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A new application of the reciprocity relations to the study of fluid flows through fixed beds

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Abstract. Creeping flow through an array of spheres with small volume fraction ϕ is studied theoretically. It is observed that it can be described macroscopically by Brinkman's equation. A generalized version of the reciprocity relations is used to determine the viscous term up to $O(\phi^2)$ for the case of random configuration and up to $O(\phi^3)$ for the case of periodic, cubic configurations of the fixed bed.

Key words: creeping flow, Brinkman's equation, porous media, array of spheres

1. Introduction

The objective of this work is to determine the effective equations describing the motion of a viscous incompressible fluid through a porous material. While at the microscale level the Navier–Stokes equations provide a complete description of the flow field, at the macroscale level the most commonly used equation is a simple empirical relation established by Darcy in 1856. In this, so-called, Darcy's law the mean pressure gradient is set to be proportional to the mean velocity, $\nabla\langle p\rangle = -(\mu/\kappa^*)\langle\mathbf{u}\rangle$, where μ is the fluid viscosity and κ^* defines the permeability of the porous medium. The validity of Darcy's law together with schemes to derive the value of the permeability have been presented by, among others, Sanchez–Palencia [1] and Keller [2], who used a multiple-scale technique to derive the averaged, or effective, equation, and Adler [3, pp.149–152], who employed a volume-averaging method.

Darcy's law seems to capture the main features of the macroscopic fluid flow; however, being first-order in the velocity, this equation is clearly incompatible with the existence of boundaries in the porous medium, where the no-slip condition must be satisfied. To resolve this paradox, a more general equation was proposed heuristically by Brinkman in 1947 by adding a viscous, second-order term, $\mu^*\nabla^2\langle\mathbf{u}\rangle$, to Darcy's law, where μ^* is some effective viscosity that is reduced to the fluid viscosity μ as the bed volume fraction ϕ tends to zero. In fact, Darcy's law can be viewed as a lower-order approximation of Brinkman's equation: the former determines the slowly-varying average velocity field far from the boundaries of the medium, while the viscous term in Brinkman's equation provides a correction, which is small in the bulk, and becomes appreciable only near the boundaries of the medium, where it allows the no-slip boundary condition to be satisfied [4, 5, 6]. Therefore, the viscous term can predict the existence of the boundary layer observed by Beavers and Joseph [7] and, far from being marginal, it is essential to predict most quantities of engineering interest. For example, in the case of a fluid flowing in a pipe filled with particles, the pressure drop and the heat- and mass-transfer coefficients are all strongly influenced, and actually sometimes determined uniquely, by the steep velocity profile near the walls that can be predicted only when the fluid flow is modelled through Brinkman's equation.

In the last twenty years, many investigators have rederived Brinkman's equation for small ϕ by averaging the Navier–Stokes equation of fluid motion, subjected to the appropriate no-slip boundary conditions on the surface of the bed particles. Among them, Hinch [8] determined Brinkman's bulk equations of motion in the dilute limit by averaging the transport equations, using a procedure that was later generalized by Kim and Russel [6] to account for multiparticle hydrodynamic interactions. For higher bed-volume fractions, however, this, like most averaging procedures, is not applicable near the external boundaries [6]. In this region, in fact, the typical length over which the velocity varies may become comparable with the typical particle–particle distance, thus invalidating the underlying assumption of separation between micro- and macro-scales, which is implicit in the averaging scheme.

Using a different approach, Rubinstein [9, 10] employed a multiple-scale technique that generalizes an approach originally proposed by Tam [11], showing convincingly that Brinkman's equation can be used even for porous media with low porosity, provided they have a very large number of scales. This seems to confirm the results of the Stokesian dynamics simulations performed by Durlovsky and Brady [12], who showed that Brinkman's equation describes qualitatively the behavior of the flow fields, even for large bed-volume fractions, although the value of the viscosity μ^* cannot be taken equal to the fluid viscosity μ.

In this work we determine the effective bulk equation of motion for flow through an array of fixed spheres located at either random or periodic positions. Details of the averaging process are presented in the next section, where the reciprocal theorem and the $O(\phi)$ results for the permeability and the Brinkman viscosity are derived. Then, in Section 3, these results are extended to higher orders in ϕ, while in Section 4 our findings are summarized and discussed.

