International Journal of Modern Physics C, Vol. 9, No. 8 (1998) 1535–1543

GRADIENT AND PERCOLATIVE CLOGGING IN DEPTH FILTRATION*

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Accepted 28 September 1998

The phenomenon of clogging in depth filtration is investigated, in which a dirty fluid is "cleaned" by the trapping of dirt particles within the pore space during flow through a porous medium. This gives rise to a self-generated gradient percolation process which exhibits a power law distribution for the density of trapped particles at downstream distance x from the input, both in idealized and lattice networks. Implications for efficient filter design are also mentioned.

Keywords: Filtration; Gradient Percolation; Clogging.

1. Introduction

Depth filtration is a process for cleaning a dirty fluid by passing it through a porous medium. The medium promotes efficient filtering by increasing the area available for trapping suspended particles and the exposure time of the suspension to the absorbing surfaces. While filtration is a ubiquitous biological, chemical, and engineering separation process and has been extensively studied,[1–3] many results are empirical or numerical and a clear relation between microscopic mechanisms and macroscopic behavior has not yet emerged.

There are two salient features of depth filtration which contribute to its unusual phenomenology. The first is the feedback between filter evolution by particle trapping and subsequent flow field modification. The other feature relates to clogging occuring preferentially upstream in the filter because particles are injected at one end of the system and proceed downstream until trapping. Thus particles rarely reach downstream pores before clogging occurs. These features lead to a basic dichotomy for filter design: a depth filter should be long for effective trapping; on the other hand, an active filter should be short, as many particles penetrate only a short distance before clogging occurs.

*This paper was presented at the 7th Int. Conf. on the Discrete Simulation of Fluids held at the University of Oxford, 14–18 July 1998.

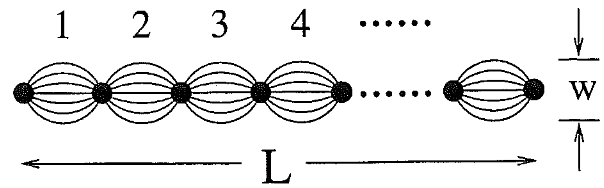

Fig. 1. The bubble model. Each line represents a separate fluid-carrying pore and fluid mixes completely at each node.

In this contribution, a microscopic description for clogging in depth filtration is presented[4] which leads to phenomenology outside classical percolation and related breakdown processes. Our approach is complementary to other microscopic modeling work which has focused primarily on drawing analogies with conventional percolation.[5–7] We first introduce a quasi-one-dimensional "bubble" model for which basic aspects of depth filtration can be found analytically. We also study depth filtration on the square lattice and discuss the relation to the bubble model description, as well as the connection to direction percolation. Implications for efficient filters are also discussed.

2. Microscopic Model

There are several basic elements in building a microscopic description of depth filtration. These include: (a) the network geometry, (b) fluid and particle transport properties of the network, (c) the trapping mechanism, and (d) particle and bond size distribution characteristics. We discuss each in turn.

2.1. *Network geometry*

We consider filtration both on a square lattice as well as on the idealized "bubble" model. The latter provides an especially convenient way to describe filter evolution during the clogging process. The bubble model consists of L series links, in which each link is a parallel bundle of w bonds and each bond represents a pore (Fig. 1). This model usefully describes the breaking of fibers,[8] extremal voltages in resistor networks,[9] and percolation itself.

2.2. *Fluid and suspended particle flow*

For filtration, we posit that each network bond has a radius r which is drawn from a specified distribution, with volumetric flow rate: (i) proportional to $r^4 \nabla p$, where ∇p is the local pressure gradient in the bond, or (ii) proportional to r^2. The former corresponds to Poiseuille flow in each bond which is driven by an external pressure gradient — the fluid analog of electrical current flow in a resistor network. The latter can be viewed as "dry" flow, in which there is a constant downstream bias in each bond. More generally, we consider the flow rate in a bond of radius r to be proportional to r^μ.

Dynamically neutral suspended particles move through the medium according to the local flow. We assume perfect mixing at each node, in which a suspended particle has a probability proportional to r_i^{μ} to enter an unblocked bond of radius r_i among the "downstream" bonds. Particles are injected singly (at infinite dilution) and tracked until each is trapped or escapes the system. After each trapping event, the new flow field is computed (if necessary) to determine the trajectory of the next suspended particle.

