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Permeability versus conductivity for porous media with wide distribution of pore sizes

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Using recent results on random resistor networks, we derive the relation between the permeability and the conductivity of liquid-saturated porous materials, presumably exact in the limit of a wide distribution of pore sizes. This relation also applies to some continuum percolation models, and the range of its universality is discussed. Some discrepancy is found with the previous analysis of Katz and Thompson [Phys. Rev. B **34**, 8179 (1986)]. Mechanisms are proposed to recover agreement with their experiments. The pore-size parameter Λ characterizing surface effects is computed.

A long-standing problem in the study of porous media is to relate the *permeability* k of a porous material saturated by a single liquid phase to its *conductivity* σ when saturated with a solution of conductivity σ_0 [k is defined by the Darcy equation $\mathbf{v}=(-k/\eta)\nabla p$—the hydraulic analog of Ohm's law, where η is the fluid viscosity, $\mathbf{v}$ is the fluid velocity, and ∇p is the pressure gradient]. Since the permeability has unit length squared, one can always write

$$k=L_0^2\frac{\sigma}{\sigma_0}, \tag{1}$$

and the problem is to relate the characteristic length L_0 to other properties of the material considered. Usually, L_0 was identified[1] as a macroscopic geometrical quantity, proportional to the *total* pore volume-to-surface ratio (hydraulic radius). Recently, two alternative approaches were proposed. Johnson, Koplik, and Schwartz (JKS) (Ref. 2) defined a new pore-size parameter Λ, as an effective pore volume-to-surface ratio, through $\Lambda=2(\int|\mathbf{E}|^2dV/\int|\mathbf{E}|^2dS)$: The weighting by the electric field $\mathbf{E}$ (solution of the potential problem in the pore space) eliminates contributions of isolated regions which do not participate in transport. They conjectured, and provided some experimental evidence, that $L_0^2=c_0(\Lambda^2/8)$, where c_0 is of order unity for a variety of materials. Katz and Thompson (KT) (Ref. 3) instead related L_0 to the *percolation diameter* δ_c, by definition such that the set of pores of diameter larger than δ_c is exactly at the percolation threshold (obviously a *microscopic* characteristic length). Using the analogy between the hydraulic and the electric transport problem, KT modeled the material by a network of cylindrical pipes of random diameters δ and lengths $1\sim\delta$, of electric and hydraulic conductances $g_e(\delta)\sim\delta$ and $g_h(\delta)\sim\delta^3$, respectively. Then they used the argument of Ambegaokar, Halperin, and Langer,[4] who approximated the conductivity σ of a network with a wide distribution of bond conductances, by the *percolation conductance* g_c such that the fraction $p(g_c)$ of bonds in the network of conductance larger than g_c is exactly the percolation concentration, e.g., $p(g_c)=p_c$ (we call the set of these bonds the percolating set). KT tried to refine this argument by approximating σ (as in Ref. 5) by its lower bound $\sigma_{\min}=Kg[p(g)-p_c]^t$ optimized by allowing g to vary in a neighborhood of g_c (t is the percolation conductivity exponent). In the limit of a wide distribution of pore sizes δ, they obtained $L_0^2=c\delta_c^2$ with $c=c_{\mathrm{KT}}=4.4\times10^{-3}$. The beauty of the idea is that δ_c can be simply measured by mercury injection experiment: At a given pressure P, only pores with $\delta>(4\gamma\cos\theta)/P$ can be filled (see Ref. 3).

Although formula (1) with $L_0^2=c_{\mathrm{KT}}\delta_c^2$ agrees remarkably well with the KT experiments, it suffers from several problems. First, the way it is derived is unsatisfactory, since KT optimize a lower bound: As a result, for a given model, the coefficient c_{KT} is only approximate. Both conceptually, and for comparison with experiments, a correct derivation is needed. Then, one can ask if it is really *predictive:* (i) does it depend on the details of the network used to model the total pore space? (ii) how "wide" the distribution of pore sizes has to be (iii) how faithful are conductance network models, and the electric versus hydraulic analogy? In Ref. 6, point (i) was addressed by the same approximate method that KT used, and the relation between Λ and δ_c was discussed.

