RAPID COMMUNICATIONS

PHYSICAL REVIEW E VOLUME 53, NUMBER 4 APRIL 1996

Transition in particle capture in deep bed filtration

Claude Ghidaglia
Laboratoire de Physique et Mécanique des Milieux Hétérogènes, URA 857 au CNRS, ESPCI, 10 rue Vauquelin, 75231 Paris CEDEX 05, France

Lucilla de Arcangelis
Laboratoire de Physique et Mécanique des Milieux Hétérogènes, URA 857 au CNRS, ESPCI, 10 rue Vauquelin, 75231 Paris CEDEX 05, France
and Dipartimento di Fisica, Unità dell'Istituto Nazionale per la Fisica della Materia, Università dell'Aquila, via Vetoio, 67010 Coppito, L'Aquila, Italy

John Hinch
Department of Applied Mathematics and Theoretical Physics, University of Cambridge, Silver Street, Cambridge, CB3 9EW, United Kingdom

Élisabeth Guazzelli
Laboratoire de Physique et Mécanique des Milieux Hétérogènes, URA 857 au CNRS, ESPCI, 10 rue Vauquelin, 75231 Paris CEDEX 05, France

(Received 5 December 1995)

Deep bed filtration has been experimentally and numerically studied for small particles flowing through a random packing of larger spheres under laminar flow conditions and in the absence of Brownian motion. The study of the particle penetration depths as a function of the ratio between the diameter of the small mobile particles and the diameter of the large fixed spheres reveals a transition in particle capture which is proposed to be analyzed as a phase transition phenomenon.

PACS number(s): 47.55.Kf, 47.55.Mh

Filtration is the process of separating solid particles in suspension from a carrier fluid by passing the fluid through a filtering medium which retains part or all the injected solids. Despite its long history as a well-known industrial process, with widespread applications in modern industry from waste water treatment to the refining of liquid aluminum, the complex phenomena involved have not been thoroughly understood. Various kinds of filtration exist and can be classified as medium, cake, cross flow, or deep bed filtration. In the case of deep bed filtration, the particles in suspension are smaller than the pores of the filtering medium which they are able to penetrate before being captured at various depths within the bed [1–6]. The transportation and capture of the solid particles in the filtering medium are due to hydrodynamic, gravitational, molecular, Brownian, or electrical forces, acting alone or in combination. The present work focuses on a statistical study of the deep bed filtration of small non-Brownian particles of diameter d in suspension by a random packing of glass spheres of larger diameter D. The objective of this study is to characterize the transition in particle capture by considering the hydrodynamic and gravitational forces and the geometric structure of the packing alone. The problem of the interactions between particles is examined in another study.

The model experiment used to study deep bed filtration is based on a visual observation of small marked particles flowing through a random fixed bed of larger glass spheres, made optically transparent by matching the refractive index of the suspending fluid to that of the glass spheres [7]. The small marked particles were made from acrylic resin with a density 1.19 ± 0.01 g/cm^3. The particles were coated with a fine grain layer of gold using a metal deposition device. The diameters of the particles, d, were chosen to be of the order of several hundred micrometers to facilitate observation and to ensure that electrostatic, Brownian, and molecular forces were negligible compared to hydrodynamic and gravitational effects. For each batch of particles, the distribution of diameters was obtained by computer treatment of an image of the particles projected onto a plane and found to be approximately Gaussian.

Seven batches of small gilt particles with a measured mean diameter and standard deviation (from 240 ± 20 μm to 870 ± 60 μm) were used. The fixed bed consisted of a random packing of glass spheres. Three batches of glass spheres were used, having diameters $D = 4.0$, 5.0 and 6.0 ± 0.1 mm. Partial layers of larger spheres of diameter $D = 10 \pm 0.1$ mm were created at the extremities of the packing in order to induce disorder throughout the bed and to avoid the formation of crystalline zones which would alter the random structure of the medium [8]. Measurements of the porosity along the bed always gave a value of 0.39 ± 0.01 which confirmed the random nature of the packing. The chosen suspending fluid was a mixture of 60% dibutyl phthalate and 40% butyl benzyl phthalate (Santicizer 160 made by Monsanto) with a density $\rho_f = 1.07 \pm 0.15$ g/cm^3, a viscosity $\eta = 29 \pm 1$ cP and a refractive index of 1.520 ± 0.005, equal to that of the glass spheres at 25 °C.

