Surface permeability of particulate porous media.

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The dispersion process in particulate porous media at low saturation levels takes place over the surface elements of constituent particles and, as we have found previously by comparison with experiments, can be accurately described by super-fast non-linear diffusion partial differential equations. To enhance the predictive power of the mathematical model in practical applications, one requires the knowledge of the effective surface permeability of the particle-in-contact ensemble, which can be directly related with the macroscopic permeability of the particulate media. We have shown previously that permeability of a single particulate element can be accurately determined through the solution of the Laplace-Beltrami Dirichlet boundary-value problem. Here, we demonstrate how that methodology can be applied to study permeability of a randomly packed ensemble of interconnected particles. Using surface finite element techniques we examine numerical solutions to the Laplace-Beltrami problem set in the multiply-connected domains of interconnected particles. We are able to directly estimate tortuosity effects of the surface flows in the particle ensemble setting.

I. INTRODUCTION

Liquid transport in particulate porous media, such as sand, is customarily classified into fully saturated, funicular and pendular regimes of spreading[1–4]. The first two regimes of the liquid dispersion occur at relatively high saturation levels $s > s_c$ 10%, where saturation s is defined as the ratio of the liquid volume V_L to the volume of available voids V_E in a sample volume element V, $s = \frac{V_L}{V_E}$. At high saturation levels, above the critical value s_c, liquid transport takes place in the pore space either fully or partially filled by the liquid.

Our prime concern here is the special case of liquid dispersion at low saturation levels. As the saturation level drops below the critical value, s s_c, that is to the value relevant to the pendular regime of spreading, the liquid volumes in the porous matrix become isolated[3–4]. As a result, at low saturation levels, the liquid is only contained in the pendular rings formed at the locations of the particle contacts and on the particle rough surfaces, and the liquid transport can only occur over the matrix surface elements, as is illustrated in Fig. 1.

Our main concern here is the wetting cycle, when the liquid spreads over a dry porous matrix or over a matrix with a very low background saturation level up to s_r 2%. These conditions are similar to those in the case studied previously experimentally and theoretically in[6]. The main driving force of the dispersion process, as is often the case during the wetting cycle, is capillary pressure developed at the moving front in the process of wetting of dry porous matrix, while the liquid bridges play a role of variable liquid reservoirs of uniform surface curvature.

The analysis of this regime of wetting, which is crucial for studies of biological processes and spreading of non-volatile liquids in arid natural environments and industrial installations, has shown that the liquid dispersion has many distinctive features and can be accurately described by the so-called superfast non-linear diffusion equation[5,6].

Theoretically, the superfast non-linear diffusion equation belongs to a special class of mathematical models. Unlike in the standard porous medium equation[7], in this special case, the non-linear coefficient of diffusion $D(s)$

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many particles of the porous medium, the diffusion process in the slow creeping flow conditions can be described by the following non-linear diffusion equation

$$\frac{\partial s}{\partial t} = \nabla \{ D(s) \nabla s \}; \quad t > 0; \qquad (1)$$

where

$$D(s) = \frac{D_0(s)}{(s \quad s_0)^{3/2}}; \quad s > s_0;$$

for $D_0 > 0$.

The details of derivation of (1) can be found in[5,6], here we note that, the resultant governing non-linear equation (1) directly follows from the conservation of mass principle

$$\frac{\partial (s)}{\partial t} + \nabla \quad Q = 0; \qquad (2)$$

where is porosity defined as $= \frac{V_E}{V}$, which is further assumed to be constant, and Q is the macroscopic flux density. The macroscopic flux density Q is defined in such a way that the total flux through the surface of a macroscopic sample volume element is given by the surface integral $\int Q \quad n\, dS$, where n is the normal vector to the surface of the sample volume element.

