) = -\mathbf{F}n = \mu\alpha^2 \langle \mathbf{v} \rangle(\mathbf{x}). \quad (1.1)$$

The permeability $k = \alpha^{-2}$ follows by definition from (1.1) since \mathbf{F} is linear (Stokes flow) with respect to the viscosity μ and the average velocity $\langle \mathbf{v} \rangle(\mathbf{x})$. The essence of Brinkman's argument was that a test sphere at \mathbf{x}_1 sees its surroundings as an effective medium of permeability k , with the governing equation obtained by adding a Darcy resistance term to the Stokes and continuity equations for incompressible viscous flow:

$$-\nabla p + \mu \nabla^2 \mathbf{v} - \mu\alpha^2 \mathbf{v} = \mathbf{0}, \quad \nabla \cdot \mathbf{v} = 0. \quad (1.2)$$

The drag on the sphere follows from the solution of (1.2) as

$$\mathbf{F} = 6\pi\mu a [1 + \alpha a + \frac{1}{3}(\alpha a)^2] \langle \mathbf{v} \rangle(\mathbf{x}_1). \quad (1.3)$$

Finally, k for the effective medium was determined by the *ad hoc* self-consistent condition, which combines (1.1) and (1.3) to obtain

$$(\alpha a)^2 = \frac{2}{3}cB_0(\alpha a), \quad (1.4)$$

with $B_0(x) = 1 + x + \frac{1}{3}x^2$ and the volume fraction given by $c = \frac{4}{3}\pi a^3 n$.

A rigorous justification of Brinkman's analysis is possible for small c (Childress 1972; Howells 1974; Hinch 1977; Freed & Muthukumar 1978; Muthukumar & Freed 1979). The procedure is non-trivial beyond the isolated-sphere analysis, because the 'obvious' method of adding the contribution of pair interactions $F_2(\mathbf{x}_1; \mathbf{x}_2)$ to the drag on an isolated test sphere, i.e.

$$-\mathbf{F}(\mathbf{x}_1) = -6\pi\mu a \mathbf{v}(\mathbf{x}_1) + \int_{|\mathbf{x}_2 - \mathbf{x}_1| \geq 2a} P(\mathbf{x}_2 | \mathbf{x}_1) F_2(\mathbf{x}_1; \mathbf{x}_2) d\mathbf{x}_2^3, \quad (1.5)$$

leads to non-convergent integrals since F_2 decays as $|\mathbf{x}_2 - \mathbf{x}_1|^{-1}$. Even in the dilute limit, the two steps (1.1) and (1.3) cannot be decoupled, as shown by Hinch (1977), because of the strong far-field interactions. Far from the test particle at \mathbf{x}_1 , the particle at \mathbf{x}_2 acts as a point force. In a process Hinch defines as renormalization, such non-convergent far-field interaction terms can be incorporated into a new effective porous medium surrounding the test particle at \mathbf{x}_1 . The Fourier-transform approach taken by Freed and Muthukumar (1978, 1979), in their 'long-wavelength limit', also yields (1.3) for small c . Their technique introduces an effective medium, but the connection between the two techniques is obscured by the different mathematical approaches. In principle, their analysis can be extended to higher concentrations, but requires substantial complex algebra.

The details of the renormalization and the calculations for the permeability are presented in §2. The results for the permeability are shown in §2.2. The techniques for solving the boundary-value problems that arise from the pair interactions are

presented in the companion paper (Kim & Russel 1985, hereinafter referred to as I). Our calculations for concentrated arrays require these complete results for pair interactions in a Brinkman medium rather than the asymptotic (small- c) expressions used in the dilute-limit theories.

In §3 we present the parallel development for the bulk stress when the medium is subject to a velocity gradient ∇v . The discussion is motivated by the popular use of the Brinkman equation in the modelling of porous medium–fluid interfaces (Adler & Mills 1979; Koplik, Levine & Zee 1983), where the ratio of the Brinkman viscosity and the solvent viscosity appears as a key parameter. Currently, the various theories for this ratio (Freed & Muthukumar 1979; Koplik *et al.* 1983) disagree. For $c/(\alpha a)^3 \gg 1$, i.e. dilute arrays, our technique yields the appropriate Brinkman viscosity from the bulk stress as a monotonically increasing function of c . However, at higher concentrations the Brinkman approach cannot be justified rigorously because the velocity gradient cannot be maintained over a region that is sufficiently large compared with the particle size for the averaging procedure to be valid. Consequently, the boundary region between a porous material and a viscous fluid requires separate characterization, with the slip coefficient proposed by Beavers & Joseph (1967) being the simplest possibility.

2. The permeability of the concentrated array

In this section we review Hinch's (1977) rigorous averaged-equation formalism and introduce our approximations to calculate the permeability of a fixed array at arbitrary concentrations. At dilute concentrations our approximations become exact and our theory recovers the proper limit to $O(c^2)$. At higher concentrations the connection between our theory and Brinkman's *ad hoc* model of a single test sphere in an effective medium (i.e. a self-consistent-field model) appears in a natural way.

In the first step of the averaged equation formalism, the Newtonian constitutive equation for the fluid stress σ and the Stokes and continuity equations governing the pressure p and velocity v in an incompressible fluid are averaged over ensembles containing stationary particles. The resulting averaged equations have the same functional form, but with an extra term $\mathbf{s}(\mathbf{x})$ due to the particles:

$$\langle \sigma \rangle(\mathbf{x}) = -\langle p \rangle(\mathbf{x}) + 2\mu \langle \epsilon \rangle(\mathbf{x}) + \langle \mathbf{s} \rangle(\mathbf{x}),$$

with the rate-of-strain tensor denoted by ϵ and

$$\nabla \cdot \langle \sigma \rangle + \langle \mathbf{f} \rangle = \mathbf{0}, \quad \nabla \cdot \langle v \rangle = 0.$$

The external body force \mathbf{f} acts on the spheres to keep them fixed. The generalized function $\mathbf{s}(\mathbf{x})$ vanishes in the fluid (where the constitutive equation for σ is valid) and increments the fluid constitutive equation inside the particles to the correct value. Thus $\langle \mathbf{s}(\mathbf{x}) \rangle$ represents the particle contribution to the stress.

If the solid's constitutive equation is provided, the expression for \mathbf{s} may be written immediately. But in this paper we model the particles as rigid bodies, so the particle stresses are indeterminate, requiring a different approach as discussed later in this section.

For dilute systems the calculation proceeds through a hierarchy of conditionally averaged equations; the total contribution from the particles can be expressed in terms of the conditional average with one particle fixed, which in turn can be expressed in terms of a conditional average with two particles fixed, and so forth. The hierarchy is constructed so that an additional factor $O(c)$ appears at each level,

i.e. in the relationship between the N -particle-fixed and $(N+1)$ -particle-fixed problems.

For dilute systems a valid perturbation expansion can be constructed in principle by starting at the N -particle-fixed problem. Unfortunately, detailed solutions of the N -particle problem are limited to $N \leq 2$. Not surprisingly, explicit examples in Hinch's (1977) paper are limited to the one-particle-fixed problems (which require knowledge of the two-particle solution as discussed below). In comparison, our theory is essentially a 'two-particle self-consistent-field' approach. Consequently, we also start with the rigorous, conditionally averaged equation with one particle fixed, and introduce reasonable approximations to account for the other particles.