2. The method of solution

2.1. Statement of the problem and scaling

Let us consider the steady, slow motion of a viscous fluid of viscosity μ and unit density, $\rho = 1$, through a fixed bed of spheres of radius a, located at positions $\mathbf{r}_N$. For low solid volume fractions the influence of the spheres can be modelled through singular multipole force distributions centered at $\mathbf{r}_N$, leading to the steady-state Stokes equations of motion and the continuity equation,

$$\nabla p - \mu \nabla^2 \mathbf{u} = \mathbf{F} = \sum_N \sum_m \nabla^m \delta(\mathbf{r} - \mathbf{r}_N)(\,\cdot\,)^m \mathbf{F}_N^{(m+1)}, \tag{1}$$

$$\nabla \cdot \mathbf{u} = 0, \tag{2}$$

where p is the pressure, $\mathbf{u}$ is the fluid velocity at the point $\mathbf{r}$, $\mathbf{F}$ is the generalized force per unit volume, $\mathbf{V}^m = \mathbf{V}\mathbf{V}\ldots\mathbf{V}$ (m times) represents the mth power of a vector $\mathbf{V}$, and the symbol $(\,\cdot\,)^m$ denotes successive dot multiplications in the order prescribed by the nesting convention. Here $\mathbf{F}_N^{(m)}$ denotes the strength of the mth multipole at the center of the Nth sphere, *i.e.* $F_i^{(1)}$ is the force exerted by the sphere on the fluid, $F_{ij}^{(2)}$ the corresponding moment of dipole, $F_{ijk}^{(3)}$ the moment of quadrupole, etc. Due to the linearity of the Stokes equations, these multipole strengths are in turn proportional to the nth gradients of the unperturbed velocity $\mathbf{u}_N$, that is the velocity field at $\mathbf{r}_N$ in the absence of the Nth sphere,

$$\mathbf{F}_N^{(m)} = -\sum_n \mathbf{R}_N^{(mn)}(\,\cdot\,)^n \nabla^{n-1}\mathbf{u}_N, \tag{3}$$

where $\mathbf{R}_N^{(mn)}$ is the single-particle grand resistance matrix of the Nth sphere, which includes the influence of the other spheres as well. In the following we shall assume that the averaging volume is sufficiently far from the boundaries of the medium that the grand resistance matrix is the same for each particle, so that the subscript N can be dropped from $\mathbf{R}_N^{(mn)}$.

Now we shall proceed to take the volume average of the Stokes equations over a domain τ comprising many particles and in which, at the same time, the unperturbed velocity and pressure fields do not vary appreciably. This, in essence, requires that a separation of scales exists, that is, the macroscale over which velocity and pressure gradient vary greatly exceeds the microscale, *e.g.* the typical particle-particle distance. Only in this case, in fact, an intermediate volume τ can be defined over which the averaging is performed. Although this averaging procedure can be performed rigorously by means of a multiple-scale expansion (see Bensoussan *et al.* [13], Sanchez-Palencia [14] and Mauri [15, 16], it will not be spelled out here, as it would unnecessarily make our analysis burdensome, without adding any new insight into the physical results. The main point is that, if a large number of particles is located within the averaging volume τ, the average, or macroscale, velocity and pressure field, $\langle \mathbf{u} \rangle$ and $\langle p \rangle$ are given by:

$$\langle \mathbf{u} \rangle = \langle \mathbf{u}_N \rangle, \qquad \langle p \rangle = \langle p_N \rangle, \tag{4}$$

where the bracket denotes volume average over τ. Using these definitions, we can easily show that

$$\sum_N \langle \nabla^m \delta(\mathbf{r} - \mathbf{r}_N)\mathbf{u}_N \rangle = (-1)^m \frac{3\phi}{4\pi a^3} \nabla^m \langle \mathbf{u} \rangle. \tag{5}$$

