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Gradient clogging in depth filtration

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We investigate clogging in depth filtration, in which a dirty fluid is ''cleaned'' by the trapping of dirt particles within the pore space during flow through a porous medium. This leads to a self-generated gradient percolation process that exhibits a power-law distribution for the density of trapped particles at downstream distance x from the input. To achieve a nonpathological clogging (percolation) threshold, the system length L should scale no faster than a power of $\ln w$, where w is the system width. Nontrivial behavior for the permeability arises only in this extreme anisotropic geometry. [S1063-651X(98)51208-1]

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Depth filtration is a mechanism for cleaning a dirty fluid by passing it through a porous medium [1–3]. The medium promotes efficient filtering both by increasing the area available for the trapping of suspended particles, as well as the exposure time of the suspension to the absorbing surfaces. This mechanism is therefore widely used in a variety of biological, chemical, and engineering separation processes [1]. While filtration has been extensively studied, many of these investigations are empirical or numerical and a clear relation between microscopic mechanisms and macroscopic behavior has not yet emerged. In this Rapid Communication, we provide an intuitive, geometrical description for depth filtration which provides insights about the clogging process and leads to phenomenology outside the realm of classical percolation and related breakdown processes.

There are two salient features of depth filtration that contribute to its unusual phenomenology. First is the feedback between the continuous evolution of the medium by trapping of particles, and the subsequent modification of the flow field by trapped particles. Second, clogging occurs preferentially in the upstream end of the network. This arises simply because a particle proceeds downstream only until the first encounter with a sufficiently small pore which blocks the particle. Thus, particles rarely reach downstream pores.

Each blockage event causes a small reduction in the filter permeability, and ultimately a clogging threshold is reached where the permeability vanishes. This clogging is gradient driven, as the fraction of blocked bonds has a power-law dependence on the longitudinal co-ordinate. Such a distribution should be observable, for example, when opaque particles pass through a glass bead pack [4]. Due to the gradient, filter clogging is radically different than the failure of homogeneous disordered media [5], where the formation of breakdown paths does not have a systematic gradient. While previous studies indicated that the filter permeability vanishes as a power law near the clogging threshold, with an exponent different from that of classical percolation [6,7], we interpret the clogging process as a situation where the percolation threshold (fraction of open pores) is very close to unity and where the permeability does not have power-law behavior.

Of the many microscopic interactions that underlie filtration, we focus on size exclusion [4,8], where a particle of radius r_{particle} is trapped within the first pore encountered whose radius satisfies $r_{\mathrm{pore}} < r_{\mathrm{particle}}$. This size exclusion is the dominant effect in processes such as gel permeation in porous media and liquid chromatography. While other influences, such as van der Waals, hydrodynamic, and electrostatic interactions, etc., may be important, their faithful modeling is complex [1] and complicates the identification of the governing mechanism for a given macroscopic property. Our approach is to retain size exclusion as the only trapping mechanism in a geometric modeling of filtration and develop physical intuition for clogging from this idealized description.

The connectivity of the medium is described by the quasi-one-dimensional ''bubble'' model. This system consists of L links in series, in which each link is a parallel bundle of w bonds and each bond represents a pore (Fig. 1). This model can be viewed as a square lattice in which all perpendicular bonds are ''shorted,'' so that w can be identified as the system width. This model was introduced to account for the breaking of fibers [9] and the extremal voltages in resistor networks [10]. An appealing feature of this model in the context of percolation is that it exhibits finite-dimensional behavior when L scales as e^w. Namely, if each bond is randomly occupied with probability p, a percolation threshold p_c strictly between 0 and 1 arises, and associated critical exponents can be easily computed [10]. We adopt this bubble model as a convenient way to describe the reduced connectivity of the medium as individual bonds are blocked.

To adapt this model to filtration, we posit that each bond has a radius r drawn from a specified distribution, with a volumetric flow rate proportional to $r^4 \nabla p$ (Poiseuille flow), where ∇p is the pressure gradient in the bond. Dynamically neutral suspended particles move through the medium at a rate governed by this local flow. We assume perfect mixing at each node, in which a suspended particle has a flow induced probability proportional to r_i^4 to enter an unblocked bond of radius r_i in the next downstream bundle. The par-

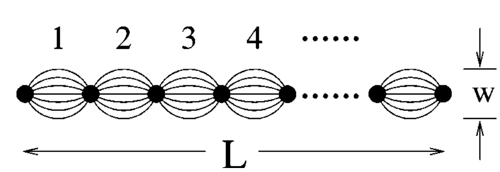

FIG. 1. Bubble model. Each line represents a separate fluid-carrying pore and fluid mixes completely at each node.