The conditionally averaged equation with a particle fixed at \mathbf{x}_1 is

$$-\nabla p(\mathbf{x} | \mathbf{x}_1) + \mu \nabla^2 \mathbf{v}(\mathbf{x} | \mathbf{x}_1) + \mathbf{f}(\mathbf{x} | \mathbf{x}_1) = -\nabla \cdot \boldsymbol{\sigma}(\mathbf{x} | \mathbf{x}_1).$$

For convenience we have dropped angular brackets from the field variables. For rigid particles the right-hand side must be manipulated (see Hinch (1977) for details and Kim (1983) for the case where \mathbf{f} is non-zero) into the following equivalent form, which requires knowledge of the stresses only at the particle surface:

$$\begin{aligned} & -\nabla p(\mathbf{x} | \mathbf{x}_1) + \mu \nabla^2 \mathbf{v}(\mathbf{x} | \mathbf{x}_1) \\ &= \int_{|\mathbf{x}_2 - \mathbf{x}_1| \geq 2a} d\mathbf{x}_2^3 P(\mathbf{x}_2 | \mathbf{x}_1) \oint_{|\mathbf{x}_2 - \mathbf{x}'| = a} dS' \boldsymbol{\sigma}(\mathbf{x}' | \mathbf{x}_1, \mathbf{x}_2) \cdot \mathbf{n}' \delta(\mathbf{x}' - \mathbf{x}). \end{aligned} \quad (2.1)$$

The forcing in (2.1) represents the disturbance, over all allowed configurations of the second test sphere, from the stress distribution on the surface of the second sphere. This is an exact expression since $\boldsymbol{\sigma}(\mathbf{x}' | \mathbf{x}_1, \mathbf{x}_2)$ is the exact conditionally averaged stress distribution with two spheres fixed (the arguments $\mathbf{x}_1, \mathbf{x}_2, \dots$ after a vertical bar indicate that the field variables are conditionally averaged over all ensembles with particles at those points). Others (e.g. Howells 1974) have used this equation as the starting point without a formal derivation. The physical interpretation of (2.1) is that the conditionally averaged pressure and velocity fields with one particle fixed must satisfy the Stokes and continuity equation, but with apparent volume forces due to the other particles. The one-particle-fixed conditional average may be related to a two-particle conditional average (this is really the definition of conditional averaging). Equation (2.1) confirms the intuitive notion that the effect of the other particles may be related *exactly* to the integral of $\boldsymbol{\sigma}(\mathbf{x} | \mathbf{x}_1, \mathbf{x}_2) \cdot \mathbf{n}$ over the surface of a test sphere at \mathbf{x}_2 . We then integrate over all permissible \mathbf{x}_2 using the conditional probability $P(\mathbf{x}_2 | \mathbf{x}_1)$ that a second test sphere is indeed at \mathbf{x}_2 .

Now we digress briefly to review Hinch's results. In the analysis of the dilute limit, non-convergent integrals appear in the solution of (2.1) if the two-sphere input on the right-hand side is approximated to $O(1)$ by taking $\boldsymbol{\sigma}(\mathbf{x}' | \mathbf{x}_1, \mathbf{x}_2)$ as the stress field for two spheres in the pure solvent. Hinch (1977) showed that the convergence problem does not occur if the spheres are placed in an effective medium with properties accurate to $O(c)$. The procedure, which Hinch calls *renormalization*, is accomplished mathematically by placing additional terms on the left-hand side of (2.1) so that the homogeneous equation describes the effective medium. The equality is preserved by modifying the right-hand side with a distribution of singularities that are mathematically equal to the new terms on the left-hand side.

The elimination of the non-convergent integrals may be traced to two distinct factors; (1), the right-hand side of the renormalized problem decays more rapidly than in the original problem and, (2), the particle-particle interactions are weaker

in the effective medium. As discussed by Hinch (1977), the first factor alone ensures convergence in the bulk stress (Batchelor & Green 1972*b*) and sedimentation (Batchelor 1972) problems, whereas the second factor plays the crucial role in the porous medium. The elegant aspect of Hinch's theory becomes quite apparent in the computational process, where one finds that in all problems the distribution of singularities that arises from the intuitive modification toward the effective medium are precisely those that cancel the divergent terms in the two-sphere solution. Stated in another way, the non-convergent terms in the far field were never there, but arose because the effective medium between two widely separated test spheres was erroneously approximated as the pure solvent.

For concentrated arrays, such rigorous analysis is not feasible, but we shall take a similar approach and conjecture that the (unknown) conditionally averaged problem with two spheres fixed may be approximated by two spheres interacting through an effective medium. In the dilute theory, the single-particle solution is used to determine the effective-medium parameters. Here, motivated by the dilute analysis, we assume that the effective medium for the two-sphere problem can be determined from a convergence requirement in the far field. Thus (2.1) is renormalized as

$$\begin{aligned}
 & -\nabla p(\mathbf{x} | \mathbf{x}_1) + \mu \nabla^2 v(\mathbf{x} | \mathbf{x}_1) - \mu \bar{\alpha}^2 v(\mathbf{x} | \mathbf{x}_1) \\
 &= \int_{|\mathbf{x}_2 - \mathbf{x}_1| \geq 2a} d\mathbf{x}_2^3 \left\{ P(\mathbf{x}_2 | \mathbf{x}_1) \oint_{|\mathbf{x}_2 - \mathbf{x}'| = a} dS' \boldsymbol{\sigma}(\mathbf{x}' | \mathbf{x}_1, \mathbf{x}_2) \cdot \mathbf{n}' \delta(\mathbf{x}' - \mathbf{x}) \right. \\
 &\quad \left. - \mu \bar{\alpha}^2 v(\mathbf{x}_2 | \mathbf{x}_1) \delta(\mathbf{x} - \mathbf{x}_2) \right\} - \int_{a \leq |\mathbf{x}_2 - \mathbf{x}_1| \leq 2a} d\mathbf{x}_2^3 \mu \bar{\alpha}^2 v(\mathbf{x}_2 | \mathbf{x}_1) \delta(\mathbf{x} - \mathbf{x}_2), \quad (2.2)
 \end{aligned}$$

with the boundary conditions

$$v(\mathbf{x} | \mathbf{x}_1) = 0 \quad \text{at } |\mathbf{x} - \mathbf{x}_1| = a, \quad v(\mathbf{x} | \mathbf{x}_1) \rightarrow v(\mathbf{x}_1) \quad \text{as } |\mathbf{x} - \mathbf{x}_1| \rightarrow \infty.$$

The new term on the left-hand side of (2.2) converts the solvent into a Brinkman medium with a permeability $\bar{\alpha}^{-2}$ (as yet to be determined). The equivalent distribution of monopoles on the right-hand side has simply been written in terms of the Dirac delta function.

The problem has thus been reduced to calculating the drag on a test sphere at \mathbf{x}_1 governed by (2.2). As in the dilute case, the explicit calculation of the velocity field $v(\mathbf{x} | \mathbf{x}_1)$ itself can be avoided through use of the analogue for the Brinkman equation to Faxén's (1922) law for the Stokes equation, which gives the drag on a test sphere of radius a centred at \mathbf{x}_1 in the ambient field $v(\mathbf{x}_1)$ as $6\pi\mu a(1 + \frac{1}{6}a^2 \nabla^2) v^\infty(\mathbf{x})|_{\mathbf{x}=\mathbf{x}_1}$. Howells (1974) has shown that, for the Brinkman equation, the Stokesian factor $6\pi\mu a(1 + \frac{1}{6}a^2 \nabla^2)$ should be replaced by

$$6\pi\mu a \{ B_0(\alpha a) + B_2(\alpha a) a^2 \nabla^2 \}, \quad (2.3)$$

with
$$B_0(x) = 1 + x + \frac{1}{3}x^2, \quad B_2(x) = \frac{e^x - B_0(x)}{x^2}.$$

For the non-homogeneous equation (2.2), $v^\infty(\mathbf{x})$ includes both the imposed velocity and the velocity field generated by the singularities. The latter is readily obtained by replacing the delta functions in (2.2) with the Green dyadic (derived by Howells 1974) $\mathcal{G}(\mathbf{x}; \bar{\alpha})/8\pi\mu$.