In this paper, we obtain exact expressions for the length L_0 in Eq. (1) for several models. Strictly, it applies only to materials whose total pore space can be modeled by some network close to particular percolation situations, such as (i) a three-dimensional (3D) network with a distribution of (electric and hydraulic) bond conductances which is wide on a logarithmic scale (ii) more standard percolation situations were only a fraction $p\simeq p_c$ of the bonds in the network are conducting but with an accumulation of small conductances $P(g)\sim g^{-\alpha}$ with α sufficiently close to 1. Situation (i) is considered by KT and qualitatively justified in some rocks where log-normal distributions of pore sizes occur. Situation (ii) arises in some continuum percolation models, like the 3D Swiss cheese model.[7] As we discuss, the critical exponent involved is *not* t. We find that the KT relation $L_0^2=c\delta_c^2$ holds only for models such that the electric and hydraulic percolation sets are *identical* (models I and II below). In that case, and in situation (i), the factor c is independent of the topology of the network and of details of the disorder, and weakly dependent on the geometry. This comes from the universality of the percolation fixed point for wide distributions. Also in that case the parameter Λ is found to be close to L_0. However, we find that if the shape

of the pores (or their lengths) also varies widely, the order of magnitude of L_0 decreases from δ_c towards the *typical* diameter δ_{typ}, and simultaneously Λ becomes very different from L_0.

The present analysis is based on the results of two recent works, one by Tyc and Halperin[8] and the other by the author[9] on random resistor networks. It was proposed that, if ln$g$ is widely distributed, the conductivity σ takes the universal asymptotic form[8]

$$\sigma \cong K g_c [g_c P(g_c)]^y , \tag{2}$$

where K is a constant depending only on the topology of the network, and y depends only on its dimensionality. All details of the (smooth) distribution $P(g)$ are irrelevant, except the value $P(g_c)$, at the condition that $g_c P(g_c) \ll 1$ and $gP(g)$ be slowly varying in a large neighborhood of g_c (e.g., of appreciable total weight). A useful model to study is $g = g_0 \exp(\lambda x)$ with a fixed distribution $D(x)$ of x: one has then $g_c P(g_c) = D(x_c)/\lambda \ll 1$ if λ is large. We have shown[9] by several arguments that $y = (d-2)\nu$ for $d \leq 6$, where ν is the percolation correlation length exponent. In three dimensions $y \cong 0.88$. This precise value for y allows us to also apply Eq. (2) to distributions with a weight $(1-p)\delta(g)$ at $g=0$, and a tail for small conductances $P(g) \sim g^{-\alpha}$. This gives $\sigma \sim (p-p_c)^{t'}$ with $t' = t(\alpha) = (d-2)\nu + 1/(1-\alpha)$, for α such that $t(\alpha) < t$, a result previously obtained by Machta.[10] Although the exponent is correctly predicted, the amplitude K probably becomes (weakly) α dependent (ln$g$ is widely distributed only if α is close to 1). In Swiss-cheese-like continuum percolation models, transport is controlled by narrow necks of conductance $g \sim d^z$, and the size parameter d is such that $P(d=0) > 0$. The corresponding exponent is $t' = \max[t, (d-2)\nu + z]$.

Let us now compute L_0 from Eq. (2). The network of pores is modeled, as in Ref. 6 by a network of interconnected cylinders of diameters δ and length l, chosen according to some distribution $P(\delta, l)$. The electric conductance of a cylinder is $g_e(\delta, l) = \pi\delta^2/4l$ and the hydraulic conductance is $g_h(\delta, l) = \pi\delta^4/128l$. Surface conduction processes are neglected here, and the pressure gradient is supposed to be very small in the regions connecting the cylinders; one probably also needs $l \gg \delta$. Several models can be considered:

Model I. δ and l are simply proportional: $l = a\delta$ with a given a (this is the case considered by KT). A pore is characterized by a single (random) length scale. Then $g_e(\delta) = a_e \delta$ and $g_h(\delta) = a_h \delta^3$. In that case both percolation conductances $g_{e,c}$ and $g_{h,c}$ correspond to the percolation diameter δ_c (such that the fraction of pores of diameter larger than δ_c is p_c). One can compute the distribution $P_e(g_e) = P(\delta)[dg_e(\delta)/d\delta]^{-1}$ and, similarly, $P_h(g_h)$. From Eq. (2) one has, in the limit of a very wide distribution of diameters δ

$$k = \frac{g_h(\delta_c)}{g_e(\delta_c)} \left(\frac{d \ln g_e(\delta)}{d \ln \delta} \bigg|_{\delta=\delta_c} \right)^y \left(\frac{d \ln g_h(\delta)}{d \ln \delta} \bigg|_{\delta=\delta_c} \right)^{-y} \frac{\sigma}{\sigma_0} , \tag{3}$$

all network-dependent or distribution-dependent factors cancel out in this calculation, and we are left with

$$k = \frac{\delta_c^2}{32} \left(\frac{1}{3} \right)^y \frac{\sigma}{\sigma_0} = c_{\mathrm{I}} \delta_c^2 \frac{\sigma}{\sigma_0} , \tag{4}$$

with $c_{\mathrm{I}} \cong 11.8 \times 10^{-3}$. This is larger than the KT result by almost a factor of 3. However, as pointed out in Ref. 6, there is an inconsistency in the precise way KT carried out their calculation, and for the same model Banavar and Johnson obtained instead $c'_{\mathrm{KT}} = 7.68 \times 10^{-3}$. The final exact result is thus roughly 1.5 larger than using the approximation of Ref. 5.

