Hydrodynamics of Particulate Motion in Porous Media

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We develop a general model for flow of suspended particles in a porous medium. The model can take into account the effects of the morphology of the porous medium (including pore surface roughness and fractality), the chemistry of the flowing fluid, and all important forces between the particles and the pore surface. If surface deposition and pore plugging by size exclusion take place, the process is a percolation phenomenon which may belong to a different universality class than that of random percolation. The model is applied to two classes of problems, flow of solid, electrically charged colloidal particles, and that of stable emulsion droplets in a porous medium.

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Flow of suspended particles in porous media is relevant to a wide variety of natural and industrial processes and has been studied for a long time.[1,2] Examples include hindered diffusion in membranes,[3] hydrodynamic chromatography,[4] deep bed filtration,[5] ground water contamination,[6] flow of dilute stable emulsions,[7,8] polymer flooding,[9] migration of fines[10] (electrically charged colloidal particles), and gel permeation. The efficiency of such processes depends crucially on the availability of open pore space which provides transport paths for the flowing particles, the interaction of the particles with the surface of the pores, and the chemistry of the flowing fluid (i.e., its ionic strength and pH).

Classical approaches to modeling of such phenomena[1] have been based on the macroscopic continuum conservation equations. These purely phenomenological models generally ignore the details of the morphology of the pore space, i.e., its pore size distribution, pore surface structure (e.g., its roughness), and pore connectivity, and also provide no insight on how the physicochemical forces and the chemistry of the flowing fluid affect the transport of the suspended particles. One usually has to introduce several adjustable parameters into the models, and also needs reasonable estimates of such parameters whose experimental or theoretical determination is not always possible. In this Letter we propose a new approach to the problem of flow of suspended particles in porous media. We combine a network model of porous media with molecular-dynamics-like simulations to determine the exact paths of the particles in the pore space from which all the important properties can be calculated.

We represent the porous medium by a simple-cubic network in which each bond, assumed to be a cylindrical tube of constant length, represents a pore of the medium. The effective radius R of each pore is distributed according to a pore size distribution $f(R)$. However, unlike all previous simulations of flow in random networks, we assume that the surface of the pores is *not* smooth. It is now well established that[11] the surface of many porous media, e.g., sandstone, is rough and, at least over certain length scales, can be characterized by a fractal dimension $D_s > 2$. We show below that, for a realistic simulation of flow of suspended particles in porous media, one has to incorporate some measure of surface roughness and fractality into the model. To do so, we assume that the surface of each pore is covered by overhangs of various heights h where typically[11-13] $h/R \leq 0.05$. However, the overhangs provide large surface area for the attraction, adsorption, and retention of the suspended particles as shown below.

We assume that there is Poiseuille flow in each pore, so that the fluid-flow problem is isomorphic to current flow in a random-resistor network and, thus, for a given set of boundary conditions, the nodal pressures can be calculated. The suspended particles are assumed to be hard spheres whose effective radii are selected from a distribution $g(r_p)$. Particle concentration in the flowing fluid is assumed to be low enough for the suspension to be dilute. For nondilute cases, the particle-particle interactions may be important, and although our model would still be applicable, the computations would be more intensive. If the particles and/or the surface of the pores are electrically charged, their electrical charges Q_p and Q_s are also specified.

The particles are injected into the network at randomly selected points on the upstream plane of the network. For each pore of radius R, we select a random number δ, uniformly distributed in (r_p,R), and place the center of the particle at that point. We inject *simultaneously* M particles at time $t=0$, another M particles at time Δt, and so on (in our simulations reported below, $M \sim 150$). For each particle within a pore we write $\sum_i F_i = 0$ and $\sum_i I_i = 0$, where F_i and I_i denote, respectively, the various forces and their corresponding torques acting on the particles. Some of the forces that are included are gravitational, inertial, molecular dispersion, double-layer interaction, drag, and Brownian forces. Because the flow of fluid within the pore is slow enough and the particles are not usually too large, the flow field near the particles can be represented by a linear shear flow for which exact expressions for drag force F_D and torque were derived by

Goldman, Cox, and Brenner,[14] and were employed in the simulations. Combining the force and torque balances yields a differential equation of the form $H(y, dy/dz, d^2y/dz^2)=0$, with z being the axial position of the center of the particle and $y=R-r$, where r is the radial position of the particle's center. For every particle within every pore we solve this equation numerically to determine its exact path. If the path of a particle does not intersect the surface of the pore, the particle will pass through the pore and arrive at the next node. From particles' paths, we calculate the time for passing through the pore and, hence, various properties of the suspended particles (e.g., their effective diffusivity). Each time a particle enters a new pore, its new path is calculated.