Thus the velocity

$$v(\mathbf{x}_1) - \int_{|\mathbf{x}_2 - \mathbf{x}_1| \geq 2a} d\mathbf{x}_2^3 \left\{ P(\mathbf{x}_2 | \mathbf{x}_1) \left[\oint_{|\mathbf{x}_2 - \mathbf{x}'| = a} dS' \sigma(\mathbf{x}' | \mathbf{x}_1, \mathbf{x}_2) \cdot \mathbf{n}' \cdot \frac{\mathcal{F}(\mathbf{x}' - \mathbf{x}; \bar{\alpha})}{8\pi\mu} \right] - \mu \bar{\alpha}^2 v(\mathbf{x}_2 | \mathbf{x}_1) \cdot \frac{\mathcal{F}(\mathbf{x} - \mathbf{x}_2; \bar{\alpha})}{8\pi\mu} \right\} + \int_{a \leq |\mathbf{x}_2 - \mathbf{x}_1| \leq 2a} d\mathbf{x}_2^3 \mu \bar{\alpha}^2 v(\mathbf{x}_2 | \mathbf{x}_1) \cdot \frac{\mathcal{F}(\mathbf{x} - \mathbf{x}_2; \bar{\alpha})}{8\pi\mu}. \quad (2.4)$$

can be used to obtain the following expression for the drag on the test sphere at \mathbf{x}_1 :

$$F = 6\pi\mu a B_0(\bar{\alpha}a) v(\mathbf{x}_1) + \int_{|\mathbf{x}_2 - \mathbf{x}_1| \geq 2a} d\mathbf{x}_2^3 \left\{ -P(\mathbf{x}_2 | \mathbf{x}_1) F_2(\mathbf{x}_1; \mathbf{x}_2; \bar{\alpha}) + \mu \bar{\alpha}^2 v(\mathbf{x}_2 | \mathbf{x}_1) 6\pi\mu a \{ B_0(\bar{\alpha}a) + B_2(\bar{\alpha}a) a^2 \nabla^2 \} \frac{\mathcal{F}(\mathbf{x} - \mathbf{x}_2; \bar{\alpha})}{8\pi\mu} \right\} \Bigg|_{\mathbf{x} = \mathbf{x}_1} + \int_{a \leq |\mathbf{x}_2 - \mathbf{x}_1| \leq 2a} d\mathbf{x}_2^3 \mu \bar{\alpha}^2 v(\mathbf{x}_2 | \mathbf{x}_1) 6\pi\mu a \{ B_0(\bar{\alpha}a) + B_2(\bar{\alpha}a) a^2 \nabla^2 \} \frac{\mathcal{F}(\mathbf{x} - \mathbf{x}_2; \bar{\alpha})}{8\pi\mu} \Bigg|_{\mathbf{x} = \mathbf{x}_1} \quad (2.5)$$

The functional form of the terms due to $v(\mathbf{x}_1)$ and the distribution are in accordance with the above explanation. Although not as straightforward, the contribution from the surface distribution $\sigma(\mathbf{x}' | \mathbf{x}_1, \mathbf{x}_2)$ is precisely $F_2(\mathbf{x}_1; \mathbf{x}_2; \bar{\alpha})$, the difference between the two-particle and one-particle drags, since the Faxén law for the drag on sphere 1 with another sphere at \mathbf{x}_2 requires that sphere 2 be represented by precisely the surface distribution of singularities on the right-hand side of (2.2).

In the far field the method of reflections determines the excess drag $F_2(\mathbf{x}_1; \mathbf{x}_2; \bar{\alpha})$ to leading order as

$$v(\mathbf{x}_1) \cdot 6\pi\mu a B_0(\alpha a) \left[\frac{3}{4} a B_0(\bar{\alpha}a) \mathcal{F}(\mathbf{x} - \mathbf{x}_2; \bar{\alpha}) \right]. \quad (2.6)$$

This term, the contribution from the first reflection, decays as $|\mathbf{x}_2 - \mathbf{x}_1|^{-3}$ and must be cancelled by the corresponding component in the subtraction term. Therefore the permeability parameter for the effective medium must be

$$(\bar{\alpha}a)^2 = \frac{3}{2} c B_0(\bar{\alpha}a). \quad (2.7)$$

This resembles Brinkman's closure scheme for a test sphere in an effective medium. However, in our approach the effective medium applies to both the two-particle-fixed problem and the inhomogeneous equation governing the one-particle-fixed problem. Thus this approach preserves the near-field interactions not considered by conventional self-consistent-field theories. $F(\mathbf{x}_1)$, the drag on the test sphere at \mathbf{x}_1 as determined by (2.5), and local homogeneity determine the permeability as

$$\mu/k = P(\mathbf{x}_1) F(\mathbf{x}_1). \quad (2.8)$$

Readers familiar with Hinch's (1977) paper may note that our (2.2) has fewer terms than his (8.2). The answer is found in the fact that in the dilute-limit theory the renormalization is dictated solely by the far-field region. There, altering the near-field behaviour, for example with viscosity jumps, will change the relative contributions from various physical effects and the remainder integral, without changing the final result. Thus the extra terms in Hinch's (8.2) reduce the contribution from the remainder integral or, equivalently, the far-field interactions. A thorough analysis of the change in the relative contributions from the near-field and remainder terms as the near field is modified is presented in Kim (1983).

At higher concentrations the modification of the near-field medium (left-hand side

of (2.2)) the distributions of singularities on the right-hand side are not necessarily equivalent. Furthermore, ‘jumps’ in the effective-medium parameters, as used in the dilute-limit theories, are no longer tractable mathematically. We extend the effective medium into the surface of the spheres and thereby eliminate the additional terms.

An alternative rearrangement is presented in Kim (1983). In an attempt to reduce the contribution from the remainder integral, i.e. to include a better approximation of the far field, a distribution of degenerate quadrupoles was placed on the right-hand side of (2.2). In the dilute theory, the associated viscosity modification

$$\bar{\mu} = \mu[1 + \frac{5}{2}cB_2(\bar{\alpha}a)]^{-1} \sim \mu(1 - \frac{5}{2}c) \quad \text{as } \bar{\alpha} \rightarrow 0$$

exactly compensates the modification of the far-field contribution. However, at higher concentrations, $\bar{\mu}/\mu \ll 1$ leads to substantial underestimation of the stress in the near field, and consequently an unrealistically low prediction for the drag. In fact, at separations of less than a diameter other spheres are excluded from the gap, and the pair interact much as if in the pure solvent, rather than the effective medium. We conclude that (2.2) is the best renormalization with constant effective parameters, i.e. the best compromise between the constraints imposed by the near-field and far-field interactions.

2.1. Asymptotic and numerical calculations for the drag

We can now calculate the permeability. At a given concentration (2.7) determines the renormalization. The symmetry of the two-sphere geometry permits the terms in the integrand of (2.5) to be expressed in terms of uniform streams parallel and perpendicular to the sphere–sphere axis \mathbf{R} . For example,

$$\mathbf{F}_2(\mathbf{x}_1; \mathbf{x}_2; \bar{\alpha}) = 6\pi\mu a \mathbf{v}(\mathbf{x}_1) \cdot [X_F(R; \bar{\alpha}) \mathbf{R}\mathbf{R} + Y_F(R; \bar{\alpha}) (\boldsymbol{\delta} - \mathbf{R}\mathbf{R})]. \tag{2.9}$$

For convenience, we will define \mathbf{G} as the subtraction terms, and write a similar decomposition

$$\mathbf{G}(\mathbf{x}_1; \mathbf{x}_2; \bar{\alpha}) = 6\pi\mu a \mathbf{v}(\mathbf{x}_1) \cdot [X_G(R; \bar{\alpha}) \mathbf{R}\mathbf{R} + Y_G(R; \bar{\alpha}) (\boldsymbol{\delta} - \mathbf{R}\mathbf{R})].$$

Equation (2.5) then becomes

$$\begin{aligned} \mathbf{F} &= 6\pi\mu a B_0(\bar{\alpha}a) \mathbf{v}(\mathbf{x}_1) \\ &\quad - \int_{|\mathbf{x}_2 - \mathbf{x}_1| \geq 2a} d\mathbf{x}_2^3 \{P(\mathbf{x}_2 | \mathbf{x}_1) \mathbf{F}_2(\mathbf{x}_1; \mathbf{x}_2; \bar{\alpha}) - P(\mathbf{x}_1) \mathbf{G}(\mathbf{x}_1; \mathbf{x}_2; \bar{\alpha})\} \\ &\quad + \int_{a \leq |\mathbf{x}_2 - \mathbf{x}_1| \leq 2a} d\mathbf{x}_2^3 P(\mathbf{x}_1) \mathbf{G}(\mathbf{x}_1; \mathbf{x}_2; \bar{\alpha}) \\ &= 6\pi\mu a B_0(\bar{\alpha}a) \mathbf{v}(\mathbf{x}_1) \\ &\quad - \int_{|\mathbf{x}_2 - \mathbf{x}_1| \geq 2a} d\mathbf{x}_2^3 \{P(\mathbf{x}_2 | \mathbf{x}_1) - P(\mathbf{x}_1)\} \mathbf{F}_2(\mathbf{x}_1; \mathbf{x}_2; \bar{\alpha}) \\ &\quad - 6\pi\mu a P(\mathbf{x}_1) \mathbf{v}(\mathbf{x}_1) \frac{4}{3}\pi \int_{2a}^{\infty} [(X_F - X_G) + 2(Y_F - Y_G)] R^2 dR \\ &\quad + 6\pi\mu a P(\mathbf{x}_1) \mathbf{v}(\mathbf{x}_1) \frac{4}{3}\pi \int_a^{2a} [X_G + 2Y_G] R^2 dR. \tag{2.10} \end{aligned}$$

Since the integrals converge, the angular integrations that were performed in arriving at (2.10) are rigorously justified.