Model II. l is kept fixed ($l = l_0$) and δ is widely distributed: This leads again to Eq. (4) with the same definition of δ_c and $c_{\mathrm{II}} = \frac{1}{32}(2)^{-y} \cong 16.9 \times 10^{-3}$. Again, it is larger by a factor of 1.5 than Banavar and Johnson's result,[6] who considered the same model.

Model III. If the pores have some well-defined curvature, one can still use the cylinder approximation with $l = \sqrt{R\delta}$. If δ has a wide continuous distribution, then from Eq. (3) and since $g_e \sim \delta^{3/2}$ and $g_h \sim \delta^{7/2}$, one again recovers Eq. (4) with $c_{\mathrm{III}} = \frac{1}{32}(\frac{3}{7})^y \cong 14.8 \times 10^{-3}$. The length scale R does not appear in the final result. Let us consider another case where, as in the Swiss cheese model, the diameter δ has a probability $(1-p)$ to be zero with a continuous distribution for $\delta > 0$, of finite nonzero density $P(\delta \to 0)$. The conductivity vanishes as a function of $(p-p_c)$ with exponent $t' = \nu + \frac{3}{2}$ and the permeability with exponent $e' = \nu + \frac{7}{2}$. As usually stated, in this regime $k \sim (\sigma/\sigma_0)^x$, and we find $x \simeq 1.84$. However, it is also interesting to write the relation between k and σ as in Eq. (1). As explained above, one can still use Eq. (2) keeping in mind that the constant K now depends weakly on some details of the distribution of conductances. One obtains $L_0^2 = c\delta_c^2$ with $c = (K_h/K_e) \times 14.8 \times 10^{-3}$ and δ being still the percolation diameter. For reasons of continuity in the limit $\alpha \to 1$, the ratio of the two constants K_h and K_e in Eq. (2) corresponding to the electric and hydraulic cases, respectively, is likely to be close to unity. Thus, for the Swiss cheese model in 3D, we expect the constant c to be of the same order of magnitude than in models I and II. This is true only if both permeability and conductivity exponents are in the regime $t(\alpha) < t$. This would not be true for the inverted Swiss cheese model.[7]

The characteristic length Λ introduced by JKS can be computed in the limit of wide distributions of pore sizes. It is obtained[2] by introducing a surface conductivity Σ and expanding the conductivity of the system to first order in $\epsilon = \Sigma/\sigma_0$ since $\sigma(\Sigma) = \sigma(\Sigma=0)[1 + (2/\Lambda)\epsilon]$. The conductance $g(\Sigma)$ of a cylinder becomes: $g(\Sigma) = g(\Sigma=0)[1 + (4/\delta)\epsilon]$. Let us consider model I. The percolation conductance still corresponds to δ_c, but is now $\ln g_c(\Sigma) = \ln g_c(0) + (4/\delta_c)\epsilon$. From Eq. (2) one easily gets: $\ln\sigma(\Sigma) - \ln\sigma(0) = [(4+4y)/\delta_c]\epsilon \equiv (2/\Lambda)\epsilon$. Thus we obtain the relation $\Lambda = \delta_c/2(1+y)$ for model I. It is identical to the result of Ref. 6, with t replaced by y, but this is presumably the exact result. Similarly, model II leads to $\Lambda = \delta_c/(2+y)$. The corresponding constants c_0 appearing in the relation $k = c_0(\Lambda^2/8)(\sigma/\sigma_0)$ are, respectively, $c_{0\mathrm{I}} = 32(1+y)^2 c_{\mathrm{I}} \cong 1.34$ and $c_{0\mathrm{II}} = 8(2+y)^2 c_{\mathrm{II}} \cong 1.12$. For model III, our conclusions agree with those of Ref. 11.