Finally, taking the average of the Stokes equations (1) and (2) and using the above relations (4) and (5), we find the following Brinkman-like equation:

$$\nabla_\imath \langle p \rangle = -\mu \kappa_{\imath\jmath}^{-1} \langle u_\jmath \rangle - 2\xi_{\imath kl} \nabla_k \langle u_l \rangle + \eta_{\jmath\imath kl} \nabla_\jmath \nabla_k \langle u_l \rangle, \tag{6}$$

$$\nabla_\imath \langle u_\imath \rangle = 0, \tag{7}$$

where $\kappa_{\imath\jmath}, \xi_{\imath\jmath k}$ and $\eta_{\imath\jmath kl}$ are the permeability, coupling and viscosity tensors, respectively, and further

$$\kappa_{\imath\jmath}^{-1} = \frac{3\phi}{4\pi a^3 \mu} R_{\imath\jmath}^{(11)}, \tag{8}$$

$$\xi_{\imath\jmath k} = \frac{3\phi}{8\pi a^3} (R_{\imath\jmath k}^{(12)} - R_{\jmath\imath k}^{(21)}), \tag{9}$$

$$\eta_{\imath\jmath kl} = \mu(\delta_{\imath k}\delta_{\jmath l} + \delta_{\imath l}\delta_{\jmath k}) + \frac{3\phi}{4\pi a^3}(R_{\imath\jmath kl}^{(22)} - R_{\jmath\imath kl}^{(13)} - R_{\imath k\jmath l}^{(31)}). \tag{10}$$

Since our averaging procedure implies a separation of scales, we have tacitly assumed that $|\mathbf{u}| \gg l|\nabla \mathbf{u}|$, where $l = \tau^{1/3}$ is the linear dimension of the averaging volume. Therefore, the three terms at the right-hand side of Equation (6) are of decreasing magnitude, thus justifying why we have neglected higher-order velocity-gradient terms. From here we see that the only components of the grand resistance matrix that are of relevance to us are $\mathbf{R}^{(11)}, \mathbf{R}^{(12)}, \mathbf{R}^{(21)}, \mathbf{R}^{(13)}, \mathbf{R}^{(31)}$ and $\mathbf{R}^{(22)}$.

Now, before we go further, let us consider an important symmetry property of the grand resistance matrix.

2.2. Symmetry relations of the grand resistance matrix

The average energy dissipated per unit time and volume, $\dot{E}$, is:

$$\dot{E} = 2\mu\langle \nabla\mathbf{u} \colon \nabla\mathbf{u}\rangle + \langle \mathbf{F}\cdot\mathbf{u}\rangle. \tag{11}$$

Now, substituting expression (1) for the generalized force **F** in (11), and applying (5), we obtain

$$\dot{E} - 2\mu\langle \nabla\mathbf{u} \colon \nabla\mathbf{u}\rangle = \frac{3\phi}{4\pi a^3}\sum_m (-1)^m \mathbf{F}^{(m+1)}(\,\cdot\,)^{m+1}\nabla^m\langle\mathbf{u}\rangle, \tag{12}$$

showing that $(m+1)$th pole strength $\mathbf{F}^{(m+1)}$ is conjugated with the mth mean velocity gradient. Therefore, since these two quantities are linearly related through (3), the Onsager relations state that the proportionality term, *i.e.* $\mathbf{R}^{(mn)}$, is a symmetric matrix. To exemplify what that means, let us rewrite (3) as

$$-F_i^{(1)} = R_{ik}^{(11)}u_k + R_{ijk}^{(12)}\nabla_j u_k + R_{ijkl}^{(13)}\nabla_j\nabla_k u_l, \tag{13}$$

$$-F_{ij}^{(2)} = R_{ijk}^{(21)}u_k + R_{ijkl}^{(22)}\nabla_k u_l, \tag{14}$$

$$-F_{ijk}^{(3)} = R_{ijkl}^{(31)}u_l, \tag{15}$$

where the subscripts N have been dropped from $\mathbf{F}^{(m)}$ and **u** for simplicity. In these equations we have not considered higher velocity-gradient terms, since we saw that they are not required to determine Brinkman's effective equations. Now, due to the symmetry of the grand resistance matrix, we find:

$$R_{ij}^{(11)} = R_{ji}^{(11)}, \tag{16}$$

$$R_{ijk}^{(12)} = R_{jki}^{(21)}, \tag{17}$$

$$R_{ijkl}^{(13)} = R_{jkli}^{(31)}, \tag{18}$$

$$R_{ijkl}^{(22)} = R_{klji}^{(22)}. \tag{19}$$

Applying these symmetry relations to definition (8), we see that the permeability tensor is identically symmetric, *i.e.* $\kappa_{ij} = \kappa_{ji}$, while (9) and (10) can be rewritten as

$$\xi_{ijk} = \frac{3\phi}{8\pi a^3}(R_{ijk}^{(12)} - R_{kji}^{(12)}), \tag{20}$$

$$\eta_{ijkl} = \mu(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}) + \frac{3\phi}{4\pi a^3}(R^{(22)}_{ijkl} - R^{(13)}_{jikl} - R^{(31)}_{lkij}). \tag{21}$$

