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Rigorous bounds on the fluid permeability: Effect of polydispersivity in grain size

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Rigorous bounds on the fluid permeability (or resistance) of porous media composed of spherical grains with a continuous size distribution are computed. For any finite degree of polydispersivity, scaling the resistance bound by the square of the specific surface (relative to the monodisperse case) yields effectively universal behavior at a fixed sphere volume fraction. A new proposition regarding an exact relationship between the permeability and another effective parameter, the trapping constant associated with diffusion-controlled reactions among traps, is employed to assess the accuracy of the rigorous bound.

I. INTRODUCTION

The flow of a fluid through a porous medium plays an important role in a variety of technological problems such as oil and gas recovery, hydrology, gel chromatography, filtration, and biological membranes, to mention but a few examples. A key macroscopic property of interest for describing slow viscous flow through porous media is the fluid permeability k defined through Darcy's law. The fluid permeability depends upon the microstructure of the medium through an infinite set of statistical correlation functions. Unfortunately, for random porous media, this set of functions is never completely known and hence an exact theoretical determination of k for general microstructures is out of the question.

Theoretical approaches to predicting k of random porous media fall primarily into one of two categories: determination of effective-medium approximations[1-4] or of rigorous bounds.[5-12] In this paper, we focus on the latter. Since the papers of Prager[5] and Doi,[6] which describe the derivation of variational upper bounds on k, a considerable amount of effort has been put forth to find improved upper bounds[7-9,11,12] and to derive lower bounds.[10,12] These bounds involve the microstructure of the medium via its first few statistical correlation functions. Theoretical calculations of these bounds have thus far been limited to equisized (monodispersed) distributions of spheres.[5,8-12] The evaluation of rigorous bounds on k for porous media composed of spherical grains with a polydispersivity in grain size has heretofore not been carried out.

One aim of this paper is to compute and study the so-called "two-point interfacial-surface" upper bound on k, obtained by Doi[6] and, more recently, by Rubinstein and Torquato,[12] for such a model. Interestingly, scaling the inverse permeability or resistance k^{-1} by the square of the specific surface (relative to a monodispersed system) gives, for the size distribution employed here, effectively universal behavior at a fixed volume fraction. We also employ a new proposition concerning the relationship between the trapping constant k_D associated with diffusion-controlled reactions among static traps and the fluid permeability k for the same microgeometry to assess the accuracy of the interfacial-surface bound.

II. INTERFACIAL-SURFACE UPPER BOUND AND MODEL-SYSTEM CORRELATION FUNCTIONS

A. Interfacial-surface upper bound

Doi,[6] and later Rubinstein and Torquato,[12] using a different variational approach, found that the fluid permeability k for statistically isotropic media of *general topology* with porosity ϕ_1 and specific surface s was bounded from above by

$$k^{(2)}=\frac{2}{3}\int_0^\infty r\left(F_{vv}(r)-\frac{2\phi_1}{s}F_{sv}(r)+\frac{\phi_1^2}{s^2}F_{ss}(r)\right)dr. \tag{1}$$

The functions $F_{vv}(r)$, $F_{sv}(r)$, and $F_{ss}(r)$ are the void–void, surface–void, and surface–surface two-point correlation functions, respectively. Following Rubinstein and Torquato,[12] who derived four different classes of bounds, we refer to (1) as a two-point "interfacial-surface" upper bound. This bound has been evaluated only for two models: monodispersed overlapping spheres[6] and monodispersed impenetrable spheres.[8]

B. Model system and correlation functions

We shall consider the evaluation of (1) for flow around a bed of overlapping spherical grains with a continuous distribution in radius R characterized by the probability density function $f(R)$. This is a good model of *consolidated* porous media such as a sandstone, which is characterized not only by an interconnected fluid phase but an interconnected solid phase.

Now, in order to compute (1) we need to know the one- and two-point correlation functions for our model.