If the path does take a particle to the surface of a pore, then depending on the signs of Q_s and Q_p one of the two events may take place. Suppose that Q_s and Q_p are of opposite sign, and consider a particle on the rough surface of the pore in the vicinity of an overhang (Fig. 1). It can be shown that[13] if the inequality

$$F_S(2r_ph-h^2)^{1/2} > 10.205\pi r_p^2\tau_w(r_p-h)+3.776\pi r_p^3\tau_w \quad (1)$$

is obeyed, then the particle will attach itself to the surface. Here F_S is the attractive surface interaction force which, for a particle sufficiently close to the surface, is dominated by the London force F_L, and τ_w the local shear stress at the surface. The first term on the right-hand side of (1) is the contribution of net drag (hydrodynamic) force acting through the center of the particle, whereas the second term is the contribution of the moment M_D around the center of the particle. Without any surface roughness and overhangs, the inequality (1) is always violated, i.e., no *permanent* attachment or adsorption takes place, and an attached particle will simply roll on the surface and leave the pore. Thus, for every particle that has arrived on the surface of a pore, inequality (1) is checked. If it is obeyed, then we attach the particle to the surface and record its position. Because of the attachment, the flowing fluid exerts a drag force on the deposited particle which increases the resistance to flow in that pore, which is equivalent to a reduction in the effective radius of the pore. We estimate the new effective radius of the pore from the change in the pressure drop along the pore, which has already been calculated rigorously by Goldman, Cox, and Brenner,[14]

$$\pi R_a^2\Delta P_a=\pi R_b^2\Delta P_b+10.205r_p^2\tau_w, \quad (2)$$

where ΔP_a (R_a) is the pressure drop (pore radius) *after* deposition of the particle, and ΔP_b (R_b) its corresponding value *before* deposition. From Eq. (2) we can calculate the new effective radius of the pore.

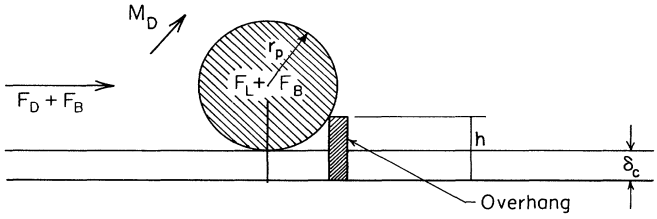

FIG. 1. Forces and torques acting on a particle close to the pore surface in the presence of an overhang. F_B and F_L are the body and London forces, respectively.

However, if Q_s and Q_p are of the same sign, then there are repulsive forces between the particles and the surface at a certain separation distance δ_c between the pore surface and that of the particle. To calculate δ_c, we find the distance δ_c from the pore surface at which the net force acting on the particle changes sign (becomes repulsive). If $\delta_c > h$, where h is the height of the overhang in the vicinity of the particle, the particle can have no contact with the pore surface (overhang) and will pass through the pore. If $\delta_c < h$, the inequality (1) (in which h is replaced by $h-\delta_c$) is checked to see whether the conditions for particle deposition are favorable.