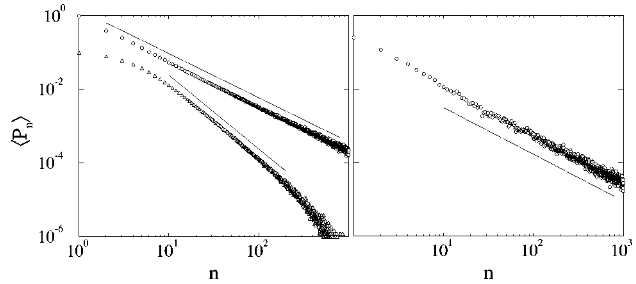

FIG. 2. (a) Trapping probability $\langle P_n \rangle$ vs n at the percolation threshold for a bubble model of width $w=50$ for coincident and uniform distributions of particle and bond radii. Shown are data, based on 10^4 configurations, for $(a,b)=(0,1)$ (○) and $(a,b)=(0.7,1.0)$ (△), with the latter divided by 10 for visualization. The straight lines have slopes $-6/5$ and -2 respectively. (b) Trapping probability, based on injecting 4×10^6 particles into an unperturbed 500×1000 square lattice whose axes are oriented at 45° with respect to the average flow, for coincident, uniform particle and bond radius distributions on (0,1). The straight line has slope -1.27.

ticles, whose radii are also drawn from a distribution, are injected singly and tracked until each is trapped or escapes the system. Upon capture, the particle is defined to block the bond completely so that there is no further fluid flow in this bond. More realistically, particle trapping may not completely block a bond, but rather, the permeability should be reduced to a nonzero value. However, this partial blocking has the same asymptotic behavior as in complete blockage, since large-distance properties are governed by the smallest particles for which the state of the already modified pore space is relatively unimportant. After each blockage event, the new flow field is computed to determine the trajectory of the next suspended particle.

This system exhibits three regimes of behavior. For pores typically smaller than particles (subcritical), the particles get trapped almost immediately and rapid clogging ensues. Conversely, for pores typically larger than particles (supercritical), a steady state is eventually reached for a finite length system, in which the smallest pores are blocked and the suspension flows freely through the remaining unblockable pores. These cases can be viewed as corresponding to poor filter performance. At the boundary between these regimes is the critical case, where the particle and pore radius distributions overlap substantially. Here, particle trapping is gradual, with considerable penetration of the medium before clogging is reached. This may be viewed as efficient filtration because of the large number of particles filtered before clogging and the relatively long filter lifetime. Thus, both from practical and theoretical perspectives, the critical case is the most interesting.

For simplicity and concreteness, consider a uniform distribution of both particle and bond radii in the range $[a,b]$. More general continuous distributions can be straightforwardly treated, but little new qualitative insight emerges. Let us first determine the spatial distribution of trapped particles during filtration. The gradient nature of the trapping process implies that the number of blocked bonds in downstream bubbles remains small, even at the percolation threshold (see Fig. 2). We therefore employ an ''unperturbed'' approximation in which the initial bond radius distribution is used throughout the clogging process. Within this approximation and assuming Poiseuille flow, the probability that a particle of radius r gets trapped in a bubble is, for large w,

$$P_<=\frac{\int_a^r r'^4 dr'}{\int_a^b r'^4 dr'}=\frac{r^5-a^5}{b^5-a^5}. \tag{1}$$

(Exact calculation shows that this large-w form is asymptotically correct for $w\geqslant 5$.) Consequently, the probability that this particle gets trapped in the nth bubble is $P_n=(1-P_<)^{n-1}P_<$, which decays exponentially in n. Averaging over the distribution of particle radii gives

$$\begin{aligned}\langle P_n\rangle &= \int_a^b \left(1-\frac{r^5-a^5}{b^5-a^5}\right)^{n-1}\frac{r^5-a^5}{b^5-a^5}\,\frac{dr}{b-a},\\ &= \frac{1}{5}\,\frac{(b^5-a^5)}{(b-a)}\int_0^1 \frac{v(1-v)^{n-1}dv}{[v(b^5-a^5)+a^5]^{4/5}},\end{aligned} \tag{2}$$

where $v=r^5-a^5/b^5-a^5$.

Depending on the lower cutoff a, there are two different asymptotic behaviors for this trapping probability. If $a\neq 0$, the integral is elementary, while if $a=0$, the integral reduces to the β function [11] so that $\langle P_n\rangle=\Gamma(6/5)\Gamma(n)/5\Gamma(n+6/5)$, where $\Gamma(n)$ is the gamma function. In the large-n limit, these give

$$\langle P_n\rangle \simeq \begin{cases}\dfrac{b^5-a^5}{5a^4(b-a)}n^{-2}, & a\neq 0,\\ 0.1836\ldots n^{-6/5} & a=0.\end{cases} \tag{3}$$

From the denominator in the second line of Eq. (2), the crossover between the $n^{-6/5}$ and n^{-2} behaviors occurs when $v(b^5-a^5)<a^5$, or equivalently, $n>n^*=(b/a)^5$. While the exponents 6/5 and 2 are specific to the uniform radius distribution and the flow-induced bond entrance probability, the existence of the power law is generic and requires only the overlap of the bond and particle radius distributions. For example, for the Hertz distribution of particle and bond radii, $p(r)=2re^{-r^2}$, $\langle P_n\rangle\propto n^{-4/3}$.