These last expressions show that the third-order coupling tensor is antisymmetric with respect to its first and last indices, *i.e.* $\xi_{ijk} = -\xi_{kji}$, while the fourth-order viscosity tensor satisfies the following relation: $\eta_{ijkl} = \eta_{klij}$.

2.3. The leading-order averaged equations

At leading order, we neglect the influence of particle-particle interactions, so that we may determine the grand resistance matrix by studying the flow field around an isolated sphere. First, let us consider Faxen's law,

$$\mathbf{F}^{(1)}_N = -6\pi\mu a \left[1 + \frac{a^2}{6}\nabla^2\right]\mathbf{u}_N, \tag{22}$$

where $\mathbf{u}_N$ is the unperturbed fluid velocity, *i.e.* the velocity field at $\mathbf{r}_N$ in the absence of the Nth sphere. Now, comparing this relation with (13), we obtain

$$R^{(11)}_{ij} = 6\pi\mu a\delta_{ij}; \qquad R^{(12)}_{ijk} = 0; \qquad R^{(13)}_{ijkl} = \pi a^3\delta_{il}\delta_{jk}. \tag{23}$$

In addition[1], it is known that at leading order,

$$-\mathbf{F}^{(2)}_N = \frac{20}{3}\pi\mu a^3[\tfrac{1}{2}(\nabla\mathbf{u}_n + \nabla\mathbf{u}^\dagger_N)] + 4\pi\mu a^3[\tfrac{1}{2}(\nabla\mathbf{u}_n - \nabla\mathbf{u}^\dagger_N)], \tag{25}$$

where $\nabla\mathbf{u}^\dagger_N$ is the transposed of $\nabla\mathbf{u}_N$. Therefore we find

$$R^{(22)}_{ijkl} = \pi\mu a^3(\tfrac{16}{3}\delta_{ik}\delta_{jl} + \tfrac{4}{3}\delta_{il}\delta_{jk}). \tag{26}$$

Now, substituting (23) and (26) in (8), (9) and (10), we obtain:

$$\kappa^{-1}_{ij} = \frac{9\phi}{2a^2}\delta_{ij}, \tag{27}$$

$$\xi_{ijk} = 0, \tag{28}$$

$$\eta_{ijkl} = \mu(1 + \tfrac{5}{2}\phi)\delta_{ik}\delta_{jl} + \mu(1+\phi)\delta_{il}\delta_{jk}. \tag{29}$$

This shows that the flow of a Newtonian fluid through a dilute bed of solid spheres is described by the following Brinkman equation:

$$\langle\nabla p\rangle + \frac{\mu}{\kappa^*}\langle\mathbf{u}\rangle = \mu^*\nabla^2\langle\mathbf{u}\rangle, \qquad \nabla\cdot\langle\mathbf{u}\rangle = 0, \tag{30}$$

[1] Note that we can obtain the quadrupole strength by substituting (23) and (18) in (15), yielding:

$$F^{(3)}_{ijk} = -\pi\mu a^3\delta_{ij}u_k. \tag{24}$$

with permeability $\kappa^* = (2a^2)/(9\phi)$ and effective viscosity $\mu^* = \mu(1 + \frac{5}{2}\phi)$. Equation (30) shows that the viscous term in Brinkman's equation is expressed via the Einstein effective viscosity μ^*, in agreement with the result of Lundgren [17], Freed and Muthukumar [18] and Kim and Russel [6]. Not surprisingly, had the spherical particles been modelled as stokeslets, we would find [9, 10, 11] that Brinkman's viscosity equals the fluid viscosity μ.