Most of the experiments were carried out in a rectangular cell having an inside width of 125 mm, depth of 39 mm and height of 430 mm. A few preliminary experiments were also

1063-651X/96/53(4)/3028(4)/$10.00

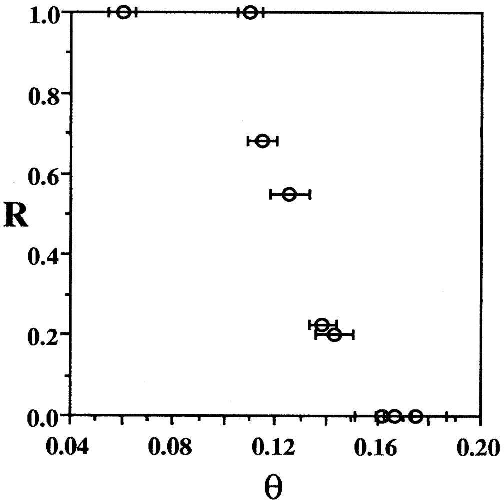

FIG. 1. Experimental results for the ratio R of the number of particles leaving to those entering the bed as a function of the ratio θ of the diameter of the small mobile particles to the diameter of the large fixed spheres.

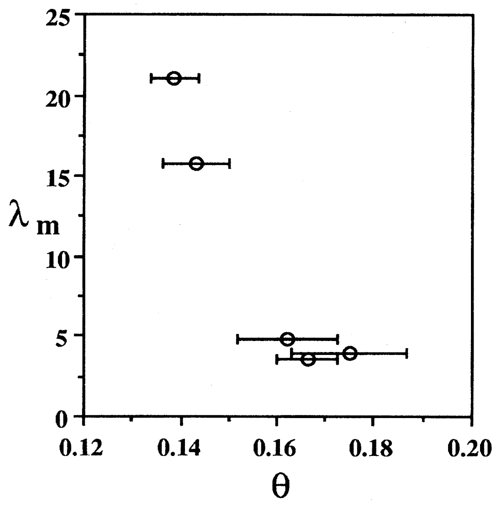

FIG. 2. Experimental results for the median value λ_m of the distribution of the penetration depth as a function of the ratio θ.

performed in another cell having inner dimensions of 100 mm by 39 mm by 550 mm. The fluid was circulated upward through the cell. At the entrance of the cell, the flow was homogenized by using different layers of porous materials. The fixed bed was held fixed by two grids inside the cell. The marked particles were injected one by one with a syringe into a layer of fluid below the bed. Each particle propagated solely in the medium; it was injected as soon as the preceding injected particle was captured or left the filter. Therefore, there was no interparticle interaction in these experiments. The cell was lit from behind to make it easier to track the particle flowing inside the packing. The initial position of the particle as it entered the bed and its final position where it was captured were recorded. From these measurements the distributions of penetration depths and of lateral dispersions were then determined. The ratio of the number of particles leaving to those entering the bed, R, which characterizes the efficiency of the filter, was also measured. A number of 200 particles was injected for each experiment to give good statistics. The experiments were found to be perfectly reproducible. Moreover, although the particles remained in the packing after capture, it was shown that their presence did not affect the penetration depth distribution. Indeed, the number of particles was so small that it did not saturate the packing. The fluid interstitial velocity was kept constant at $U=0.43\pm0.01$ cm/s for all of the experiments. The particle Reynolds number, $dU\rho_f/\eta$, was then always smaller than 0.2 and the Brownian Péclet number was always very large.