If a particle of radius r_p passes through a pore and arrives at a node, it selects a new pore of radius R with a probability proportional to the fluid flux in that pore.[15] If $r_p \geq R$, the pore entrance is blocked by the particle and the pore can no longer contribute to the flow field and the permeability of the medium. The radius of such a pore is set to be zero for the rest of the simulations. This is the so-called size-exclusion (SE) phenomenon which is routinely observed in many processes involving flow of suspended particles in porous media.[1-10] The SE phenomenon and particle adsorption on the pore surface are also responsible for the decrease in the permeability of the medium and the subsequent reduction in the efficiency of such processes. This permeability reduction is a percolation process.[5] Although in the beginning of the process most particles are normally too small to cause any pore blocking by SE, the gradual deposition of the particles and the resulting reduction in the effective radii of the pores increase the likelihood of pore blocking by SE at later stages of the process. Both particle deposition and pore blocking by SE also change the entire flow field in the network. Hence, after each pore blocking by SE, or after particle deposition (typically 3–5), we recalculate the entire flow field in the network. Thus, the entire simulation for each realization involves several thousands of pressure-field calculations (typically about 6000). A change in the morphology of the network by particle deposition and pore blocking also results in a change in the hydrodynamic forces acting on the deposited particles. Therefore, at every stage of the simulation, the inequality (1) is also checked for the *deposited* particles. The violation of (1) for such particles means that the hydrodynamic forces are strong enough to resuspend the particles in the flowing field, in which case the parti-

cles are released into the flow and their new paths are calculated. These are consistent with experimental observations.

Our model is quite general and applicable to a wide class of problems that involve flow of suspended particles through a porous medium. Not only the effect of pore-space morphology is taken into account, the chemistry of the flowing fluid and all important forces between the particles and the surface are also accounted for. For example, we can account for the pH and ionic strength of the solution since the double-layer interaction force F_{DL} is given by

$$F_{DL}=\gamma r_p \kappa (Q_s^2+Q_p^2)\left(\frac{2Q_sQ_p}{Q_s^2+Q_p^2}-e^{-\kappa\delta}\right)\frac{e^{-\kappa\delta}}{2(1-e^{-2\kappa\delta})}, \quad (3)$$

where γ is the dielectric constant of the fluid and κ the Debye-Huckel reciprocal length, which is given by $\kappa=(4\pi e^2\sum_i m_i z_i/\epsilon kT)^{1/2}$. Here k is Boltzman's constant, T the absolute temperature, z_i the valency of the ith ionic species and m_i its concentration, ϵ the porosity, and e the electron's charge. Thus, one can adjust both the ionic strength and the pH of the solution. To the best of our knowledge, this is the first comprehensive model of flow of suspended particles through a porous medium that can take into account the effects of all such factors.

We now briefly describe the application of our model to two important problems. The complete details of these and other applications can be found elsewhere.[16] The first problem is the flow of fines or colloidal particles through a porous medium. Fines migration is usually caused by the contact of the solid matrix of the porous medium with an incompatible brine solution. This releases fines into the flowing fluid which can plug pores by SE and surface deposition and severely reduce the permeability of the medium and the efficiency of oil recovery operations. Baghdikian, Sharma, and Handy[12] carefully studied the problem and reported the permeability reduction of an Ottawa sandpack as a result of injecting a suspension of kaolinite fines into the medium. Their reported pore size distribution is approximated very accurately by a log-normal distribution with an average of 18 μm and standard deviation of 12 μm, while their particle size distribution can be represented by a normal distribution with a mean of 1.84 μm and a standard deviation of 0.5 μm. The average height of an overhang in such a system is about[11,13] 60 Å (this gives rise to a fractal dimension $D_s\simeq 2.5$), the pH of the solution was about 10, the ionic strength about 0.1 mole, and the flow rate of the injected suspension about 8.6 cm^3/min. These were used in the simulations. We used a $15\times15\times15$ cubic network and averaged the results over twenty different realizations. In Fig. 2 we compare the predicted permeability with the data of Baghdikian, Sharma, and Handy,[12] and the agreement is excellent. In this figure q is defined as $q=Qt/V$, where Q is the total volumetric flow rate in the network at time t and V is the total pore volume at $t=0$. Also shown is the permeability of a system in which the surfaces of the pores have no overhangs and are smooth. Even if some particles can be attached to the surface, hydrodynamic forces acting on the particles make them roll on the surface and leave the pores. Thus, no significant permeability reduction can take place, except possibly by a few large particles that can plug pores by SE. This clearly demonstrates the significant role of a rough surface in such processes.