Brinkman's equations can also be written as a momentum conservation equation, in terms of the mean force density **f** (*i.e.* the force per unit volume exerted by the spheres onto the fluid), the mean body-couple density **g** and the mean stress tensor **T** as:

$$\mathbf{f} + \tfrac{1}{2}\nabla \times \mathbf{g} + \nabla \cdot \mathbf{T} = \mathbf{0}, \qquad \nabla \cdot \langle \mathbf{u} \rangle = 0, \tag{31}$$

with the constitutive relations:

$$\mathbf{f} = -\frac{\mu}{\kappa^*}\langle \mathbf{u} \rangle, \tag{32}$$

$$\mathbf{g} = -\chi^* \langle \omega \rangle, \tag{33}$$

$$\mathbf{T} = -\langle p \rangle \mathbf{I} + 2\eta^* \langle \mathbf{S} \rangle, \tag{34}$$

where $\langle \mathbf{S} \rangle$ is the mean rate of strain and $\omega = \frac{1}{2}\nabla \times \langle \mathbf{u} \rangle$ is the mean angular velocity of the fluid. The effective quantities that appear in the constitutive relations (32), (33) and (34) are the permeability $\kappa^* = (2a^2)/(9\phi)$, the spin, or rotational, viscosity, $\chi^* = 6\mu\phi$, and the stress viscosity $\eta^* = \mu(1 + \frac{7}{4}\phi)$. It is important to note that the effective viscosity μ^* appearing in Brinkman's equation is different from the stress viscosity η^*, the latter being defined as the ratio between the symmetric part of the deviatoric stress tensor and the rate of shear. As explained by Kim and Russel [6], this is due to the fact that the quadrupole distribution contributes an extra term to the effective viscosity η_{ijkl}[2].

The approach described in this section can be generalized to the case of dilute beds of particles of any shape. In particular, it is important to note that, whenever the bed particles lack mirror symmetry, as for screw-like particles, fluid translational and angular velocities are coupled to each other, *i.e.* $\mathbf{R}_N^{(12)}$ is not identically zero, so that, if the particles have a preferential orientation, an extra term $2\xi_{ijk}\nabla_k u_j$ will appear in the Brinkman equation [see Equation (6)]. Actually, this term is even dominant with respect to the viscous term, although it is still small compared to the permeability term.

3. Higher-order approximation

In this section we calculate the higher-order terms in Brinkman's equation. Clearly, that means that we have to account for the interactions among the spheres, which, in turn, depend on the configuration in the fixed bed. Two cases are considered, where the identical spheres are assumed to be distributed either randomly or periodically in space.

[2] η^* stems from the part of η_{ijkl} that is symmetric with respect to both the first and the last two indices. If we rearrange Equation (29) as

$$\eta_{ijkl} = \eta^*(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}) + \tfrac{1}{8}\chi^*(\delta_{jk}\delta_{jl} - \delta_{il}\delta_{jk}), \tag{35}$$

we see that μ^* comes from the contribution of the antisymmetric part of η_{ijkl}, which is proportional to the quadrupole contribution $\chi^*/8$.

3.1. Random array

In this case, the matrix $\mathbf{R}^{(11)}$ has been evaluated by, among others, Childress [19], Howells [20], Hinch [8] and Kim and Russel [6] as:

$$R_{ij}^{(11)} = 6\pi\mu a\alpha_1\delta_{ij}, \tag{36}$$

where

$$\alpha_1 = 1 + \frac{3}{2^{1/2}}\phi^{1/2} + \frac{135}{64}\phi\log\phi + 16.456\phi + o(\phi). \tag{37}$$

In addition, the coupling term $\mathbf{R}^{(12)}$ is zero, because of symmetry. As for the viscosity term, $\mathbf{R}^{(22)}$ has been evaluated by Kim and Russel [6], finding:

$$R_{ijkl}^{(22)} = \tfrac{10}{3}\pi\mu a^3\beta_1(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}) + 2\pi\mu a^3\beta_2(\delta_{ik}\delta_{jl} - \delta_{il}\delta_{jk}), \tag{38}$$

where

$$\beta_1 = 1 + \tfrac{81}{80}\phi\log\phi + 7.86\phi + O(\phi^{3/2}\log\phi), \tag{39}$$

$$\beta_2 = 1 + \tfrac{27}{64}\phi\log\phi + O(\phi^{3/2}\log\phi). \tag{40}$$

In addition, applying Faxen's law and using Equation (36), we obtain:

$$R_{ijkl}^{(13)} = \pi\mu a^3\alpha_1\delta_{il}\delta_{jk}. \tag{41}$$

Finally, proceeding as in the last section, we find the momentum-conservation equation (31) and constitutive relations (32)–(34), with permeability, spin viscosity and stress viscosity given by:

$$\kappa^* = 2a^2/(9\alpha_1\phi), \qquad \chi^* = 6\phi\mu\alpha_2, \qquad \eta^* = \mu(1 + \alpha_3\phi), \tag{42}$$

where:

$$\alpha_2 = 2\beta_2 - \alpha_1 = 1 - \frac{3}{2^{1/2}}\phi^{1/2} - \frac{243}{256}\phi\log\phi - 16.541\phi + o(\phi), \tag{43}$$

$$\alpha_3 = \tfrac{1}{4}(10\beta_1 - 3\alpha_1) = \frac{7}{4} - \frac{9}{4.2^{1/2}}\phi^{1/2} - \frac{243}{256}\phi\log\phi + 7.24\phi + o(\phi). \tag{44}$$

The velocity and pressure field of a fluid flowing through an array of spheres randomly distributed can also be described by Brinkman's Equation (30), with permeability κ^* [*cf.* Equation (42)] and effective viscosity

$$\begin{aligned}\frac{\mu^*}{\mu} &= 1 + \tfrac{1}{2}\phi(5\beta_1 + 3\beta_2 - 3\alpha_1)\\ &= 1 + \frac{5}{2}\phi - \frac{9\cdot 2^{1/2}}{4}\phi^{3/2} - 1.72\phi^2 + O(\phi^{5/2}\log\phi).\end{aligned} \tag{45}$$

Table 1. Values of the constants $\tilde{a}, \tilde{b}$ and $\tilde{c}$ for different cubic lattices

Lattice	$\tilde{c}$	$\tilde{a}$	$\tilde{b}$
s.c.	1.7601	0.2857	0.04655
b.c.c.	1.791	0.0897	0.01432
f.c.c.	1.791	0.0685	0.01271

Note that the $\phi \log \phi$-term cancels out and does not appear in (45). This expression for the effective viscosity coincides up to $O(\phi^{3/2})$-terms with that of Freed and Muthukumar [18]. The $O(\phi^2)$-term is new.

3.2. CUBIC ARRAYS

Let us consider a packed bed composed of identical spheres in a periodic cubic array. In this case, the matrix $\mathbf{R}^{(11)}$ has been evaluated by, among others, Hasimoto [21], Zick and Homsy [22] and Sangani and Acrivos [23], and is given by Equation (36), with

$$\alpha_1 = 1 - \tilde{c}\phi^{1/3} + \phi - (\tfrac{1}{5} + 630\tilde{b}^2)\phi^2 - 300\tilde{a}\tilde{b}\phi^{8/3} + O(\phi^{10/3}), \tag{46}$$

where $\tilde{a}, \tilde{b}$ and $\tilde{c}$ are constants whose values for simple, body-centered and face-centered cubic arrays can be found in Refs. [21, 24] and are listed in Table 1. Note that the constant $\tilde{c}$ equals $a\phi^{-1/3}c$, where c is defined in Refs. [21, 23], while the constants $\tilde{a}$ and $\tilde{b}$ are the constants used in Ref. [24], and equal $a^5\phi^{-5/3}a_{20}$ and $a^3\phi^{-1}b_{20}$, respectively, where a_{20} and b_{20} are defined in Ref. [23].

The coupling term $\mathbf{R}^{(12)}$ is zero, out of symmetry, while the matrix $\mathbf{R}^{(22)}$ was determined by Zuzovski *et al.* [24], who found that it is given by Equation (38) with

$$\beta_1 = 1 + (1 + 40\tilde{b})\phi + 8\tilde{a}\phi^{5/3} + (1 + 40\tilde{b})^2\phi^2 + O(\phi^{7/3}), \tag{47}$$

$$\beta_2 = 1 + \phi + \phi^2 + O(\phi^{10/3}). \tag{48}$$

From these results we find again the momentum–conservation Equation (31) and constitutive relations (32)–(34), where permeability, spin viscosity and stress viscosity are given by (42) with