There are no known analytic solutions to the Brinkman equation for the drag on two interacting spheres, since separation of variables fails for the Helmholtz equation in bispherical coordinates. However, if the two spheres are far apart, i.e. $R/a \gg 1$, accurate results can be obtained by the method of reflections, as shown in I. The integral in (2.10) can therefore be evaluated analytically in the far field.

In the near field the contributions from the algebraically complex higher reflections are no longer negligible, necessitating a numerical approach. Fortunately, the boundary collocation technique, which was developed by Gluckman, Pfeffer & Weinbaum (1971) for hydrodynamic interaction between spheres in Stokes flow, can be readily modified for the Brinkman equation. Details of the numerical procedure may be found in I. At $R = 5a$ the values for $X_F(R; \alpha)$ and $Y_F(R; \alpha)$ obtained from the method of reflections (4 reflections) and the collocation technique (12 points) agreed to five significant figures over the range of interest for α .

2.2. *Results for the permeability*

The permeability predicted by our model is examined in this subsection. In order to facilitate comparison with other work, we will present the result in terms of $F(\mathbf{x}_1)$, the drag on the test sphere at \mathbf{x}_1 , which is related to the permeability through

$$\mu/k = P(\mathbf{x}_1) F(\mathbf{x}_1). \tag{2.11}$$

Figure 1 compares our predictions of $F(\mathbf{x}_1)$ with that predicted by Hinch's dilute-limit expansion and the Carman (1937) correlation. At small c , where the permeability can be calculated analytically, our approach recovers Hinch's results. We will show the procedure to exhibit the contributions from the various terms. For $c \ll 1$, (2.7) for $\bar{\alpha}$ becomes

$$\bar{\alpha}a = \frac{3}{\sqrt{2}}c^{\frac{1}{2}} + \frac{9}{4}c + \dots \quad \text{or} \quad \mu\bar{\alpha}^2 = 6\pi\mu a P(\mathbf{x}_1) + \dots \tag{2.12}$$

Equation (2.5) can be rearranged by separating the contribution from the third reflection, a valid procedure because this contribution is convergent when inverted with the Brinkman operator. The result is

$$\begin{aligned} F &= 6\pi\mu a B_0 v(\mathbf{x}_1) \\ &+ \int_{|\mathbf{x}_2 - \mathbf{x}_1| \geq 2a} d\mathbf{x}_2^3 \{ -P(\mathbf{x}_2 | \mathbf{x}_1) F_2 + 6\pi\mu a P(\mathbf{x}_1) v(\mathbf{x}_2 | \mathbf{x}_1) \cdot \frac{3}{4}a(B_0 + B_2 a^2 \nabla_2^2)^2 \mathcal{F} \\ &+ P(\mathbf{x}_1) (\frac{3}{4}a B_0)^3 6\pi\mu a B_0 v(\mathbf{x}_1) \cdot \mathcal{F}^3 \} \\ &+ \int_{a \leq |\mathbf{x}_2 - \mathbf{x}_1| \leq 2a} d\mathbf{x}_2^3 6\pi\mu a B_0 P(\mathbf{x}_1) v(\mathbf{x}_2 | \mathbf{x}_1) \cdot (B_0 + B_2 a^2 \nabla_2^2) \frac{3}{4}a \mathcal{F} \\ &- \int_{|\mathbf{x}_2 - \mathbf{x}_1| \geq 2a} d\mathbf{x}_2^3 6\pi\mu a P(\mathbf{x}_1) v(\mathbf{x}_2 | \mathbf{x}_1) \cdot \frac{3}{4}a(B_0 + B_2 a^2 \nabla_2^2) B_2 a^2 \nabla_2^2 \mathcal{F} \\ &- \int_{|\mathbf{x}_2 - \mathbf{x}_1| \geq 2a} d\mathbf{x}_2^3 P(\mathbf{x}_1) (\frac{3}{4}a B_0)^3 6\pi\mu a B_0 v(\mathbf{x}_1) \cdot \mathcal{F}^3. \end{aligned} \tag{2.13}$$

At small c , $6\pi\mu a B_0 v(\mathbf{x}_1)$ from the homogeneous solution becomes

$$6\pi\mu a v(\mathbf{x}_1) [1 + \frac{3}{\sqrt{2}}c^{\frac{1}{2}} + \frac{15}{4}c + \dots].$$

The distribution of monopoles in the excluded volume (second integral in (2.13)) contributes

$$\frac{89}{64}c,$$

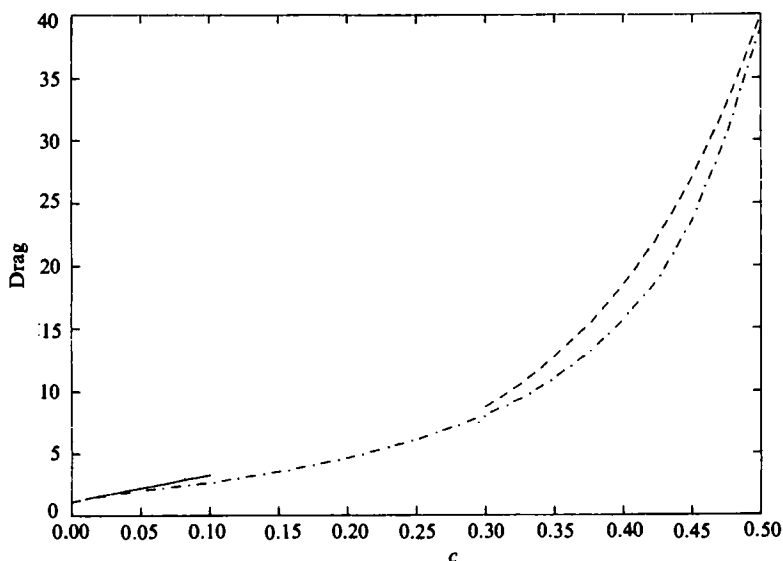


FIGURE 1. Comparison of the drag on a test sphere in a porous medium as a function of concentration c : —, dilute theory; ----, Carman correlation; - · - · -, equation (2.10).

the distribution of quadrupoles in $|\mathbf{x}_2 - \mathbf{x}_1| \geq 2a$ (the ∇_2^2 term in the third integral in (2.13))

$$-\frac{43}{64}c - \frac{171}{64\sqrt{2}}c^{\frac{3}{2}},$$

and the third reflection,

$$\frac{135}{64}c \log c + 11.987c + \frac{405}{16\sqrt{2}}c^{\frac{3}{2}} \log c + 63.231c^{\frac{3}{2}}.$$

The higher-order terms (underlined) prove useful later in estimating the region of validity of the dilute limit expansion. The leading terms sum to

$$1 + \frac{3}{\sqrt{2}}c^{\frac{1}{2}} + \frac{135}{64}c \log c + \underline{16.456c}, \tag{2.14}$$

which is the result obtained by Howells (1974) and Hinch (1977).† The remaining terms converge even if inverted with the Stokes operator. Under these circumstances, the leading-order result with the Brinkman operator coincides with the result obtained from the Stokes operator. Therefore the remainder integral asymptotes to Hinch's remainder integral.