Qualitatively similar behavior for $\langle P_n\rangle$ occurs in lattice networks. In the spirit of our unperturbed approximation, we focus on the spatial distribution of the initially injected particle. Later particles exhibit nearly the same spatial distribution of trapping location, but much more time is needed for computing this distribution, since the network permeability must be recalculated after each trapping event. For $n\gtrsim 10$, a best fit power law to the data is $\langle P_n\rangle\sim n^{-\mu}$, with $\mu\approx 1.27$ [Fig. 2(b)], fortuitously close to the bubble model exponent.

In the supercritical regime (bonds larger than particles), $\langle P_n\rangle$ exhibits near-critical behavior, except that some particles can escape from the system. Conversely, in the subcritical regime (bonds smaller than particles), Eq. (2) gives $\langle P_n\rangle\propto\exp[-n(a^5-A^5)/(B^5-A^5)]$, where (a,b) and (A,B) are, respectively, the ranges of the particle and bond radius distributions. As $a-A\to 0$, the decay length $(B^5-A^5)/(a^5-A^5)$ diverges and power-law behavior of $\langle P_n\rangle$ is recovered. It is in this sense that coincident bond and particle radius distributions correspond to a critical phenomenon.

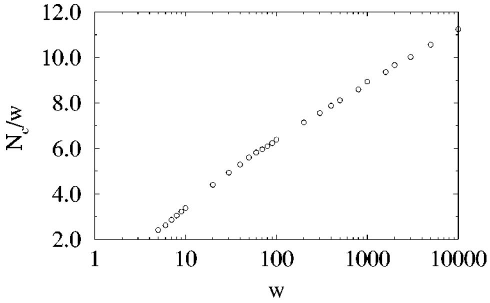

FIG. 3. Number of particles injected at percolation (normalized by w) vs ln w. The number of configurations is 10^5 for $w<10^2$, 10^4 for $10^2\leqslant w\leqslant 10^3$, and 10^3 for $w>10^3$.

Let us now determine the number of particles that need to be injected to reach the clogging (percolation) threshold; see Fig. 3. For the bubble model, this means that all bonds in a single bubble are blocked. Since $\langle P_n\rangle$ monotonically decreases in n, the probability that all w bonds are blocked in the nth bubble is nonzero only for small n. (Numerically, for $w=100$, for example, the probability of blocking in bubble n is approximately 78.2%, 15.8%, 4.44%, and 1.21% for $n=1$, 2, 3, and 4, respectively.) In the following, we therefore use the approximation that it is only the first bubble that clogs, and that both the particle and bond radius distributions are uniform on [0,1].

We first compute the number of particles that need to be injected into a bond of radius $0<r<1$ before it is blocked [12]. For N particles, the probability that all have their radii in the range $[0,r]$ is r^N. This can be reinterpreted as the probability that the maximum radius among N particles lies between 0 and r. Thus, $r^N=\int_0^r P_N(r')dr'$, with $P_N(r)=Nr^{N-1}$ the probability density that the maximum radius equals r. Thus, the average radius of this largest particle is $\langle r\rangle_N=\int_0^1 rP_N(r)dr=N/(N+1)$. Inverting this relation shows that of the order of $(1-r)^{-1}$ particles need to be injected before a particle of sufficiently large radius enters to block a bond of radius r.

For a single bubble of $w\gg 1$ bonds, the number of particles needed to block bonds whose radii are in the range $[r,r+dr]$ is $w(dr/1-r)$. Consequently, the total number of particles needed to block the bubble is

$$N_c\approx\int_{1/w}^{1-1/w} w\frac{dr}{1-r}, \tag{4}$$

where the above extreme value considerations give, for the largest and smallest bond radii in the bubble, $r_{\max}\approx 1-1/w$ and $r_{\min}\approx 1/w$, respectively. The integral is dominated by the upper limit and gives

$$N_c\propto w\ \ln w. \tag{5}$$

Notice that a naive determination of the threshold from $N_c\langle P_1\rangle=w$ gives $N_c=6w$. The logarithmic factor in Eq. (5) arises from the widest bonds for which many particles need to be injected before blocking occurs.