Particle trajectories were found to be very vertical and the lateral dispersion to be smaller than 4% of the cell width for all the experiments. This finding shows that particle transport is convective in nature and confirms that steric effects and hydrodynamic and gravitational forces are dominant. By using different combinations of small marked mobile particles and large fixed glass spheres, the dimensionless parameter $\theta=d/D$ was varied from 0.060 ± 0.005 to 0.175 ± 0.010. The ratio R as a function of θ is displayed in Fig. 1. Different experiments having the same value of θ but different values of d and D gave the same results. This observation clearly indicates that θ was the only scaling parameter in the present experimental system. For very large values of θ, the particles were captured in the first layers of the filtering medium. As θ became smaller, the particles penetrated farther inside the medium. For values of $\theta \simeq 0.16$, the penetration depth distribution decayed exponentially into the packing. As θ was further decreased, some particles passed through the packing. For very small values of θ, all the particles flowed through the packing without being captured. These findings reveal a transition in the particle capture with a threshold at θ_c, the precise value of which needs to be evaluated. A characteristic penetration length of the system can be defined by the median value of the penetration depths. The median value, λ_m, is defined as the value for which $\Sigma_{\lambda \leq \lambda_m} n(\lambda)=1/2$ where n is the number (normalized by the total number of injected particles) of particles captured at a depth λ (normalized by the diameter, D, of the spheres of the packing). This length seems to diverge as $\theta \rightarrow \theta_c$ from above as shown in Fig. 2.

These experimental results suggest an analogy with a phase transition phenomena such as percolation [9]. In this framework, the filtration probability can be defined as the probability that a particle has an infinite trajectory. In the case of a finite system such as the present experimental filter, the filtration probability is the ratio R, representing the fraction of injected particles exiting the filter. The curve $R(\theta)$ displayed in Fig. 1 decreases slowly toward zero and presents an inflection point, features which do not exist in an infinite system, where there would be a step function centered at $\theta=\theta_c$. The threshold value was determined as the value of θ where the filtration probability $R=1/2$. The measured value is the effective value 0.130 ± 0.005. An alternative method to compute θ_c is to extrapolate the tangent from the inflection point to $R=0$. This gives the effective value 0.140 ± 0.005. The threshold values estimated with the different methods match within error bars. Although no systematic experimental study of the influence of the size of the filter was performed, this measured value can be considered as close to the infinite system asymptotic value since the filter length is very large $(=60D)$. Another difference from standard percolation studies is the possibility of a spatial correlation of the wider constrictions along faults in some short-range semicrystalline order. The number of experimental data in Fig. 1 and in Fig. 2 was found not sufficient to provide accurate estimates of the critical exponent relative to the behavior of the characteristic length near threshold.

The measured threshold value differs from the geometric value $\theta_g=0.155\pm0.006$ which corresponds to the smallest possible pore formed by three identical spheres in contact [10,11]. Here the error bar comes from the dispersion in the sizes of the particles. Any looseness in the packing would increase θ_g, and thereby increase the difference between the observed experimental and the theoretical geometric threshold values. This finding suggests that the capture sites are not only determined by the pore geometry but also by the hydrodynamic and gravitational forces. Indeed, at low Reynolds numbers, a particle moving along a streamline near a surface can intercept the surface or near a stagnation point it can settle under gravity to a surface. Once in contact with the surface van der Waals forces would capture it. Thus hydrodynamics can lead to capture sites in addition to constrictions. Moreover, an external sharp flow variation can induce a relaunching of a few captured particles. This experimental observation shows the influence of flow variations on the stability of the capture sites and therefore supports the existence of hydrodynamic capture sites. These considerations reveal a deepening complexity of the capture mechanism when hydrodynamic and gravitational forces are taken into account.

In order to model the transition in particle capture, a Monte Carlo numerical simulation was performed. The filtering medium was approximated by a two-dimensional square lattice of cylindrical tubes of a given radius r and length proportional to the radius. The tube radii were chosen according to a power law distribution, $(r-0.155)^{-0.5}$, as suggested by the numerical results of Mason [12] for a random packing of spheres. The flow rate of the viscous fluid in the tubes was assumed to obey Poiseuille's law with perfect mixing at the junctions between the tubes. Gravity also acted on the particles. The mobile small particles had a Gaussian size distribution similar to that found in the experiments. Particles were injected one by one into the lattice, the probability of a particle selecting a tube being proportional to the flow rate in that tube [6]. If a particle came across a pore with a radius smaller than its own radius, it was captured and the entrance to the pore was blocked. Modeling the hydrodynamic (or gravitational) capture was far less obvious since it requires knowledge of the complex flow inside the nodes. The following simplest model was therefore adopted. If a particle reached a junction where the flow rates in the exit tubes were equal within 1%, the particle was captured with probability p_h. Because of lack of information, *ad hoc* probabilities were tested: (a) $p_h=0$ which only takes into account the constriction sites, (b) $p_h=1-\exp(-\theta^2)$ which increases with particle size as suggested by experimental observations, and (c) $p_h=0.5$ which is a random probability and does not depend upon particle size. In the following, we only present data for case (b), since case (a) could not give the capture threshold θ_c less than the geometric value θ_g, and case (c) did not give no capture $R=1$ when θ was much smaller than the threshold. The choice (b) was also motivated by the experimental observation that hydrodynamic capture had an increasing effect as the particle size decreased.