As shown in tables 1 (a, b), the higher-order contributions from the third reflection and quadrupole distribution are significant. They are less than one-tenth of the leading-order term only for extremely small values of c (4.5×10^{-5} for the third reflection and 1.26×10^{-3} for the quadrupole distribution). The result is a slowly converging series, with a limited domain of applicability for any truncation.

We now return to the comparison between our theory and the Carman correlation. At these higher concentrations, the contribution from the homogeneous solution

† There are computational errors in Hinch's (1977) calculation of the contributions from his surface distribution of monopoles and dipoles. On page 718 the factors of $-(171/1024)c$ and $(125/2048)c$ should be $(27/512)c$ and $-(125/512)c$ respectively. We note that these corrections eliminate the discrepancy between his work and Howells (1974) that was mentioned at the end of his paper.

(a)			
		Analytical expansion	
	Numerical	one-term	two-term
Homogeneous solution	1.07103	1.07083	—
Monopole in excluded volume	1.505×10^{-3}	1.391×10^{-3}	—
Integral of the third reflection	-4.903×10^{-3}	-2.584×10^{-3}	-4.494×10^{-3}
Integral of the quadrupole distribution	-7.340×10^{-4}	-6.719×10^{-4}	-7.316×10^{-4}
Remainder integral	-1.819×10^{-2}	—	—
Integral of the subtraction term in (2.5)	2.672×10^{-2}	—	—
Method of reflection ($5, \infty$)	7.114×10^{-4}	—	—

(b)			
		Analytical expansion	
	Numerical	one-term	two-term
Homogeneous solution	1.25665	1.24963	—
Monopole in excluded volume	1.764×10^{-2}	1.391×10^{-2}	—
Integral of the third reflection	-2.247×10^{-2}	-2.273×10^{-2}	-3.534×10^{-2}
Integral of the quadrupole distribution	-8.847×10^{-3}	-6.719×10^{-3}	-8.608×10^{-3}
Remainder integral	-0.1432	—	—
Integral of the subtraction term in (2.5)	0.2138	—	—
Method of reflection ($5, \infty$)	4.256×10^{-3}	—	—

TABLE 1. Contributions of the terms in (2.13) at (a) $c = 0.001$ and (b) $c = 0.01$

c	Homogeneous term	Total
0.05	1.76	1.99
0.10	2.40	2.60
0.15	3.18	3.48
0.20	4.21	4.61
0.25	5.62	6.08
0.30	7.67	8.11
0.35	10.78	11.05
0.40	15.86	15.65
0.45	24.95	23.54
0.50	43.63	39.04
$\frac{2}{3}$	∞	∞

TABLE 2. The contribution from the homogeneous solution

dominates those from the right-hand-side forcing, as shown in table 2. Since the former corresponds to Brinkman's self-consistent-field result, we essentially recover his successful comparison with the Carman correlation. The fact that the error term (the right-hand-side forcing) makes only a small contribution confirms the validity of replacing the sphere-sphere interactions with a self-consistent medium. This also implies that our model will exhibit a weak dependence on structure.

c	Random array	Periodic arrays		
		SC	BCC	FCC
0.25	6.08	8.91	9.31	9.37
0.30	8.11	11.8	12.4	12.6
0.35	11.05	15.9	16.9	17.3
0.40	15.65	20.8	22.7	23.5
0.45	23.54	28.1	31.7	33.5
0.50	39.04	36.5	43.6	47.5

TABLE 3. A comparison of the drag on a test sphere (scaled by Stokes coefficient) in the random array and periodic arrays (values from graph of Zick & Homsy 1982)

Comparison of our result for random arrays with the results obtained by Zick & Homsy (1982) for periodic arrays (table 3) illustrate this last point. In the dilute limit the drag exhibits different concentration dependence in the two systems: $c^{\frac{1}{2}}$ for random arrays and $c^{\frac{3}{2}}$ for periodic arrays. As discussed by Saffman (1973), this difference is due to the existence of the lengthscale associated with the unit cell in the periodic system. However, at higher concentrations, one might expect qualitatively similar results, since the lengthscale becomes $O(a)$ in both cases. The results in table 3 support this expectation, but also demonstrate the striking difference between the various types of packings for the periodic arrays.

Our model random array yields no analogous effect, because the term that depends on structure, i.e. the term that contains the pair-correlation function

$$P(\mathbf{x}_2 | \mathbf{x}_1) - P(\mathbf{x}_1)$$

in (2.10), makes only a small contribution. We suspect that the introduction of structure in the one-particle-fixed medium through a variable permeability reflecting the short-range structure would improve the present model, but at the cost of much greater mathematical complexity. Thus we attribute the effect of structure to short-range correlations rather than the difference in the far field between random and periodic arrays.

3. The bulk stress at porous-medium–fluid boundaries

A survey of recent literature shows that the Brinkman equation is also used as a generalization of Darcy's law which allows the matching of velocities and tractions at the boundary between fluid and porous media. Examples are Adler & Mills' (1979) model for floc rupture and the analysis of shear flow at the porous-media–fluid boundary by Koptik *et al.* (1983).

The motivation becomes apparent when one considers the boundary conditions at the interface between a fluid region and a porous medium governed by Darcy's law. Since the governing equations in the two regions are not of the same order, one cannot match all components of the velocity. In particular, the tangential component must be discontinuous; indeed experimental measurements of the apparent slip velocity, the difference between the velocity in the adjacent fluid region and the superficial velocity deep inside the Darcy medium, are available (Beavers & Joseph 1967).

Modelling the porous medium by the Brinkman equation avoids difficulties by retaining the second-order viscous-stress terms. However, the question of the proper viscosity for the Brinkman medium then arises, for, unlike the Darcy equation, the

viscosity appears as a parameter independent of the permeability. Brinkman (1947) recognized this problem, and suggested the possible use of Einstein's (1906) expression, but also provided *ad hoc* momentum-transfer arguments favouring the use of the pure-solvent viscosity. It has been recognized (Kim 1983; Koplik *et al.* 1983) that if one assumes that the $\nabla^2 v$ term in the Brinkman equation has a coefficient differing from that in the Stokes equation (i.e. the effective viscosity of the Brinkman medium differs from that of the pure solvent) then the 'viscosity ratio' can be correlated to the slip parameter of Beavers & Joseph (1967), as illustrated in §3.1.

3.1. A model problem

In this subsection we review a model problem corresponding to several experimental studies, and compare the results from the Darcy and Brinkman analyses. Beavers & Joseph (1967) have conducted experimental studies of Poiseuille flow in a channel with a permeable boundary. The geometry, as shown in figure 2, consists of an impermeable upper wall at $y = h$ and a nominal boundary between the fluid and porous medium at $y = 0$.

Beavers & Joseph described the motion in the channel by the Stokes equation and that in the porous medium by Darcy's law:

$$\frac{d^2 v}{dy^2} = \frac{1}{\mu} \frac{dp}{dx} \quad (y > 0, \text{ channel}), \quad (3.1a)$$

$$V = -\frac{k}{\mu} \frac{dp}{dx} \quad (y < 0, \text{ porous medium}), \quad (3.1b)$$

with no slip at $y = h$ and a slip boundary condition at $y = 0$:

$$\frac{dv}{dy} = \frac{\lambda}{k^{\frac{1}{2}}} (v_s - V). \quad (3.2)$$

The dimensionless slip parameter λ should depend only on the properties of the porous medium.

The fractional increase in mass-flow rate through the channel relative to that with impermeable walls was measured and compared with the expression that follows from the solution of the velocity field as

$$\Phi = \frac{3(\sigma + 2\lambda)}{\sigma(1 + \lambda\sigma)} \quad (3.3)$$

with $\sigma = h/k^{\frac{1}{2}}$. Using λ as an empirical parameter, they successfully correlated their experimental data.