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Chiew and Glandt[13] have obtained expressions for the porosity ϕ_1 and specific surface s (interface area per unit volume) for polydispersed overlapping spheres at total number density ρ:

$$\phi_1=\exp[-\rho(4\pi/3)\langle R^3\rangle], \quad (2)$$

$$s=\rho 4\pi\langle R^2\rangle\exp[-\rho(4\pi/3)\langle R^3\rangle], \quad (3)$$

where

$$\langle R^n\rangle=\int_0^\infty R^n f(R)dR \quad (4)$$

are the moments of the probability density $f(R)$.

Stell and Rikvold[14] obtained $F_{vv}(r)$ for this model as

$$F_{vv}(r)=\exp[-\rho\langle V_2(r;R)\rangle], \quad (5)$$

where

$$V_2(r;R)=\frac{4\pi}{3}R^3\left(1+\frac{3r}{R}-\frac{r^3}{16R^3}\right)H(2R-r) \quad (6)$$

and $H(x)$ is the Heaviside step function.

Expressions for the surface correlation functions F_{sv} and F_{ss} have not been obtained for this model, however. Miller and Torquato[15] have obtained these quantities for bidispersed overlapping spheres by extending the formalism of Torquato[8] to compute F_{sv} and F_{ss} (and their generalizations) for monodispersed spheres. Following this procedure, we find, for our model, that

$$F_{sv}(r)=4\pi\rho\langle R^2-(R^2/2-rR/4)H(2R-r)\rangle F_{vv}(r) \quad (7)$$

and

$$F_{ss}(r)=\left[16\pi^2\rho^2\left\langle R^2-\left(\frac{R^2}{2}-\frac{rR}{4}\right)H(2R-r)\right\rangle^2 + \frac{2\pi\rho\langle R^2H(2R-r)\rangle}{r}\right]F_{vv}(r). \quad (8)$$

Note that one can obtain corresponding results for overlapping spheres with p different sizes from the results given above by letting

$$f(R)=\sum_{i=1}^{p}\frac{\rho_i}{\rho}\delta(R-R_i), \quad (9)$$

where ρ_i and R_i are the number density and radius of type-i particles, respectively, and $\delta(R)$ is the delta function. For example, the combination of (9), with $p=2$ and the surface-correlation-function relations (7) and (8) yields the bidispersed results of Miller and Torquato.[15]

For polydispersed beds of spherical grains, there are a variety of choices available to scale k by so as to render it dimensionless. One natural scaling factor is the appropriately generalized Stokes dilute-limit permeability:

$$k_s=2\langle R^3\rangle/(9\langle R\rangle\phi_2), \quad (10)$$

where $\phi_2=1-\phi_1$ is just the sphere volume fraction. However, in the dilute limit, the bound (1) gives

$$k_0=2\langle R^3\rangle^2/(9\langle R^2\rangle^2\phi_2), \quad (11)$$

which implies that the bound is exact for dilute concentrations of spheres (be they overlapping or nonoverlapping) in the monodisperse limit. For spheres with any polydispersivity in size, on the other hand, relation (11) implies that the bound must always be greater than k_s in the dilute limit. Miller and Torquato[15] found a similar discrepancy between the interfacial-surface *lower* bound on the *trapping rate* k_D (associated with diffusion-controlled reactions among static traps) and the Smoluchowski dilute-limit result for k_D.

What is the physical significance of k_0? For fixed ϕ_2, k_0 is inversely proportional to S^2, where S is the ratio of the specific surface of a polydispersed system of overlapping spheres to that of a monodispersed system with radius $\langle R\rangle$. From (2), (3), and (11), it is easily seen that

$$k_0=2\langle R\rangle^2/(9\phi_2 S^2), \quad (12)$$

where

$$S=(\langle R^2\rangle/\langle R^3\rangle)\langle R\rangle. \quad (13)$$

In order to compute (1), one must choose a probability density function $f(R)$. The one we employ in this study is the Schulz distribution,[16]

$$f(r)=[1/(m-1)!](m/\langle R\rangle)^m R^{m-1} \times\exp(-mR/\langle R\rangle), \quad m\geqslant 1, \quad (14)$$