To obtain (1) from (2), one needs to apply the capillary pressure-saturation relationship[5,6,8] dictated by the liquid bridges behaviour

$$p = \quad p_0 \frac{A_c}{(s \quad s_0)^{1/2}} \qquad (3)$$

and the local Darcy's law[9,10] describing the surface flow in the rough layer of the particle elements

$$\frac{\ _m}{\ } \nabla u = q: \qquad (4)$$

Here, $A_c = \sqrt{\frac{3}{4}\frac{1}{\ }\frac{N_c}{\ }}$, N_c is the coordination number, that is the average number of bridges per a particle, $p_0 = \frac{2}{R}\cos \ _c$, is the coefficient of the surface tension of the liquid, $_c$ is the contact angle made by the free surface of the liquid bridge with the rough solid surface of the constituent particles, R is an average radius of the porous medium particles, q and u are the averaged local flux density and pressure in the rough surface layer, is liquid viscosity and k_m is the local coefficient of permeability of the rough surface, which is proportional to the average amplitude of the surface roughness $_R$, that is the width of the surface layer conducting the liquid flux

$$k_m \propto {}^2_R: \qquad (5)$$

One needs to emphasise here that two levels of averaging are involved in obtaining the final governing equation (1). While equations (1), (2) and (3) are 'truly' macroscopic, that is obtained by averaging using a volume element V containing many grain particles, equation (4) is only an average over some rough area of a single particle containing many surface irregularities, so that quantities q and u are also only local averages over that sample surface area.

Therefore, to transit from (4) to the macroscopic description, the spatial averaging theorem formulated in[11] should be applied. That is, using intrinsic liquid averaging $\langle ::: \rangle^l = V_l^{\ 1}\int_{V_l} d^3x$, where V_l is liquid volume within the sample volume V, one has $\langle u \rangle^l = p$ and $\langle q \rangle^l \frac{S_e}{S} = Q$. Here, S is the surface area of the sample volume V with the effective area of entrances and exits S_e. Note, the ratio S_e/S is not just a geometric property, but also takes into account the connectivity of the porous elements. For example, the effective area of entrances and exits S_e is on the surface of the spherical particle, and particles of

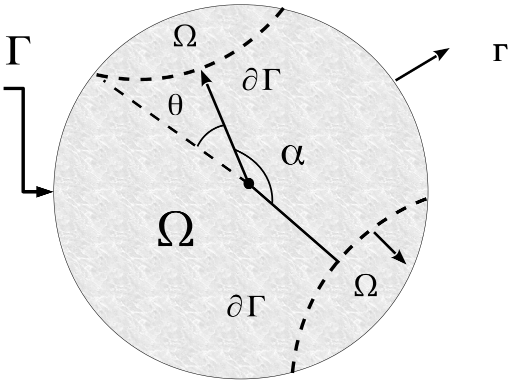

FIG. 2. Illustration of the flow and solution domains on the surface Γ of a spherical particle, and their geometric arrangements. In the picture, Ω_0 is the domain of the surface flow and the surface area covered by the liquid bridges corresponds to the domains Ω_1 and Ω_2.

are smooth[19–21], which, if it is found, allows to calculate the total flux through the particle element

$$Q_T = \sigma_R \frac{\kappa_m}{\mu} \oint_{\partial\Omega_1} \frac{\partial u}{\partial n_s}\, dl = -\sigma_R \frac{\kappa_m}{\mu} \oint_{\partial\Omega_2} \frac{\partial u}{\partial n_s}\, dl; \qquad (10)$$

where n_s is the normal vector to the domain boundaries $\partial\Omega_{1;2}$ on the surface, σ_R is the average amplitude of the surface roughness, that is the width of the surface layer conducting the liquid flux and the line integral is taken along a closed curve in Ω_0, for example the boundary $\partial\Omega_1$.

If the total flux Q_T is determined, one can define the global permeability coefficient of a single particle K_1. This can be done, if we assume that the particle has a characteristic size D and so that it can be enclosed in a volume element $V = D^3$ with the characteristic side surface area D^2. Then, the effective flux density Q can be represented in terms of K_1 (and the total flux Q_T)

$$Q = \frac{Q_T}{D^2} = -\frac{K_1}{\mu} \frac{U_2 - U_1}{D}; \qquad (11)$$

if the flow is driven by the constant pressure difference $U_2 - U_1$ applied to the sides of the volume element.