Application of the model with the Brinkman equation in the porous medium eliminates the slip condition at $y = 0$. The velocity fields in the two regions are now governed by

$$\frac{d^2 v}{dy^2} = \frac{1}{\mu} \frac{dp}{dx} \quad (y > 0, \text{ channel}), \quad (3.4a)$$

$$\frac{d^2 V}{dy^2} - \frac{V}{k} = -\frac{1}{\mu} \frac{dp}{dx} \quad (y < 0, \text{ porous medium}), \quad (3.4b)$$

with the boundary conditions

$$\begin{aligned} v &= 0 & \text{at } y &= h, \\ v &= V, \quad \mu \frac{dv}{dy} = \bar{\mu} \frac{dV}{dy} & \text{at } y &= 0. \end{aligned}$$

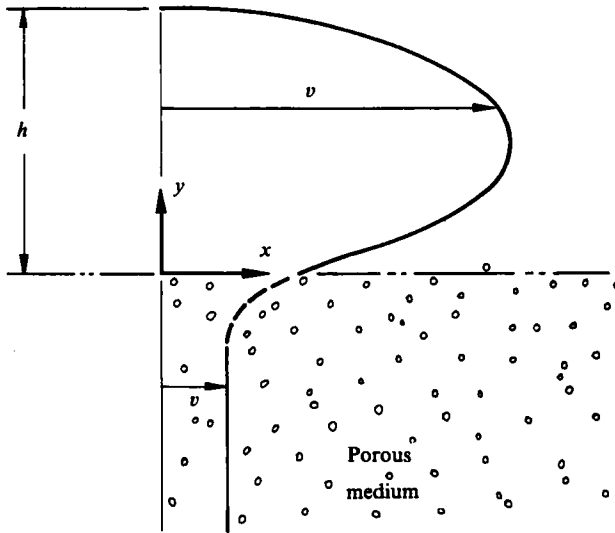


FIGURE 2. Flow geometry in the experiments of Beavers & Joseph (1967).

The resulting velocity profile determines the excess mass-flow rate as

$$\frac{3(\sigma + 2)\mu/\bar{\mu}}{\sigma\{\sigma + \mu/\bar{\mu}\}} \tag{3.5}$$

with $\sigma = h/\bar{k}^2$. We relate \bar{k} to k_0 , the permeability based on the solvent viscosity used by Beavers & Joseph (1967), by

$$\frac{\mu}{k_0} = \frac{\bar{\mu}}{\bar{k}},$$

so that (3.5) becomes

$$\Phi = \frac{3[\sigma + 2(\bar{\mu}/\mu)^{\frac{1}{2}}]}{\sigma[1 + \sigma(\bar{\mu}/\mu)^{\frac{1}{2}}]}, \quad \sigma = h/k_0^{\frac{1}{2}}, \tag{3.6}$$

which is identical with (3.3) with $\lambda = (\bar{\mu}/\mu)^{\frac{1}{2}}$. Thus the two approaches, the Darcy equation with a slip condition or the Brinkman equation, lead to similar end results. We now turn our attention to the calculation of the viscosity ratio in the Brinkman approach.

3.2. The bulk stress in the fixed array

Recognition that the Brinkman viscosity and the solvent viscosity may differ and could be related to the slip coefficient has led to a number of attempts to derive an analogue to Einstein's expression for the effective viscosity. The effective-medium theory of Freed & Muthukumar (1978) predicts the effective viscosity for dilute arrays as

$$\frac{\bar{\mu}}{\mu} = 1 + \frac{5}{2}c - \frac{9c^{\frac{3}{2}}}{2\sqrt{2}}, \tag{3.7}$$

which exceeds that for the pure solvent, but is less than the Einstein result. Koplik *et al.* (1983) calculated the energy dissipation in an extensional flow about an isolated stationary sphere to determine the effective viscosity as

$$\frac{\bar{\mu}}{\mu} = 1 - \frac{1}{2}c, \tag{3.8}$$

a value less than that for the pure solvent. In the following, we derive the Brinkman viscosity through a rigorous calculation of the bulk stress to settle the disagreement.

The advantage of determining the bulk stress rather than the energy dissipation has been discussed extensively for force-free suspensions, most notably in Landau & Lifshitz (1959) and Batchelor (1970). The energy-dissipation approach is particularly ambiguous for fixed arrays, since it is not obvious how to distinguish the energy dissipation due to the bulk straining from that due to the frictional drag.

First, we note some limitations on our rigorous approach. The volume averaging performed in the previous sections requires that the macroscale, on which velocity and pressure gradients vary, greatly exceed the microscale. Only then can a representative averaging volume with intermediate dimension l contain many particles, i.e. $nl^3 \gg 1$. A linear shear flow persists in a porous material only near a boundary or in the presence of a quadratically varying pressure field, either of which can invalidate the volume averaging (as pointed out by E. J. Hinch).

To demonstrate the consequences consider a homogeneous material with a uniform velocity gradient $\nabla \mathbf{v}$ sustained by the pressure field $p = -\mu\alpha^2 \mathbf{x} \cdot \nabla \mathbf{v} \cdot \mathbf{x}$. For pressure variations to remain insignificant within the representative volume, i.e. small relative to the viscous stresses,

$$\frac{\mu\alpha^2 l^2 |\nabla \mathbf{v}|}{\mu |\nabla \mathbf{v}|} \ll 1.$$

This means that $n^{-1/3} \ll l \ll \alpha^{-1}$, which is possible only for $cl/\alpha a \gg 1$. This is possible only at dilute concentrations where αa is $O(c^2)$. Analysis of flow near a boundary leads to the same conclusion.

The consequences are significant:

- (1) the Brinkman equation itself is valid only for dilute porous media;
- (2) for non-dilute systems ($(x/\alpha a)^3 < O(1)$) Darcy's law prevails in the bulk with a modified boundary condition necessary to account for gradients near a boundary (e.g. Saffman 1971; Ross 1983).

Detailed analyses of the region near a boundary, as yet unavailable, should provide a slip coefficient, characteristic of the porous material, coupling the interior Darcy flow to the Stokes flow in an adjacent fluid, as proposed by Saffman (1971).

We therefore calculate the bulk stress of a dilute porous medium satisfying $(\alpha a)^3 \ll c$ by determining the dipoles induced by a local velocity gradient $\nabla \mathbf{v}(\mathbf{x}_1)$. Following Batchelor (1970), the contribution of the rigid particle to the stress, $\Sigma^{(P)}$, is determined by applying the divergence theorem to a volume average of the stress field:

$$\Sigma_{ij}^{(P)} = P(\mathbf{x}_1) \int_{S_P} \sigma_{ik} n_k x_j \, dA - P(\mathbf{x}_1) \int_{V_P} \left(\frac{\partial \sigma_{ik}}{\partial x_k} \right) x_j \, dV. \quad (3.9)$$

with $\nabla \cdot \boldsymbol{\sigma} = -\mathbf{f}$. The second term in (3.9) would not be present for force-free particles. The force distribution \mathbf{f} has the zeroth moment \mathbf{F}^{ext} , which keeps the particles fixed, and (for rigid particles) an indeterminate dipole moment. However, only the surface moment in (3.9) is significant physically, since only that term interacts with the velocity gradient. Thus the problem becomes that of computing the stress dipoles,

$$\mathbf{S} = \frac{1}{2} \int_{S_P} [(\boldsymbol{\sigma} \cdot \mathbf{n}) \mathbf{x} + \mathbf{x}(\boldsymbol{\sigma} \cdot \mathbf{n})] \, dA \quad \text{and} \quad \mathbf{T} = \frac{1}{2} \int_{S_P} [(\boldsymbol{\sigma} \cdot \mathbf{n}) \mathbf{x} - \mathbf{x}(\boldsymbol{\sigma} \cdot \mathbf{n})] \, dA,$$

with the velocity gradient decomposed into rate-of-strain \mathbf{E} and vorticity $\boldsymbol{\Omega}$ fields. We shall consider the rate-of-strain case first.