The constant c depends on the (disordered) geometry

(although in general not of the topology of the underlying network). However, for very different models (models I, II, or III) it remains of the same order of magnitude (by contrast with the pore size which varies over several orders of magnitude). Thus, there is some universality in the relation $L_0^2 = c\delta_c^2$. Note that short-range statistical correlations in the geometry do not change this relation. Similarly $\Lambda^2/8$ remains remarkably close to L_0^2 (c_0 is close to its value for a single cylinder $c_0 \equiv 1$). It is important to determine the stability of these results when more parameters are added to describe the disorder of the material. For instance, in any realistic material, one cannot expect the length of the pores (or the necks) to be exactly a given function of the diameter δ without any fluctuations, as supposed in models I–III.

We still consider a network of cylindrical pipes, and we choose for each cylinder both diameter and length (δ, l) according to a ln-normal distribution (for simplicity) with a 2 by 2 covariance matrix V. Let us consider as an example, the case where $\ln\delta$ and $\ln(\delta/l)$ have independent normal distributions of variances v and v', respectively. This corresponds to uncorrelated disorder on the diameter and on the aspect ratio δ/l. We suppose that v is large, so that both electric and hydraulic conductances are widely distributed. The variances of $\ln g_e$ and $\ln g_h$ are $v_e = v + v'$ and $v_h = 9v + v'$, respectively. From Eq. (2) the electrical conductivity behaves for large v_e as $\sigma/\sigma_0 = K g_{e,c} [\Phi(x_c)/\sqrt{v_e}]^y$ where $\Phi(x) = (2\pi)^{-1/2} \exp(-x^2/2)$ is the normal distribution and x_c is such that $\int_{x_c}^{\infty} \Phi(x)dx = p_c$ ($x_c > 0$ in 3D, and $x_c \cong 0.77$ for a cubic lattice). A similar relation holds for the permeability. The new feature is that the percolation conductance $g_{e,c}$ does not correspond simply to δ_c. One can define δ_c^* and l_c^* such that $g_{e,c} = (\pi/4)(\delta_c^{*2}/l_c^*)$ and $g_{h,c} = (\pi/128)(\delta_c^{*4}/l_c^*)$. However, now $\delta_c^* \neq \delta_c$ contrarily to models I–III. It is easy to obtain L_0 in Eq. (1) as

$$L_0^2 = \tfrac{1}{32} \exp[x_c(\sqrt{9v+v'} - \sqrt{v+v'} - 2\sqrt{v})] \times (\sqrt{v+v'}/\sqrt{9v+v'})^y \delta_c^2 , \quad (5)$$

the exponential factor being precisely $(\delta_c^*/\delta_c)^2$. In the limit $v' = 0$ one recovers the result of model I. When the disorder on the aspect ratio δ/l is small compared to the disorder on δ, expanding Eq. (5) to first order in v'/v, one gets the dominant contribution

$$c \cong c_1 \exp[-\tfrac{1}{3} x_c \sqrt{v} (v'/v)] .$$

This becomes nonnegligible as soon as $v' \sim \sqrt{v}$. Now, let the ratio v'/v vary from 0 to ∞: the factor involving y varies slowly between 0.38 and 1. On the other hand, since $x_c\sqrt{v} = \ln\delta_c - \langle\ln\delta\rangle$, the exponential factor varies between 1 and $(\delta_{\text{typ}}/\delta_c)^2$, where δ_{typ} is the typical diameter $\exp(\langle\ln\delta\rangle)$: Thus, when the disorder on the aspect ratio dominates the disorder on the diameter, the characteristic length becomes the *typical* diameter which can be *much smaller* than the percolation diameter.

One can consider more general situations. Let $x_1 = \ln\delta - \langle\ln\delta\rangle$ and $x_2 = \ln l - \langle\ln l\rangle$ be Gaussian variables such that $\langle x_i x_j\rangle = V_{ij}$. Then a short calculation leads to

$$\frac{k}{\sigma} = (v_e/v_h)^{y/2} \delta_c^2 \exp[x_c(\sqrt{v_h} - \sqrt{v_e} - 2\sqrt{V_{11}})]$$

with $v_h = 16V_{11} - 8V_{12} + V_{22}$ and $v_e = 4V_{11} - 4V_{12} + V_{22}$. The extension of model II where δ and l have independent ln-normal distributions of variances v and v', respectively, corresponds to $V_{11} = v$, $V_{22} = v'$, $V_{12} = 0$, and model II itself to $v' = 0$. Model III with a widely varying curvature also corresponds to some choice of the V_{ij}. In all cases one finds that the prefactor (with exponent y) varies very slowly, but that due to the exponential factor the length scale L_0 is progressively reduced from δ_c to $\delta_{\text{typ}} = \delta_c \exp(-x_c\sqrt{V_{11}})$ as V_{22} increases.