B. Surface permeability of a sphere in the case of azimuthally symmetric domain boundaries

Consider now a spherical particle in an azimuthally symmetric case, when the domain boundaries $\partial\Omega_1$ and $\partial\Omega_2$ are oriented at the reflex angle $\alpha = \pi$ and have a circular shape. We use a spherical coordinate system with its origin at the particle centre and the polar angle θ counted from the axis of symmetry passing through the centre of the circular contour $\partial\Omega_1$. In this case, the Dirichlet boundary value problem (8)-(9) admits an analytical solution, so that particle permeability can be determined explicitly. Indeed, problem (8)-(9), if we assume that the liquid pressure distribution u is a function of θ only and independent of the azimuthal angle, is equivalent to

$$\frac{1}{\sin\theta} \frac{\partial}{\partial\theta} \left(\sin\theta \frac{\partial u}{\partial\theta} \right) = 0; \qquad \theta_0 < \theta < \pi - \theta_1; \qquad (12)$$

with the boundary conditions

$$u|_{\theta=\theta_0} = U_1; \quad u|_{\theta=\pi-\theta_1} = U_2. \qquad (13)$$

The analytic solution to problem (12)-(13) after applying the boundary conditions can be represented in the following form

$$u = \sigma_0 (U_2 - U_1) \ln \left(\frac{\sin\theta}{\sin\theta_0} \frac{1+\cos\theta_0}{1+\cos\theta} \right) + U_1; \qquad (14)$$

where

$$\sigma_0 = \frac{1}{\ln \left\{ \frac{\sin\theta_1}{\sin\theta_0} \frac{1+\cos\theta_0}{1-\cos\theta_1} \right\}}.$$

One can now calculate the total flux and the permeability, using its definition (11),

$$Q_T = -\frac{K_1}{\mu} D (U_2 - U_1) = -2\pi \sin\theta_0\, \sigma_R \frac{k_m}{\mu} \left. \frac{\partial u}{\partial\theta} \right|_{\theta=\theta_0}$$

$$= -(U_2 - U_1)\, 2\pi \sigma_R \sigma_0 \frac{k_m}{\mu}. \qquad (15)$$

So that, taking $D = 2R$,

$$K_1 = \pi \sigma_0 \frac{\sigma_R}{R} k_m. \qquad (16)$$

the main diffusion equation? If we approximate the permeability coefficient K by K_1 obtained in the azimuthally symmetric case at ${}_1$ = ${}_0$, (17), and, using an approximate relationship between the radius of curvature R sin ${}_0$ of the boundary contour @${}_1$ and the pendular ring volume[2], one can show that

$$\sin^2 {}_0 \quad {}^2_0 = \sqrt{s \quad s_0}:$$

Therefore, finally

$$K(s) \quad 2\frac{R}{R}\frac{k_m}{j\ln(s \quad s_0)j}: \qquad (18)$$

As it follows from (18), the distinctive particle shape results in logarithmic correction to the main non-linear superfast-diffusion coefficient $D(s) = \frac{D_0(s)}{(s \quad s_0)^{3=2}}$, such that

$$D(s) / \quad \frac{1}{j\ln(s \quad s_0)j(s \quad s_0)^{3=2}}:$$

Apparently, the correction will mitigate to some extent the divergent nature of the dispersion at the very small saturation levels s s_0, smoothing out the characteristic dispersion curves.

C. Surface permeability of a chain of spheres in the case of azimuthally symmetric domain boundaries

Consider now how the problem can be formulated in the case of several particles arranged in a single chain, as is illustrated in Fig. 3 in the case of two coupled by the bridge particles. To create the flow in the system of two coupled particles, one can set pressure di 626rnsure di 626rnsure di 626rnsure di 626rnsur48(suc)27(h)-23903(the)-4

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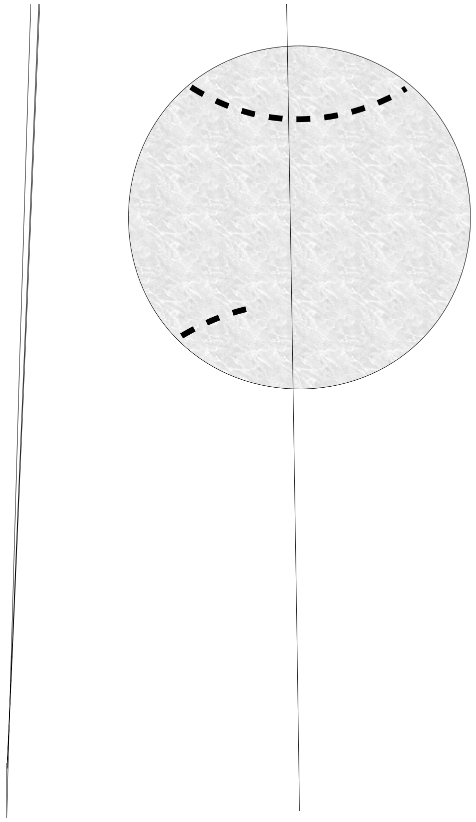