The isolated-particle case is straightforward, and, as in the permeability problem, will appear as the homogeneous solution in the pair-interaction problem. With the two spheres in the pure solvent, the pair-interaction contribution to the dipoles, $\mathbf{S}_2(\mathbf{x}_1; \mathbf{x}_2)$, decays as R^{-1} . This differs from the R^{-3} result for force-free suspensions (Batchelor & Green 1972*a*). In the fixed array the sphere at \mathbf{x}_2 produces a monopole field, so that the excess stresslet \mathbf{S}_2 decays as

$$\mathbf{S}_2(\mathbf{x}_1; \mathbf{x}_2; \boldsymbol{\theta}) = \frac{20}{3}\pi\mu a^3 \left\{ \frac{3}{4}a\boldsymbol{\theta} \cdot (\mathbf{x}_2 - \mathbf{x}_1) \right\} \cdot \frac{1}{2}[\nabla\mathcal{J}(\mathbf{x} - \mathbf{x}_2) + \nabla\mathcal{J}(\mathbf{x} + \mathbf{x}_2)^T]. \quad (3.10)$$

The fact that this non-convergent interaction cannot be renormalized with a bulk stress correction, i.e. a viscosity change, indicates a fundamental difference between fixed arrays and force-free suspensions. The far-field behaviour exhibited in (3.10) suggests the Brinkman renormalization (2.2), with $(\bar{a}a)^2 = \frac{2}{3}cB_0(\bar{a}a)$ as before.

The details of the velocity field may be bypassed by using the appropriate Faxén law. The Brinkman stresslet is obtained by applying $\frac{20}{3}\pi\mu a^3 \{C_0(\alpha a) + C_2(\alpha a)a^2\nabla^2\}$ to the ambient rate-of-strain field and the rate-of-strain field forced by the right-hand-side singularities in (2.2), with

$$C_0(x) = \frac{1+x+\frac{2}{5}x^2+\frac{1}{15}x^3}{1+x} \quad \text{and} \quad C_2(x) = \frac{e^x - (1+x)C_0(x)}{(1+x)x^2}.$$

The stresslet from the renormalized problem is

$$\begin{aligned} \mathbf{S} = & \frac{20}{3}\pi\mu a^3 C_0(\bar{a}a) \boldsymbol{\theta}(\mathbf{x}_1) + \int_{|\mathbf{x}_2 - \mathbf{x}_1| \geq 2a} d\mathbf{x}_2^3 \{P(\mathbf{x}_2 | \mathbf{x}_1) \mathbf{S}_2(\mathbf{x}_1; \mathbf{x}_2; \bar{\boldsymbol{\alpha}}) \\ & + \mu\bar{\boldsymbol{\alpha}}^2 v(\mathbf{x}_2 | \mathbf{x}_1) \frac{5}{12}a^3 (C_0 + C_2 a^2 \nabla^2) (\nabla\mathcal{J} + \nabla\mathcal{J}^T) |_{\mathbf{x} - \mathbf{x}_1} \\ & + \int_{a \leq |\mathbf{x}_2 - \mathbf{x}_1| \leq 2a} d\mathbf{x}_2^3 \mu\bar{\boldsymbol{\alpha}}^2 v(\mathbf{x}_2 | \mathbf{x}_1) \frac{5}{12}a^3 (C_0 + C_2 a^2 \nabla^2) (\nabla\mathcal{J} + \nabla\mathcal{J}^T) |_{\mathbf{x} - \mathbf{x}_1}. \end{aligned} \quad (3.11)$$

The far-field behaviour of \mathbf{S}_2 is determined by the method of reflections as $\frac{20}{3}\pi\mu a^3 \{C_0(\alpha a) + C_2(\alpha a)a^2\nabla^2\}$ operating on the rate-of-strain fields of the first, second and third reflection velocities from sphere 2. The leading-order terms in these velocities are monopoles and dipoles:

$$\begin{aligned} & -\frac{3}{4}a(B_0 + B_2 a^2 \nabla^2) (\boldsymbol{\theta} \cdot (\mathbf{x}_2 - \mathbf{x}_1)) \cdot \mathcal{J}(\mathbf{x} - \mathbf{x}_2) + \frac{5}{8}a^3 (C_0 + C_2 a^2 \nabla^2) \boldsymbol{\theta} \cdot \nabla\mathcal{J}(\mathbf{x} - \mathbf{x}_2), \\ \text{(2nd reflection)} \quad & -\frac{3}{4}a(B_0 + B_2 a^2 \nabla^2) \frac{5}{8}a^3 (C_0 + C_2 a^2 \nabla^2) \boldsymbol{\theta} \cdot \nabla\mathcal{J}(\mathbf{x}_2 - \mathbf{x}_1) \cdot \mathcal{J}(\mathbf{x} - \mathbf{x}_2) \\ \text{(3rd reflection)} \quad & + (-\frac{3}{4}aB_0)^3 \boldsymbol{\theta} \cdot (\mathbf{x}_2 - \mathbf{x}_1) \cdot \mathcal{J} \cdot \mathbf{I} \cdot \mathcal{J}(\mathbf{x} - \mathbf{x}_2). \end{aligned} \quad (3.12)$$

The underlined terms are cancelled by the subtraction terms, leaving a convergent integral in (3.11).

The axisymmetry about the sphere-sphere axis \mathbf{R} allows us to write the linear relation between the stresslet and the ambient rate-of-strain in terms of three scalar functions (Brenner 1972):

$$\begin{aligned} S_{ij} = & \frac{3}{2}X_S(R_i R_j - \frac{1}{3}\delta_{ij}) (R_k R_l - \frac{1}{3}\delta_{kl}) e_{kl}(\mathbf{x}_1) \\ & + \frac{1}{2}Y_S(R_i \delta_{jl} R_k + R_j \delta_{il} R_k + R_i \delta_{jk} R_l + R_j \delta_{ik} R_l - 4R_i R_j R_k R_l) e_{kl}(\mathbf{x}_1) \\ & + \frac{1}{2}Z_S(\delta_{ik} \delta_{jl} + \delta_{jk} \delta_{il} - \delta_{ij} \delta_{kl} + R_i R_j \delta_{kl} + \delta_{ij} R_k R_l \\ & - R_i \delta_{jl} R_k - R_j \delta_{il} R_k - R_i \delta_{jk} R_l - R_j \delta_{ik} R_l + R_i R_j R_k R_l) e_{kl}(\mathbf{x}_1). \end{aligned} \quad (3.13)$$

Equation (3.13) is a rearrangement of the K , L and M decomposition of Batchelor & Green (1972*a*). We define our X , Y and Z functions so that they correspond to the

following canonical flows: axisymmetric extensional flow, hyperbolic straining in a plane containing the sphere–sphere axis and hyperbolic straining in the plane perpendicular to the sphere–sphere axis (respectively). We substitute this decomposition into (3.11) and, after angular integration, obtain

$$\begin{aligned}
 \mathbf{S} = & \frac{20}{3}\pi\mu a^3 C_0(\bar{a}a) \boldsymbol{\theta}(\mathbf{x}_1) + \int_{|\mathbf{x}_2-\mathbf{x}_1| \geq 2a} d\mathbf{x}_2^3 \{P(\mathbf{x}_2|\mathbf{x}_1) - P(\mathbf{x}_1)\} \mathbf{S}_2(\mathbf{x}_1; \mathbf{x}_2; \bar{a}) \\
 & + 4\pi\mu a^3 c \boldsymbol{\theta}(\mathbf{x}_1) \int_2^\infty [X_S - X_G] + 2(Y_S - Y_G) + 2(Z_S - Z_G)] R^2 dR \\
 & - 4\pi\mu a^3 c \boldsymbol{\theta}(\mathbf{x}_1) \int_1^2 [X_G - 2Y_G + 2Z_G] R^2 dR.
 \end{aligned} \tag{3.14}$$

The last two integrals are dimensionless, with R scaled by a . The subscript S denotes the stresslet and the subscript G again denotes the subtraction term.