which normalizes to unity. The moments of the Schulz distribution are

$$\langle R^n\rangle=[(n+m-1)!/(m-1)!\,m^n]\langle R\rangle^n. \quad (15)$$

Therefore, by increasing m, the variance decreases, i.e., the distribution becomes sharper. In the monodisperse limit, $m\to\infty$, $f(R)=\delta(R-\langle R\rangle)$. From (13) and (15), one finds that the specific surface ratio S for the Schulz distribution is given by

$$S=m/(m+2). \quad (16)$$

Equation (16) leads to the interesting conclusion that a polydispersed system with finite m has a smaller specific surface than a monodispersed one at fixed ϕ_2.

III. CALCULATION OF THE INTERFACIAL-SURFACE BOUND

Here we shall compute the bound (1) for overlapping spheres with the radii R distributed according to the continuous Schulz distribution (14). Such calculations require the use of the correlation functions (2), (3), (5), (7), and (8) for this model.

In Fig. 1, we plot the scaled inverse permeability (or scaled fluid resistance) k_s/k for several values of the parameter m as a function of the particle volume fraction as obtained from (1). Thus the curves presented represent rigorous *lower* bounds on k_s/k. Recall that k_s is the exact dilute-limit permeability as given by (10). Note that as the degree of polydispersivity increases (i.e., as m decreases) for fixed ϕ_2, the scaled resistance decreases. This is expected behavior since, as observed earlier, the specific surface s decreases as m decreases for fixed ϕ_2. Again, for

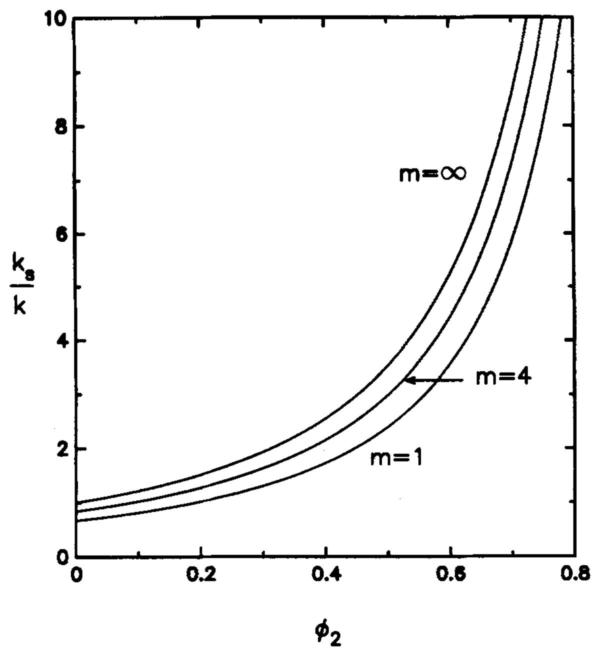

FIG. 1. Two-point interfacial-surface lower bound on the scaled fluid resistance k_s/k for polydispersed, overlapping spherical grains versus the particle volume fraction ϕ_2 as computed from Eq. (1). The radii are distributed according to the Schulz distribution (14). The cases $m = 1$, 4, and ∞ are shown.