FIG. 3. Illustration of the solution domains in a system of two coupled spherical particles and their geometric arrangements.

where

$$^{(2)}_{0} = \frac{1}{2\ln\frac{1+\cos\quad_{0}}{1\quad\cos\quad_{0}}}:$$

One can now calculate total flux and define permeabil-

III. SURFACE PERMEABILITY OF A RANDOMLY PACKED PARTICLE ENSEMBLE

In real systems, the particles are interconnected randomly, so that the effects of tortuosity should substantially affect the permeability of the system[1,15–17]. To analyse those effects, we consider an ensemble of spherical particles randomly packed, as is shown in Fig. 4. The randomly packed configuration of approximately 3000 – 7000 particles has been generated by means of a molecular dynamics technique by applying a constant force to every particle placed in a box with reflecting boundaries (in the perpendicular direction to the box side), and interacting via the Lennard-Jones potential with different characteristic length scales R distributed normally, that is with the probability of the particle radius $W(R) \propto \exp\left(-\frac{(R-R_0)^2}{R^2}\right)$ at $R/R_0 = 0.3$. In this study, there were particles with three different characteristic dimensions $R_1 = 1.3R_0$, $R_2 = R_0$ and $R_3 = 0.7R_0$. The resultant porosity in the configurations was about 48%.

To obtain the configuration, the particle temperature controlled by the thermostat has been gradually reduced to bring the system to a minimum energy, frozen state. A representative sample volume element with dimensions L_x^B, L_y^B, L_z^B then was cut off the system, as is illustrated in Fig. 4, containing $N_S = 13 - 17$ particles, see Table I for details. We have generated several statistically independent sample configurations, and, as in the previous examples, set constant pressure difference $U_2 - U_1$ at the boundaries of the sample elements, Figs. 4 and 6.

The Laplace-Beltrami method then has been applied after establishing the position of the liquid bridges coupling the particles in the sample. Two particles (of radii R_1 and R_2) are assumed to be coupled by a liquid bridge if the distance between their centres r was only slightly larger than the sum of their radii

$$R_1 + R_2 \leq r < R_1 + R_2 + 0.05\max(R_1, R_2).$$

The size of a single liquid bridge footprint H_B

particles in the sample may significantly increase computational time to obtain highly resolved numerical solutions, while at the same time would not substantially rey

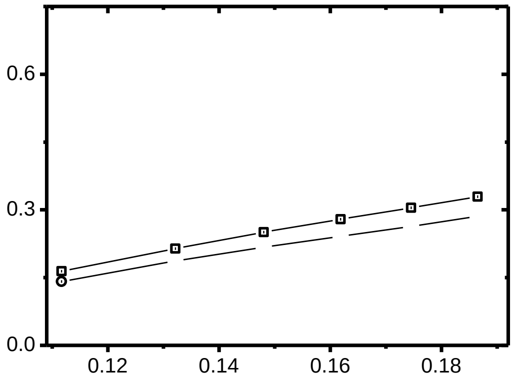

FIG. 8. Reduced total flux $Q_T = S_0 Q_0$, $Q_0 = p_0 \ {}_R \frac{m}{} \frac{U_2 \quad U_1}{L^B_z}$ and $S_0 = L^B_x L^B_y$, as a function of $_0(H_B = 2R_0)$. The error bar indicates the statistical error, which is expected due to the fluctuations of the number of particles in the samples.

have described.

As before, we are going to find a weak solution to a system of the Laplace-Beltrami equations

$$_0^{(k)} u_k = 0$$

defined on each particle domain $_0^{(k)}$, as in in Fig. 6. On the internal boundaries of the domains we set up continuity conditions, for example on $@_3^{(2)}$ and $@_2^{(3)}$

$$u_2 \, j_{@_2^{(3)}} = u_3 \, j_{@_3^{(2)}} = \text{const}; \qquad (36)$$

$$\oint_{@_2^{(3)}} r \; u_2 \; n_{s_2} j_{@_2^{(3)}} \, dl = \oint_{@_3^{(2)}} r \; u_3 \; n_{s_3} j_{@_3^{(2)}} \, dl : \quad (37)$$

While on a few external boundaries, Dirichlet boundary conditions are set.

The numerical solution allows to calculate the total flux through the system by summing up the fluxes pas

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