The collocation solution for X , Y and Z and other details of the numerical work are discussed in I. As in the permeability problem, the method of reflections was used when the two results agreed to at least five significant figures. For small c , (3.14) reduces to

$$\begin{aligned}
 & 2\mu[1 + \frac{5}{2}c + (\frac{81}{32} \log c + 21.041) c^2 + O(c^{\frac{3}{2}} \log c)] \boldsymbol{\theta}(\mathbf{x}_1) \\
 & + P(\mathbf{x}_1) \int_{R \geq 2a} d\mathbf{x}_2^3 \{P(\mathbf{x}_2|\mathbf{x}_1) \mathbf{S}_2(\mathbf{x}_1; \mathbf{x}_2) - 5\mu c \mathbf{E}_1(\mathbf{x}_1; \mathbf{x}_2) \\
 & - 5\mu c (\frac{3}{4}a)^3 [\boldsymbol{\theta}(\mathbf{x}_1) \cdot (\mathbf{x}_2 - \mathbf{x}_1)] \cdot \mathcal{J}^2 \cdot \frac{1}{2} [\nabla_2 \cdot \mathcal{J}(\mathbf{x}_2 - \mathbf{x}_1) + \nabla_2 \cdot \mathcal{J}(\mathbf{x}_2 - \mathbf{x}_1)^T]\} \\
 & \sim [P(\mathbf{x}_1)]^{-1} 2\mu \{\frac{5}{2}c + (\frac{81}{32} \log c + 19.66) c^2 + 6.59c^{\frac{3}{2}} \log c\} \boldsymbol{\theta}(\mathbf{x}_1).
 \end{aligned} \tag{3.15}$$

$\mathbf{E}_1(\mathbf{x}_1; \mathbf{x}_2)$ is the rate of strain at \mathbf{x}_1 due to the disturbance field of sphere 2. As in the permeability problem, the $\log c$ factors originate for the third reflection.

We now determine the relation between the stress dipole \mathbf{T} and the vorticity field $\boldsymbol{\Omega}(\mathbf{x}_1)$. The procedure for \mathbf{T} is quite similar, and the details will not be repeated. As before, the velocity field may be bypassed by using a Faxén law for \mathbf{T} . This time we apply

$$\frac{4\pi\mu a^3 e^{\alpha a}}{1 + \alpha a}$$

to the ambient vorticity $\boldsymbol{\Omega}(\mathbf{x}_1)$ and the vorticity field driven by the right-hand-side singularities in (2.2). For small c the linear relation between \mathbf{T} and $\boldsymbol{\Omega}$ reduces to

$$\mathbf{T} = -4\pi\mu a^3 (1 + \frac{27}{4}c \log c) \boldsymbol{\Omega}(\mathbf{x}_1). \tag{3.16}$$

The bulk stress follows from (3.15) and (3.16) as

$$-p\boldsymbol{\delta} + 2\mu c \{\frac{5}{2} + (\frac{81}{32} \log c + 19.66) c + 6.59c^{\frac{3}{2}} \log c\} \boldsymbol{\theta}(\mathbf{x}_1) - 3\mu c (1 + \frac{27}{4}c \log c) \boldsymbol{\Omega}(\mathbf{x}_1).$$

Therefore our analysis predicts that the Brinkman viscosity is greater than the solvent viscosity. The coefficient for the rate-of-strain term agrees to $O(c)$ with that of Freed & Muthukumar (1978) and, consequently, not with Koplik *et al.* (1983).

The discussion of §3.1 implies that the slip parameter should satisfy $\lambda > 1$. Table 4 summarizes the experimental results of Beavers & Joseph (1967), most of which yield $\lambda < 1$. However, $c(k/a^2)^{\frac{1}{2}} \ll 1$ for all cases, so that the large velocity gradients near the boundary invalidate the averaging used in our continuum approach. Thus there are as yet no experiments at conditions for which the theory is valid and no theory relevant to the existing experiments.

System	λ	k/a^2	c	$c(k/a^2)^{\ddagger}$
A	0.78	0.23	0.21	0.023
B	1.45	0.21	0.22	0.021
C	4.00	0.25	0.20	0.025
Aloxite	0.1	0.024	0.44	0.0016
Aloxite	0.1	0.014	0.49	0.0008

TABLE 4. Comparison of the slip parameter λ (experiment of Beavers & Joseph 1967)

4. Conclusions

The effective permeability of an array of fixed spheres has been determined over concentrations ranging from dilute to almost closest packing by adapting rigorous techniques for renormalizing non-convergent interactions in dilute systems. The extension into higher concentrations has introduced a natural connection with the self-consistent-field approach. We have shown that the self-consistent component dominates, thereby both recovering and explaining Brinkman's (1947) successful comparison with the Carman correlation at the higher concentrations.

In the porous-medium-fluid boundary problem we have shown that the Brinkman approach is invalid for dense arrays. Unless $c/(\alpha a)^3 \gg 1$, the large gradients on the microscale invalidate the averaging. Nield (1983) has reached the same conclusion for the boundary conditions in the Rayleigh-Darcy convection problem. For dilute arrays we conclude that the bulk stress depends on both rate-of-strain and vorticity fields, with the Brinkman viscosity in the rate-of-strain problem agreeing to $O(c)$ with the work of Freed & Muthukumar (1977). Finally, we note that there are as yet no experiments at conditions for which the theory is valid and no predictive theory relevant to the existing experiments.

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REFERENCES

- ADLER, P. M. & MILLS, P. M. 1979 *J. Rheol.* **23**, 25.
 BATCHELOR, G. K. 1970 *J. Fluid Mech.* **41**, 545.
 BATCHELOR, G. K. 1972 *J. Fluid Mech.* **52**, 245.
 BATCHELOR, G. K. & GREEN, J. T. 1972a *J. Fluid Mech.* **56**, 375.
 BATCHELOR, G. K. & GREEN, J. T. 1972b *J. Fluid Mech.* **56**, 401.
 BEAVERS, G. S. & JOSEPH, D. C. 1967 *J. Fluid Mech.* **30**, 197.
 BRENNER, H. 1972 *Chem. Engng Sci.* **27**, 1069.
 BRINKMAN, H. C. 1947 *Appl. Sci. Res.* **A1**, 27.
 CARMAN, W. 1937 *Trans. Inst. Chem. Engrs* **15**, 150.
 CHILDRESS, S. 1972 *J. Chem. Phys.* **56**, 2527.
 EINSTEIN, A. 1906 *Ann. der Phys.* **19**, 289.
 FAXÉN, H. 1922 *Ark. Mat. Astron. Fys.* **17**, no. 1.
 FREED, K. F. & MUTHUKUMAR, M. 1978 *J. Chem. Phys.* **68**, 2088.

- GLENDINNING, A. B. & RUSSEL, W. B. 1982 *J. Coll. Interface Sci.* **89**, 124.
- GLUCKMAN, M. J., PFEFFER, R. & WEINBAUM, S. 1971 *J. Fluid Mech.* **50**, 705.
- HAPPEL, J. & BRENNER, H. 1965 *Low Reynolds Number Hydrodynamics*. Prentice-Hall.
- HASHIN, Z. 1964 *Appl. Mech. Rev.* **17**, 1.
- HINCH, E. J. 1977 *J. Fluid Mech.* **83**, 695.
- HOWELLS, I. D. 1974 *J. Fluid Mech.* **64**, 449.
- KIM, S. 1983 Ph.D. Dissertation, Princeton University.
- KIM, S. & RUSSEL, W. B. 1985 *J. Fluid Mech.* **154**, 253.
- KOPLIK, J., LEVINE, H. & ZEE, A. 1984 *Phys. Fluids* **26**, 2864.
- LANDAU, L. D. & LIFSHITZ, E. M. 1959 *Fluid Mechanics*. Pergamon.
- MUTHUKUMAR, M. & FREED, K. F. 1979 *J. Chem. Phys.* **70**, 5875.
- NIELD, D. A. 1983 *J. Fluid Mech.* **128**, 37.
- ROSS, S. M. 1983 *AIChE J.* **29**, 840.
- SAFFMAN, P. G. 1971 *Stud. Appl. Maths* **50**, 93.
- SAFFMAN, P. G. 1973 *Stud. Appl. Maths* **52**, 115.
- ZICK, A. A. & HOMS, G. M. 1982 *J. Fluid Mech.* **115**, 13.