We performed simulations for system sizes $L_h\times L$ ranging from 10×30 to 60×180 (the unit length was chosen to be the characteristic length of the experimental sphere packing, i.e., the diameter of the glass bead), where the particles were injected from the shorter side of the rectangular system as in the experiments. A small number of particles were injected into the same network, about 10% of the total number of entry tubes, then a new configuration was generated and the data were averaged over several configurations (80 000 for $L=30$ and 3000 for $L=180$). For each system size, particles with different θ were injected and the filtration probability, $R=n_{\text{out}}/n_{\text{injected}}$, and the average penetration length were calculated. Of course quantitative comparison between experimental and numerical data is not appropriate since the simulations were performed in two dimensions whereas experiments were in three.

In order to give an accurate estimate of the threshold value θ_c, we applied a method often used in percolation. We evaluated the average penetration length both taking into account the particles exiting the system, and just considering those captured. At the filtration threshold, these two quantities should scale in the same way as function of L, which allowed us to determine θ_c. By doing so, we obtained a value of $\theta_c\simeq0.151\pm0.002$, slightly smaller than the geometric threshold (though these two values can be considered to match within error bars). This result confirms the important role of hydrodynamic capture and suggests that it would be interesting to investigate further the dependence of p_h on the particle size.

To evaluate the critical exponent, ν, of the penetration length we found that the log-log plot as function of $(\theta-\theta_c)$ did not give reliable results. We then chose to collapse the data of the filtration probability R for different system sizes as function of $(\theta-\theta_c)L^{1/\nu}$. For the determined value of θ_c the best collapse was obtained with $\nu=1.82\pm0.07$ (see Fig. 3). This exponent is different from the one for random or directed percolation in two dimensions, a result that could be expected due to the crucial role played by hydrodynamics.

In this work, deep bed filtration was experimentally studied for small particles flowing through a random packing of larger spheres under laminar flow conditions and in the absence of Brownian motion. Steric effects and hydrodynamic and gravity forces were dominant. The study of the particle penetration depths as a function of the ratio, θ, between the diameter of the small mobile particles and the diameter of the large fixed spheres revealed a transition in particle capture which was analyzed as a phase transition phenomenon in analogy with percolation. The ratio θ was shown to be the only scaling parameter in the present experimental system. The measured threshold value of θ was found to be smaller than the geometric threshold value, a plausible explanation being the existence of relaunchable hydrodynamic capture sites in addition to geometric sites. Monte Carlo simulations were developed jointly with the experimental study. Simple ingredients were implemented to reproduce geometric and hydrodynamic capture sites. The numerical threshold was

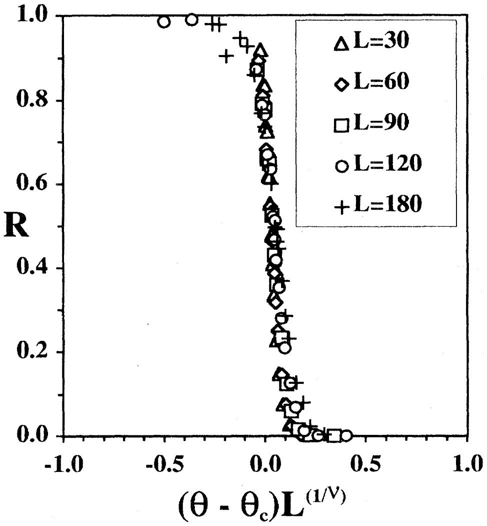

FIG. 3. Numerical data collapse of the filtration probability R as function of $(\theta-\theta_c)L^{1/\nu}$ for different system sizes L. The best collapse is obtained for $\theta_c \sim 0.151$ and $\nu \sim 1.82$.

found to be slightly smaller than the geometrical value θ_g. However the numerical values are close, suggesting that further study of the hydrodynamic capture probability p_h is needed. This will require knowledge of the complex flow inside the pores. Finally, we showed that the critical behavior of the penetration length differed from the well-known results for random and directed percolation.