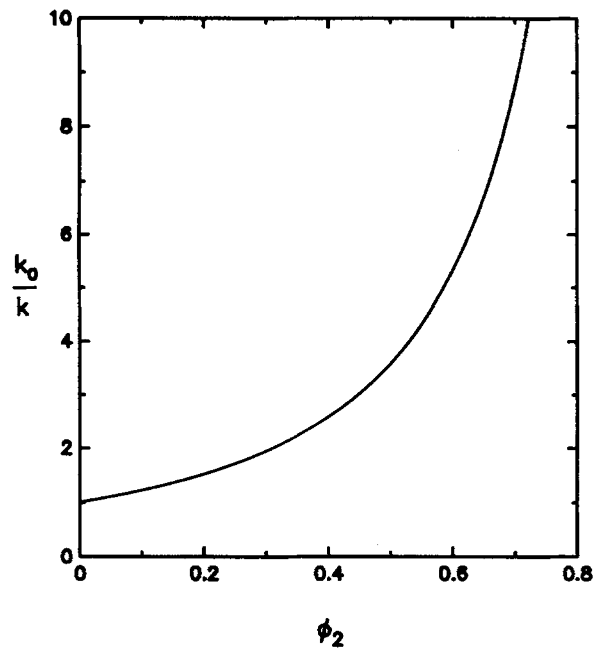

FIG. 2. Two-point interfacial-surface lower bound on the scaled fluid resistance k_0/k [where k_0 is given by Eq. (12)] for polydispersed, overlapping spherical grains versus the particle volume fraction ϕ_2 as computed from Eq. (1). The radii are distributed according to the Schulz distribution (14). The cases $m = 1$, 4, and ∞ are shown. All curves, to an excellent approximation, collapse onto the monodisperse curve ($m = \infty$).

reasons mentioned above, only the monodisperse limit ($m=\infty$) of the bound gives the exact result as $\phi_2 \to 0$.

Interestingly, if one multiplies $1/k$ by k_0, the exact dilute-limit behavior of the upper bound on the permeability given by (11) or (12), all the curves, to an excellent approximation, collapse onto the monodispersed curve, regardless of the degree of polydispersivity. This is shown in Fig. 2 for $m = 1$, 4, and ∞, where it is seen that, on the scale of the figure, the results for any value of m are virtually indistinguishable from one another. Thus, given the monodisperse bound on the scaled resistance for overlapping spheres, one can obtain any corresponding polydisperse bound on the inverse permeability, at the same value of ϕ_2, by dividing the former bound by the simple expression for k_0, Eq. (12). Recall that k_0 is inversely proportional to the square of the relative surface area S. There is no reason to believe that this simple scaling will apply to bounds for other model microstructures or to the exact expression for the inverse permeability.

IV. RELATION BETWEEN PERMEABILITY AND TRAPPING CONSTANT

The problems of diffusion-controlled reactions among perfectly absorbing traps of slow viscous flow in porous media share a common feature: screening effects, at small solid volume fractions, lead to expansions for the steady-state trapping constant k_D and fluid permeability k, which are nonanalytic in ϕ_2.[1,17] (In the trapping problem, reactant is being produced at a constant rate and diffuses in the trap-free region but is instantly absorbed in contact with any trap. The steady-state trapping constant k_D is proportional to the ratio of the rate of production to the mean concentration field.[6,18,19]) No one, however, has ever considered investigating the possibility of a deeper relationship between these two different physical parameters.

Here we use a new proposition regarding the relationship between the fluid resistance k^{-1} and trapping constant k_D for an isotropic porous medium of general topology having a fluid region of porosity ϕ_1, namely,[20]

$$k^{-1} \geqslant k_D. \qquad (17)$$

That is, the fluid resistance bounds the trapping constant from above for the same microgeometry. This should prove to be a useful relationship, since in some cases one property may be easier to measure or predict than the other. The proposition, which in its general form for *anisotropic* media is a tensor relation,[20] was motivated by several observations. First, rigorous two-point interfacial-surface bounds for k^{-1} and k_D, which are valid for arbitrary topology,[12,18] satisfy relation (17). Second, for a disconnected or nonpercolating fluid phase of general microgeometry, it is known that k_D is finite[21] while k^{-1} must be infinite, thus expression (17) is obeyed in such instances. Third, inequality (17) is satisfied for random arrays of spheres at low but nondilute concentrations[1,17] and for periodic arrays of spheres for arbitrary densities.[22,23] Fourth, two-point bounds for k^{-1} and k_D for irregularly shaped particles with cusps satisfy the inequality (17) (see Ref. 22). A rigorous proof of the general form of this proposition for *anisotropic* porous media will be forthcoming.[24]