2.3. *Trapping mechanism*

Of the many microscopic interactions that underlie filtration, we consider size exclusion,[4,5,7] where a particle of radius r_{particle} is trapped within the first bond encountered with $r_{\text{bond}} < r_{\text{particle}}$. Upon trapping, the particle is defined to block the bond completely so that there is no further fluid flow in this bond. This size exclusion is the dominant effect in gel permeation in porous media and liquid chromatography. More realistically, trapping of a too-large particle should only partially block a bond. Additionally, van der Waals and other electrostatic interactions lead to the attachment of relatively small particles onto pore walls.

Each trapping event reduces the filter permeability and ultimately a clogging threshold is reached where the permeability vanishes. For appropriate particle and bond size distributions, this clogging is gradient driven, as the fraction of blocked bonds may have a power-law dependence on longitudinal co-ordinates. Due to this gradient, filter clogging is fundamentally different from homogeneous breakdown processes[10] and the filter permeability vanishes near the clogging threshold in an unusual manner.[5,6]

2.4. *Particle and bond characteristics*

We consider distributed radii of both particles and bonds. For bonds typically smaller than particles, particles get trapped almost immediately — this is "cake" filtration. The situation of bonds typically larger than particles is more subtle. If there exist particles which are smaller than the smallest bond, then a steady state is eventually reached for a finite system in which the smallest bonds are blocked and the suspension flows freely through the remaining unblockable bonds. If the lower limits of the bond and bond radius distributions coincide, but particles are typically still smaller than bonds, a percolation-like clogging process arises. Finally, if the particle and bond radius distributions overlap substantially, "gradient" trapping occurs, with power law penetration of the medium before clogging is reached.

For simplicity and concreteness, we consider uniform distributions of particle and bond radii in the ranges $[0, b]$ (with $b \leq 1$) and $[0, 1]$, respectively. More general continuous distributions can also be straightforwardly treated.

3. Bubble Model Clogging

We discuss the spatial distribution of the trapped particles, the properties of "escapee" particles as a function of distance traveled, and the nature of the clogging threshold. The second attribute is of practical interest, since a goal of many filtration processes is to remove particles beyond a specified size from a suspension.

3.1. *Distribution of trapped particles*

To determine the spatial distribution of trapped particles during filtration, notice that the gradient nature of the trapping process implies that the number of blocked bonds in downstream bubbles remains small, even at percolation (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 a flow rate proportional to r_i^μ, where r_i is the radius of the ith bond, the probability that a particle of radius r gets trapped in a bubble of w bonds is

$$P_< = w\int_0^r dr_1 \int_0^1 dr_2 \cdots \int_0^1 dr_w \, \frac{r_1^\mu}{r_1^\mu + r_2^\mu + \cdots + r_w^\mu}\,. \tag{1}$$

This gives the probability that the particle enters a bond of radius $r_1 < r$ among w bonds. Introducing $z_i = r_i/r_1$ and writing the integrand in spherical co-ordinates, the asymptotic large-z behavior is given by