One further possibility to be investigated, both experimentally and numerically, is the effect of the distribution about the mean value of the diameters of the mobile particles. Preliminary numerical results suggest that, in the case of monodisperse mobile particles, the curve $R(\theta)$ presents a sharp discontinuity at threshold. As soon as a dispersion in mobile particle size is introduced, a continuous and smooth variation of $R(\theta)$ is observed. This might suggest that the smooth nature of the transition is induced by this size dispersion.

Finally, the existence of relaunchable hydrodynamic capture sites is essential in understanding the collective effects which arise when particles are injected by packets [13]. In that latter case, in contrast with the single particle injection procedure which allowed time for the particles in hydrodynamic capture sites to become bonded by van der Waals forces, the local velocity variations induced by a nearby moving particle can produce local relaunching of a particle captured in a hydrodynamic site. These relaunchings explain the deeper penetration of packets of particles inside the filter.

The work of C. Ghidaglia was sponsored by the Centre de Recherches de Voreppe (Pechiney) and by the Centre National de la Recherche Scientifique. It was also undertaken under the auspices of the Groupement de Recherche du CNRS "Physique des Milieux Hétérogènes Complexes," the European network "Cooperative Phenomena in Complex Systems" and a NATO collaborative Research grant on Particle Mixing by Percolation. We wish to thank A. Coniglio, G. M. Homsy, J. Koplik, and S. Roux for discussions. We would also like to thank F. Latrémolière, M. Nicolas, L. Oger, I. Raynal, and S. Tomlin for valuable help and O. Brouard, J.-C. Guibert, M. Mayeux, R. Porcher, and D. Vallet for technical assistance. We also thank F. Lacour for help in coating the acrylic beads. The Santicizer 160 was donated by Monsanto and the Acrilic beads were supplied by du Pont de Nemours and Orkem Norsolor.

[1] C. Tien, *Granular Filtration of Aerosols and Hydrosols* (Butterworths, Boston, 1989).
[2] J. P. Herzig, D. M. Leclerc, and P. LeGoff, Ind. Eng. Chem. **62**, 8 (1970).
[3] C. Tien and A. C. Payatakes, AIChE J. **25**, 737 (1979).
[4] J. Dodds, G. Baluais, and D. Leclerc, in *Disorder and Mixing*, edited by E. Guyon, J.-P. Nadal, and Y. Pomeau (Kluwer Academic Publishers, Dordrecht, 1988), p. 163.
[5] D. Houi, in *Hydrodynamics of Dispersed Media*, edited by J.-P. Hulin, A.-M. Cazabat, and E. Carmona (Elsevier Science Publishers, Dordrecht, 1990), p. 155.
[6] M. Sahimi, G. R. Gavalas, and T. T. Tsotsis, Chem. Eng. Sci. **45**, 1443 (1990).
[7] C. Ghidaglia, E. Guazzelli, and L. Oger, J. Phys. D **24**, 2111 (1991).
[8] D. Bideau, J.-P. Troadec, and L. Oger, C. R. Acad. Sci. Paris **297**, 219 (1983).
[9] D. Stauffer, *Introduction to Percolation Theory* (Taylor and Francis, London, 1990).
[10] J. A. Dodds, J. Colloid Interface Sci. **77**, 317 (1980).
[11] P. Meakin and R. Jullien, J. Phys. (Paris) **51**, 2673 (1990).
[12] G. Mason, J. Colloid Interface Sci. **35**, 279 (1971).
[13] C. Ghidaglia, L. de Arcangelis, E. H. Hinch, and E. Guazzelli, Phys. Fluids **8**, 6 (1996).