$$\alpha_2 = 1 + \tilde{c}\phi^{1/3} + \phi + (\tfrac{11}{5} + 630\tilde{b}^2)\phi^2 + O(\phi^{8/3}), \tag{49}$$

$$\begin{aligned}\alpha_3 = {} & \tfrac{7}{4} + \tfrac{3}{4}\tilde{c}\phi^{1/3} + (\tfrac{7}{4} + 100\tilde{b})\phi \\ & +20\,\tilde{a}\phi^{5/3} + (\tfrac{53}{20} + 200\tilde{b} + \tfrac{1745}{2}\tilde{b}^2)\phi^2 + O(\phi^{7/3}).\end{aligned} \tag{50}$$

Finally, Brinkman's equation (30) is obtained, with effective viscosity

$$\begin{aligned}\frac{\mu^*}{\mu} = {} & 1 + \tfrac{5}{2}\phi + \tfrac{3}{2}\tilde{c}\phi^{1/3} + (\tfrac{5}{2} + 100\tilde{b})\phi^2 + 20\tilde{a}\phi^{8/3} \\ & +(\tfrac{43}{10} + 200\tilde{b} + 4945\tilde{b}^2)\phi^3 + O(\phi^{10/3}).\end{aligned} \tag{51}$$

As we see, for cubic arrays we are able to determine the effective viscosity up to $O(\phi^3)$, while for random arrays we can only determine it up to $O(\phi^2)$. This is obviously due to the fact that the multipole strengths of spheres in a periodic array are known more accurately than for a random configuration.

4. Conclusions and discussion

In this article, creeping flow through porous media has been shown to be described macroscopically by Brinkman's equation. The effective transport coefficients, namely the permeability, coupling and viscosity tensors, are expressed in terms of six tensors of the single-particle grand resistance matrix, connecting the multipole strengths at the center of a particle with the velocity gradients of the unperturbed flow field. Only four of these six tensors are independent, as the remaining two can be determined by means of a new generalized version of the reciprocity relations. In this way, the Brinkman viscosity was determined up to $O(\phi^2)$- and $O(\phi^3)$-terms for random and cubic arrays of spherical particles, respectively.

The main contribution of this work is to show how to use the reciprocity relations (16)–(19) in conjunction with known results about the single-particle grand resistance matrix to obtain constitutive equations with a high degree of accuracy. This method can be applied to a variety of problems, such as heat- and mass-transport in porous media and suspension flow. As a further example of application, let us consider the motion of a dilute suspension of neutrally buoyant spheres of radius a. In this case, the governing equation (1) is supplemented by the constitutive relations (13)–(15) for the multipole strengths, with u_i replaced with $(u_i - U_i)$, where U_i denote the particle translational velocities. Now, considering that the suspended particles are force- and torque-free, applying Faxen's law and using the reciprocity relation (18), we obtain at leading order,

$$F_{ij}^{(2)} = \frac{10\pi}{3}\mu a^3(\nabla_i u_j + \nabla_j u_i); \qquad F_{ijk}^{(3)} = \frac{\pi}{6}\mu a^5 \delta_{ij}\nabla^2 u_k. \tag{52}$$

Finally, substituting these results, together with $\mathbf{F}^{(1)} = \mathbf{0}$ in (1), and averaging, using (5), we obtain

$$\nabla\langle p\rangle = \mu(1 + \tfrac{5}{2}\phi)\nabla^2\langle \mathbf{u}\rangle - \tfrac{1}{8}\mu a^2\phi\nabla^4\langle \mathbf{u}\rangle; \quad \nabla\cdot\langle \mathbf{u}\rangle = 0. \tag{53}$$

Now, it is clear that the biharmonic term in Equation (53) further stabilizes the diffusive part of the Stokes equation, and therefore plays a similar role as the viscous term of Brinkman's equation. As the biharmonic term is $O(a^2/l^2)$ times smaller than the viscous term, it can be neglected in the bulk, while it becomes important near the boundaries, perhaps providing some ground for the introduction of a slip boundary condition associated with the effective Stokes equation. The new fourth-order equation, however, requires an additional boundary condition for **u** which, at the moment, is not known. Therefore, in view of this difficulty, we prefer to consider Equation (53) as a conjecture and reserve a more detailed analysis of the biharmonic correction to the effective Stokes equation for future studies.

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