$$P_< \sim w\int_0^r r_1^{w-1} dr_1 \int_0^{1/r_1} z^{w-2-\mu} dz\,. \tag{2}$$

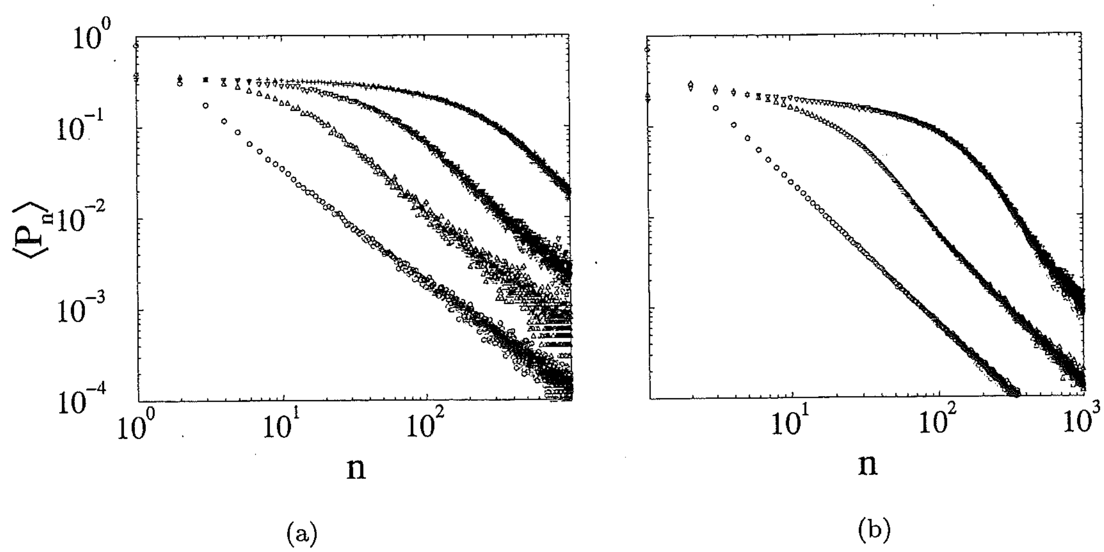

Fig. 2. Trapping probability $\langle P_n \rangle$ versus n at percolation for uniform bond and particle radius distributions $[0, 1]$ and $[0, b]$, respectively, for (a): 10^4 configurations of the bubble model of width $w = 10$ with Poiseuille flow, and (b) 10^3 configurations of a 50×1000 square lattice with "dry" flow ($\mu = 2$). Shown are: (a) $b = 1$ ($\circ$), 0.8 (Δ), 0.7 (∇) and 0.6 (+) (the straight line has slope $-6/5$); (b) $b = 1$ ($\circ$), 0.5 (Δ), and 0.4 (∇) (the straight line has slope $-3/2$).

Evaluating the z integral shows that there is a change in behavior when $w = \mu + 1$ and the final result for the trapping probability is

$$P_< \propto \begin{cases} r^w\,, & w < \mu + 1\,; \\ r^\mu \ln r\,, & w = \mu + 1\,; \\ r^{\mu+1}\,, & w > \mu + 1\,. \end{cases} \tag{3}$$

Thus for small co-ordination number w (appropriate for a lattice), the entrance probability is w dependent, while for large w (appropriate for the bubble model), the entrance probability depends only on μ.

For the bubble model, the last trapping probability in Eq. (3) can also be obtained from the (mean-field) probability of picking a bond whose radius is less than r. This is given by the integral

$$P_< = \frac{\displaystyle\int_0^r x^\mu\, dx}{\displaystyle\int_0^1 x^\mu\, dx} = r^{\mu+1}\,. \tag{4}$$

The probability that this particle gets trapped in the nth bubble is therefore $P_n = P_<(1 - P_<)^{n-1}$, which decays exponentially in n. Averaging over the distribution of particle radii gives

$$\begin{aligned} \langle P_n \rangle &\propto b^{-1} \int_0^b (1 - r^\nu)^{n-1}\, r^\nu dr\,, \\ &\propto b^{-1} n^{-(\nu+1)/\nu} \int_0^{bn^{1/\nu}} y^\nu e^{-y^\nu} dy\,, \end{aligned} \tag{5}$$

where $\nu = \mu + 1$ and $y = rn^{1/\nu}$. When the upper limit of the integral is large, the integral approaches a constant. When the upper limit is small, the exponential factor can be ignored and evaluating the resulting integral leads to the asymptotic behaviors for the trapping probability

$$\langle P_n \rangle \propto \begin{cases} n^{-(\nu+1)/\nu}/b\,, & bn^{1/\nu} > 1\,; \\ b^\nu\,, & bn^{1/\nu} < 1\,. \end{cases} \tag{6}$$

The crossover between these limiting behaviors occurs at a correlation length $n^* \sim b^{-\nu}$, at which point the two asymptotes join smoothly.

3.2. *Escapee size distribution*

The radius distribution of particles which reach downstream distance n is determined by Eq. (5). Here we consider uniform particle and bond radius distributions on $[0, 1]$. Particles which are trapped in bubble $n' > n$ can be viewed as "escapees" for the nth bubble. The probability that a particle of radius r escapes the nth bubble is

$$\sum_{n'>n}^{\infty} P_n = \sum_{n'>n}^{\infty} r^{\nu}(1-r^{\nu})^{n-1},$$
$$\sim \exp(-nr^{\nu}) . \tag{7}$$

Normalizing, this can be recast as the radius distribution of escapees from the nth bubble, Q_n

$$Q_n \propto n^{1/\nu} \exp(-nr^{\nu}) . \tag{8}$$

From this distribution, the average radius of an escapee from bubble n decays as $n^{-1/\nu}$. Thus the escapee size decreases relatively slowly with n: this effect would be even more pronounced if the particle radius distribution were uniform on $[0, b]$. This means that a depth filter with size exclusion as the only trapping mechanism should be relatively long if substantial reduction in the escapee particle size is desired.