The parameter Λ can also be computed for these models. After a straightforward calculation one finds, in the case corresponding to formula (5)

$$\Lambda = \frac{\delta'}{2(1+y)} \quad \text{with } \delta' = \delta_c \exp\left[x_c\left(\frac{v}{\sqrt{v+v'}} - \sqrt{v}\right) - \frac{vv'}{2(v+v')}\right], \quad (6)$$

and in the general case

$$\Lambda = \frac{\delta_c}{2(1+yw/v_e)} \exp\left[x_c\left(\frac{w}{\sqrt{v_e}} - \sqrt{V_{11}}\right) - \frac{\det V}{2v_e}\right],$$

with $w = 2V_{11} - V_{12}$ and v_e as above. Two cases can be distinguished.

(i) $\det V = 0$: then, there is a functional relation between l and δ of the type $l = R^{1-\beta}\delta^{\beta}$ ($\beta = 1, 0, \frac{1}{2}$ in models I–III, respectively) where R is some fixed length. The (dominant) exponential factor in the expression of Λ is then $\exp\{x_c\sqrt{V_{11}}[(2-\beta)/(|2-\beta|) - 1]\}$ which has to be compared with the analogous factor in the expression of L_0 from the previous paragraph: $\exp[\frac{1}{2} x_c\sqrt{V_{11}}(|\beta - 4| - |\beta - 2| - 2)]$. There are three regimes: if $\beta < 2$ then $\Lambda \cong L_0 \cong \delta_c$, if $2 < \beta < 4$ then $\Lambda \cong \delta_{\text{typ}}^2 \delta_c^{-1}$, but $L_0 \cong \delta_{\text{typ}}^{\beta-2} \delta_c^{3-\beta}$ if $\beta > 4$, $\Lambda \cong L_0 \cong \delta_{\text{typ}}^2 \delta_c^{-1}$. These results have obvious interpretations: The hydraulic and electric percolation sets are identical for $\beta < 2$ and for $\beta > 4$. However, for $2 < \beta < 4$, these two sets are unrelated; then L_0 and Λ split apart.

(ii) $\det V \neq 0$: the term $\det V$ in the exponential factor dominates the behavior of Λ. As soon as v' in Eq. (6) is nonzero, Λ becomes a very bad approximation to L_0. For instance for $v = 20$, the increase of v'/v from 0 to 0.2 already leads in Eq. (6) to a variation of the ratio Λ/L_0 of one order of magnitude (by contrast the ratio L_0/δ_c varies by 5%).

In conclusion, although the experimental data presented by Katz and Thompson remarkably agree with

$L_0^2 = c_{KT}\delta_c^2$ over six decades, our theoretical predictions, for the same kind of model as they consider (models I–III), differs from theirs by a factor of 3 (or 4). This is out of their error bars but still the prediction is not far from the experiments. It would be interesting to understand the physical origin of this deviation. As we have shown, this deviation can still be accounted for in the context of a wide distribution of pore sizes inside the class of "random cylinder" models: an additional disorder on the length, or the aspect ratio of the pores decreases L_0 from the percolation diameter to the typical diameter. From Eq. (5), L_0^2 decreases by a factor of 3 if, for example, $(v,v'/v)=(20,1.8)$ or (100,0.5); simultaneously, if this scenario is correct, the JKS length Λ should become very different from L_0. Independent experimental determinations of L_0, Λ, and δ_c on the same sample of rock do not exist at present, but are clearly needed. Of course, many other effects could also account for the deviation. First, one expects "finite-width" corrections to Eq. (2) if the distribution of pore sizes is not wide enough. A theoretical estimation of these corrections is needed. Also, if cylinders are replaced by objects of more complex shape, slightly different factors arise in Eq. (4), but more importantly, the experimental determination of δ_c (which uses the Washburn equation in a cylindrical geometry[3]) has to be slightly corrected. Finally, to determine whether the initial percolation arguments of KT can really be applied to transport in rocks, we suggest experiments on samples of intermediate size. This is because, in the context of random resistor networks, if $1 \ll L \ll \lambda^{\nu}$ where λ is the width of lng, then the finite-size percolation conductance is an excellent approximation to the conductance of the sample,[9] even if λ is not extremely large. The ratio $G_h(L)/G_e(L)$, which for finite L fluctuates from sample to sample, should then be strongly correlated to $c\delta_c^2(L)$ for the same sample.

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