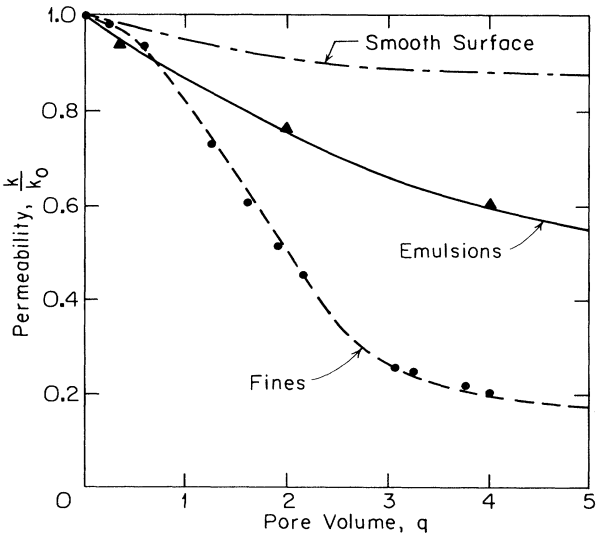

FIG. 2. Comparison of predicted normalized permeability with the experimental data (circles and triangles). Long-dashed–short-dashed curve is for fines migration with smooth pore surface.

To compare these results with the random percolation model, we plot in Fig. 3 the results in terms of the fraction p of unplugged pores, the standard practice in percolation.[17] Here a plugged pore is either one which is blocked by SE or one whose hydraulic conductivity has reduced to a very small value because of surface deposition. In contrast with random percolation, the permeability curve is convex. To check the scaling behavior of k near the percolation threshold p_c, we fitted the results with $k/k_0\sim(p-p_c)^\mu$ to estimate the critical exponent μ. In contrast with random percolation,[17] we *cannot* use finite-size scaling and estimate μ by doing simulations at p_c, since the permeability reduction in this problem is *process dependent*. We found $\mu\simeq 1$, which should be compared with $\mu\simeq 2$ for random percolation.[17] A mean-field-like approximation also predicts that[16] $\mu=\frac{1}{2}$, whereas the same approximation predicts that $\mu=1$ for random percolation. Thus, not only does our simulation identify this problem as a percolation process, it also in-

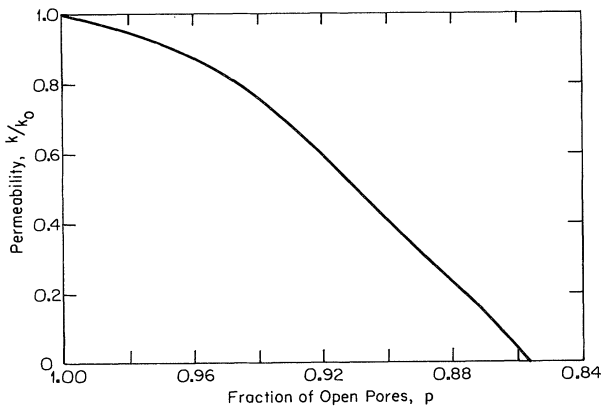

FIG. 3. Variations of permeability (during fines migration) with the fraction of unplugged pores.

dicates that it may belong to a different universality class than the random percolation.

The second application is to the problem of flow of stable emulsion droplets in porous media. The displacement of oil by another miscible fluid is often unstable and inefficient if the viscosity of the displacing fluid is less than that of oil. To decrease the viscosity contrast and increase the efficiency of the process, emulsion droplets either are injected into the displacing fluid or are generated *in situ* in the porous medium. However, while the viscosity contrast decreases, emulsion droplets can deposit on the surface of the pores and decrease k. Soo and Radke[8] provide precise data on permeability reduction during this process. Their pore size distribution was reported to be log-normal with an average of 15.3 μm and standard deviation of about 1 μm, while their emulsion size distribution was reported to be log-normal with an average of 3 μm and standard deviation of 0.23 μm. These were employed in the simulations. Soo and Radke[8] also demonstrated experimentally that emulsion droplets, unlike solid particles, do *not* deposit on top of one another, but deposit as a monolayer. Thus, the main difference between this simulation and fines migration is that only the formation of monolayers is allowed in this case. The results are also shown in Fig. 2 and the agreement is very good. In general, the permeability reduction in this problem is not as severe as in fines migration, because only monolayers of deposited droplets are formed.

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