3.3. *The percolation threshold*

Percolation means that all bonds in a single bubble are blocked. There are two different behaviors depending on whether the largest particle radius $b = 1$ or $b < 1$.

3.3.1. *The case* $b = 1$

For $b = 1$, clogging is controlled by the spatial gradient in the trapping probability and the probability that all w bonds are blocked in the nth bubble is nonzero only for small n. Within the approximation that it is only the first bubble that clogs, there is a logarithmic anomaly in the w dependence of the number of particles needed to block the system. This ultimately stems from extreme statistics.

To appreciate this feature, let us first compute the number of particles that must be injected into a bond of radius $0 < r < 1$ before it is blocked. This is a basic extreme statistics exercise.[11] For N particles, the probability that all have their radii in the range $[0, r]$ is r^N. This can be re-interpreted as the probability that the maximum radius among N particles lies between 0 and r. Hence $r^N = \int_0^r P_N(r')\,dr'$, with $P_N(r) = Nr^{N-1}$ the probability density that the maximum radius equals r. The average radius of the largest particle in an ensemble of N is $\langle r \rangle_N = \int_0^1 rP_N(r)\,dr = N/(N+1)$. Inverting this relation shows that an order of $(1-r)^{-1}$ particles must be injected before a particle of sufficiently large radius enters to block a bond of radius r.

For a single (the first) 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

$$N_c \approx \int_{r_{\min}}^{r_{\max}} w \frac{dr}{1-r} , \tag{9}$$

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

$$N_c \propto w \ln w\,. \tag{10}$$

The logarithm arises from the widest bonds in the first bubble for which many particles need to be injected before blocking occurs. For a large isotropic system, the percolation threshold therefore is $p_c = 1 - N_c/Lw$, with Lw the total number of bonds in the system. This gives a value p_c which is very close to 1. To obtain a threshold less than unity requires exponential anisotropy in which $L \sim \ln w$.

3.3.2. *The case $b < 1$*

Since the trapping probability is constant (2nd line of Eq. (6)), blockage should equally likely occur in any bubble within the correlation length. Correspondingly, the number of removed bonds at the percolation threshold is proportional to the correlation length times the system width, i.e.,

$$N_c \propto w b^{-\nu}\,. \tag{11}$$

However, for $b < 1$ percolation can occur only if all the bonds in a single bubble have radii less than b. Since this occurs with probability b^w, this means that a single blockable bubble will arise in a system whose length L satisfies $Lb^w \gtrsim 1$. If percolation is to occur, this system length should be less than the correlation length. This gives the pathologically stringent constraint $\nu < w$. This pathology stems from the mean-field transverse character of the bubble model.

4. Square Lattice Clogging

Consider a square lattice whose axes are oriented at 45° with respect to the external bias. Similar to the bubble model, if a particle penetrates to large distance, it effectively encounters an unperturbed bubble model of width $w = 2$ at each node. Using Eq. (3) for a system of co-ordination number 2 and following the derivation of Eq. (5), gives $\langle P_n \rangle$ decaying as $n^{-3/2}$ for the case of coincident particle and bond radius distributions. For the case where $b < 1$, a crossover behavior similar to that in the bubble model is observed (Fig. 2(b)). While this result is most applicable to the first particle injected into the network, later-injected particles appear to exhibit nearly the same spatial distribution of trapping location. Qualitatively similar behavior for $\langle P_n \rangle$ occurs for the square lattice in which there is Poiseuille flow in each bond and in which there is a hydrodynamic bias in the network. The network simulation with Poiseuille flow is a relatively time-consuming simulation since the network permeability must be recalculated after each trapping event.