TABLE I. Effective parameters for transport around overlapping spheres with radii distributed according to the Schulz distribution (14); $m = \infty$ (monodisperse), $m = 4$, and $m = 1$. Tabulated for several values of the sphere volume fraction ϕ_2 is the two-point interfacial-surface lower bound on the scaled fluid resistance k_s/k as computed from relation (1) and the scaled trapping constant $k_D/k_{D,s}$ as computed from (19). The results for $k_D/k_{D,s}$ obtained from Ref. 15, combined with inequality (18), give the best estimate of the scaled fluid resistance to date for this geometry.[26]

	$m=\infty$		$m=4$		$m=1$	
ϕ_2	k_s/k	$k_D/k_{D,s}$	k_s/k	$k_D/k_{D,s}$	k_s/k	$k_D/k_{D,s}$
0.1	1.22	1.97	1.02	1.89	0.82	1.80
0.2	1.51	2.91	1.27	2.74	1.20	2.57
0.3	1.93	4.19	1.63	3.91	1.30	3.62
0.4	2.55	6.11	2.15	5.65	1.73	5.16
0.5	3.53	9.20	2.99	8.42	2.40	7.62
0.6	5.22	14.64	4.42	13.30	3.56	11.91
0.7	8.57	25.66	7.26	23.12	5.85	20.49
0.8	16.86	53.78	14.33	48.02	11.57	42.12
0.9	51.54	173.63	43.61	153.39	35.33	132.76

A practically important question is how sharp is inequality (17)? This is a difficult question to answer at this point since there are very few geometries for which we have exact results. For periodic arrays and random arrays at low but nondilute concentrations, (17) is relatively sharp. For such dispersions, however, the more restrictive inequality

$$k^{-1}k_s \geqslant k_D k_{D,s}^{-1} \tag{18}$$

holds, where k_s and $k_{D,s}$ are dilute-limit results for k and k_D, respectively. For example, for simple cubic lattices,[22,23] $k_s/k = 1.212$ and $k_D/k_{D,s} = 1.211$ at $\phi_2 = 0.001$; $k_s/k = 2.810$ and $k_D/k_{D,s} = 2.606$ at $\phi_0 = 0.064$; $k_s/k = 4.29$ and $k_D/k_{D,s} = 3.62$ at $\phi_2 = 0.125$; and $k_s/k = 15.4$ and $k_D/k_{D,s} = 7.82$ at $\phi_2 = 0.343$. It is important to emphasize that such bounds are considerably sharper than any "direct" variational bounds on k^{-1} or k_D that have been evaluated thus far (see the bounds of Refs. 12 and 23).

Miller and Torquato[15] have recently obtained a highly accurate analytical expression for the trapping constant of overlapping spherical traps with a continuous size distribution (the same model considered in the previous sections) by extending the results of Richards.[25] For such a microgeometry, they found that[26]

$$\frac{k_D}{k_{D,s}} = \frac{\eta}{\phi_1\phi_2}\frac{1}{1-\sqrt{\pi}ye^{y^2}\,\mathrm{erfc}(y)}, \tag{19}$$

where

$$k_{D,s} = 3\phi_2\langle R\rangle/\langle R^3\rangle, \tag{20}$$

$$y = 2\rho^{1/2}\langle R^2\rangle/\langle R\rangle^{1/2}, \tag{21}$$

$$\eta = \rho(4\pi/3)\langle R^3\rangle. \tag{22}$$

Equation (19) was found to be in good agreement with "exact" computer-simulation data and hence, to a good approximation, may be regarded as exact.

In Table I, we compare our calculations of the two-point interfacial-surface *lower* bound on the scaled resistance k_s/k and the scaled trapping constant $k_D/k_{D,s}$ as obtained from (19). If we invoke inequality (18), then an obvious conclusion is that the two-point bound on the scaled resistance is not very sharp. Thus two-point bounds are insufficient to yield accurate estimates of the fluid permeability or resistance, i.e., one must rely upon higher-order variational bounds to give good estimates of k. Moreover, result (19), in light of the inequality (18), must be regarded as the best estimate of the scaled fluid resistance k_s/k for this polydispersed geometry.

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