Simulations on the bubble model indicate that this logarithmic w dependence for N_c is independent of the precise form of the entrance probability for a particular bond and similar details. Thus, if the system size increases isotropically, the percolation threshold $p_c=1-N_c/Lw$ approaches 1, where Lw is the total number of bonds in the system. To obtain a threshold value less than unity requires exponential anisotropy in which $L\sim\ln w$. This result is qualitatively robust with respect to different particle and bond radius distributions. For example, for the Hertz distribution, following analogous computations to those just outlined gives $N_c\propto w(\ln w)^2$. For the square lattice, on the other hand, simulations indicate that N_c is linearly proportional to the system width. This corresponds to a percolation threshold that scales as $1-1/L$. Thus, either L should be constant, or an alternative relation between the bond and particle radius distributions may be appropriate to define criticality for a finite-dimensional network.

Finally, consider the behavior of the permeability during filtration, for which the bubble model again provides a useful description. From the series geometry of the bubble model, the inverse permeability κ can be written as $\kappa^{-1}=\Sigma_{n=1}^{L}(\kappa_n)^{-1}$, where κ_n denotes the permeability of the nth bubble. When a single particle is injected, the permeability of the nth bubble decreases by $\delta\kappa_n$, where the radius-average change in this permeability can be written as

$$\delta\kappa_n=\int_0^1 dr r^4\left(\frac{r^4}{\int_0^1 r^4 dr}\right)\left[\int_r^1(1-u^5)^{n-1}du\right]. \tag{6}$$

The last factor is the probability that a particle of radius greater than r reaches the nth bubble, the second factor is the probability that this particle enters a bond of radius r in bubble n, and the first factor gives the change in κ_n when a bond of radius r is blocked. From scaling, this integral varies as n^{-2}, a result that holds for any entrance probability rule which is proportional to the bond permeability. Simulations on the square lattice also clearly show an n^{-2} dependence for κ_n.

Thus, $\kappa_n\propto w-A/n^2$, where A is proportional to the total number of particles injected. Here $A\to w$ corresponds to clogging (up to logarithmic factors) so that we identify $A-w$ with $p-p_c$. The inverse permeability now becomes $\kappa^{-1}\sim\int_1^L dn(w-A/n^2)^{-1}$. This integral is approximately constant and gives $\kappa^{-1}\sim L/w$, except close to clogging. To estimate the integral in this limit, note that for n close to 1, the integrand is dominated by the divergence in the denominator, while for $n>(A/w)^{1/2}$ the second term in the denominator can be neglected. Splitting the integral according to this prescription gives

$$\kappa^{-1}=\int_1^{n*}\frac{dn}{w-A/n^2}+\int_{n*}^{L}\frac{dn}{w}, \tag{7}$$

with $n*=(A/w)^{1/2}$. The first integral is estimated by defining $v=wn^2/A$ and treating the resulting slowly varying factor of $v^{1/2}$ in the numerator as constant compared to the divergent factor $1/(v-1)$. We thereby obtain

$$\kappa^{-1}\sim\frac{L}{w}\left[1-\left(\frac{A}{w}\right)^{1/2}\frac{1}{L}\ln\left(\frac{w}{A}-1\right)\right], \tag{8}$$

where the correction term in κ^{-1} is manifestly positive near the clogging threshold ($A\rightarrow w$ from below). This crude estimate shows that the permeability of an isotropic system ($L \propto w$) is essentially unaffected by individual bond blocking events until one bubble is nearly completely blocked, after which κ discontinuously drops to zero. If, however, $L \sim \ln w$, then κ decreases logarithmically in $A-w$, or $p-p_c$.

In summary, depth filtration is a gradient-controlled process for which the bubble model provides a simple geometrical description. For coincident bond and particle radius distributions, the number of particles trapped a distance n downstream varies as $n^{-\mu}$, where μ is distribution dependent, but invariably between 1 and 2. From extreme value considerations, the length must scale logarithmically in the system width to have a percolation threshold strictly less than unity. For this geometry, the permeability varies logarithmically in $(p-p_c)$. This may explain previous simulation results of a threshold value p_c close to unity and a permeability rapidly varying in concentration near p_c [4].

The gradient nature of filtration has ramifications for efficient filter design. A relatively wide system $w \gg L$ allows a finite fraction of the medium to actually trap particles, i.e., $p_c<1$. On the other hand, the radius-average probability that a particle escapes a system of length L vanishes as $L^{1-\mu}$ for $\langle P_n\rangle \sim n^{-\mu}$. Thus, a small escape rate and p_c strictly less than unity, both desired properties of a depth filter, cannot simultaneously be satisfied in a spatially homogeneous medium. A more appropriate design would be a filter with a longitudinally varying local permeability, which effectively cancels the gradient in particle trapping.

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