The location of the clogging (percolation) threshold on the square lattice now reflects features of directed percolation. For coincident uniform particle and bond

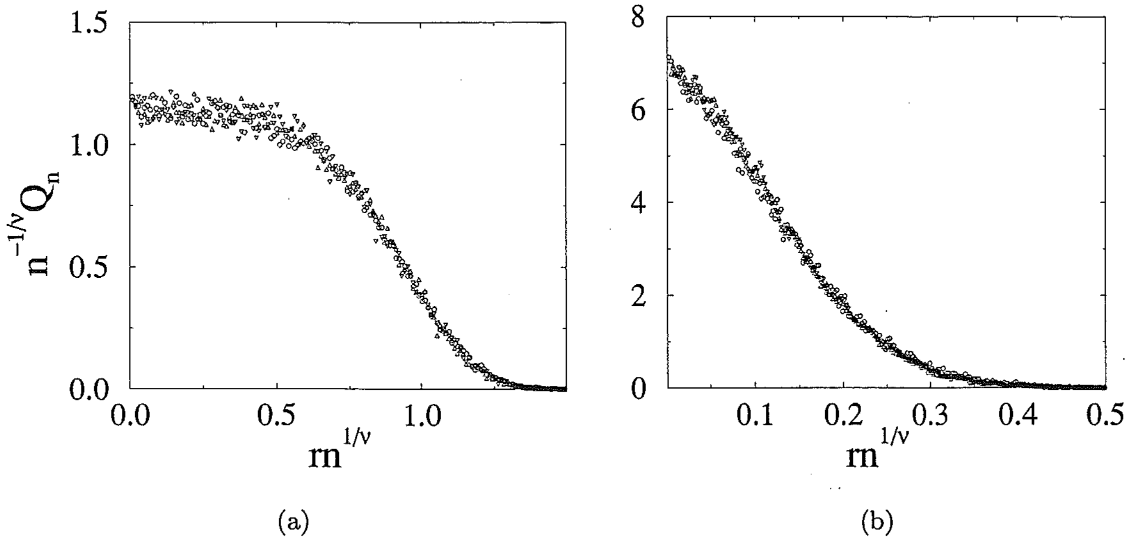

Fig. 3. Escapee size distribution as a function of downstream distance at percolation for uniform bond and particle radius distributions $[0, 1]$ for (a): 10^4 configurations of the bubble model of width $w = 10$ with Poiseuille flow, and (b) 10^3 configurations of a 50×1000 square lattice with "dry" flow ($\mu = 2$). Shown are the scaled distributions $n^{-1/\nu}Q_n$ versus scaled radius $rn^{1/\nu}$ for $n = 50$ ($\circ$), $n = 200$ (Δ), and $n = 800$ (∇).

radius distributions on $[0, 1]$, of the order of $2w$ particles need to be injected to clog the filter. If, however, the particle radius distribution is uniform on $[0, b]$, then there is substantial penetration of particles within the correlation length, just as in the bubble model. As $b \to 0.35$ which corresponds to $1 - p_c$ for directed percolation on the 45° square lattice, clogging is never achieved. This arises simply because a fraction of bonds which is greater than p_c can never be blocked. Finally, the radius distribution of particles which reach a downstream distance n exhibits the same qualitative scaling behavior as in the bubble model, although with a somewhat different shape (Fig. 3(b)).

5. Conclusions and Outlook

In depth filtration a porous medium is gradually clogged by the trapping of particles as a suspension passes through the pore space. The bubble model provides a convenient description for this gradient-controlled dynamical process. For coincident bond and particle radius distributions, the number of particles trapped a distance n downstream varies as $n^{-\lambda}$, where λ depends on the particle and bond radius distributions, but is invariably between 1 and 2. Corresponding to this upstream bias, clogging is achieved when relatively few particles have been injected. For particles smaller than bonds on average, the filter can hold many more particles and is thus substantially penetrated before clogging is reached. These features should have ramifications for efficient filter design. In principle, the slow reduction in the size of escaped particles means that a good filter should be long. On the other hand, in a long filter the downstream end will be relatively inactive. Reconciling these two competing features may best be accomplished by a filter with a longitudinally varying local permeability which effectively cancels the gradient in particle trapping.

There are many interesting directions for further study. Perhaps the most important is to extend the infinite-dilution modeling presented here to the case of a finite-density suspension. This would allow a determination of the time to clogging, as well as quantify the amount of fluid which can be clarified before clogging. It should also prove worthwhile to consider more realistic microscopic particle trapping mechanisms. As mentioned previously, these include the partial blockage of pores when a particle gets trapped, as well as the trapping of fine particles on pore walls by electro-chemical forces.

Acknowledgments

We thank J. Koplik and P. L. Krapivsky for helpful discussions. We also gratefully acknowledge NSF grant DMR-9632059 and ARO grant DAAH04-96-1-0114 for financial support.

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