

## General Probabilistic Approach to the Filtration Process

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We show experimentally that clogging is basically a matter of the probability of the presence of particles. We describe this process as a function of the main variables of the process, namely, the ratio of particle to mesh hole size, the solid fraction, and the number of grains arriving at each mesh hole during one test, with the help of a simple model, the predictions of which are in very good agreement with our experimental data.

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The flow of suspensions through porous structures, which leads to a partial or full filtration, is widely used in industry either to purify fluids or to separate species in chromatography, microfluidics, or nanofluidics [1]. Filtration has also a major impact in the environment (water and wastewater treatment, drilling well productivity, pollutant storage in soils, unbalance of ecosystems or floods due to accumulation of fine sediments in gravel-bed streams, etc.) [2–4].

At the origin of these processes is clogging, namely, the fact that the suspended particles at some time jam somewhere in the porous medium and never again start moving. Although it may be enhanced by attraction forces between the particles and the solid surfaces, clogging also occurs for noncolloidal particles with a diameter smaller than the pore size, which means that it is essentially a collective effect. Despite its practical importance, only a few studies have focused on the foundations of this phenomenon in specific conditions [5,6], while most other previous approaches were phenomenological [3,4,7]. Here from simple experiments with suspensions of noncolloidal particles in different fluids flowing under gravity through a sieve, we show that clogging is basically a matter of probability; i.e., a sufficient number of particles must be present at the same time at the right place. We describe this process with the help of a simple model, which very well captures our experimental data and might serve as a general basis for modeling filtration in any other situations.

The suspensions were made of glass beads of uniform ( $\pm 3\%$ ) diameter  $d_0$  in the range 0.5–7 mm in either a viscoplastic gel (i.e., roughly speaking, solid for a stress below 30 Pa and liquid otherwise) or a Newtonian glycerol solution (viscosity 0.745 Pa s). The volume fraction ( $\phi$ , the ratio of solid to sample volume) was smaller than 40%. In this range of  $\phi$ , the suspension behavior is governed mainly by the liquid phase rather than by a possible granular structure. Because of the viscosity of our fluids (and the smaller size of the particles used with the glycerol solution), sedimentation was negligible within the duration of our tests. A given volume of suspension ( $\Omega = 0.7$  l, except when explicitly mentioned) was poured in a cylinder (di-

ameter:  $\delta = 10$  cm) closed by a thin plastic film just above a sieve [square mesh with hole size ( $d$ ) in the range 0.63–16 mm and mesh width  $l$ ]. Then the film was withdrawn, leaving the suspension to flow through the sieve under gravity. After the end of the flow, the mass ( $m$ ) of particles remaining in the sieve was weighed, which provided the residue  $R$ , equal to  $m/M$ , in which  $M$  is the mass of particles initially in the suspension. The total number of particles in the suspension was  $N = 6\Omega\phi/\pi d_0^3$ , and the number of mesh holes was  $N_0 = \pi\delta^2/4(d+l)^2$ , from which we deduce the average number of particles arriving at each mesh hole:  $N_e = N/N_0$ . We also carried out similar experiments with dry grains at a solid fraction approximately equal to 60%.

The first striking point of our experiments is that the result of the filtration process is not as simple (binary) as we could expect, i.e., with either no particle at all or an accumulation of all of the particles at the sieve for, respectively, a particle size smaller or larger than the mesh size. We instead get a residue ranging from 0 to 1 as  $\phi$  increases or  $D = d/d_0$  decreases. Moreover, the reproducibility of the data, in particular, when  $R$  is significantly different from 0 and 1, is not very good: The results fluctuate significantly around an average value. For example, the residue measured for 30 trials under the same experimental conditions (mesh hole: 8 mm, particle diameter: 5 mm,  $\phi = 10\%$ ) varied between 0.2 and 0.3. This may seem surprising, since we are dealing with a fluid flow with fixed initial and boundary conditions, which is *a priori* a very reproducible process. However, an exact definition of the initial conditions should involve the precise distribution of the particles in the fluid, and this distribution, in fact, conditions these data fluctuations. The results presented in the following correspond to data averaged over three trials.

Looking at the variations of  $R$  as a function of  $D$  for a given volume of suspension (see Fig. 1), we see that for each  $\phi$  there exists a region of large residues ( $R \approx 1$ ) and a region of small residues ( $R \approx 0$ ) with a transition around a critical ratio  $D_c = (d/d_0)_c$ . Surprisingly,  $D_c$  is significantly larger than 1, which means that the particles may

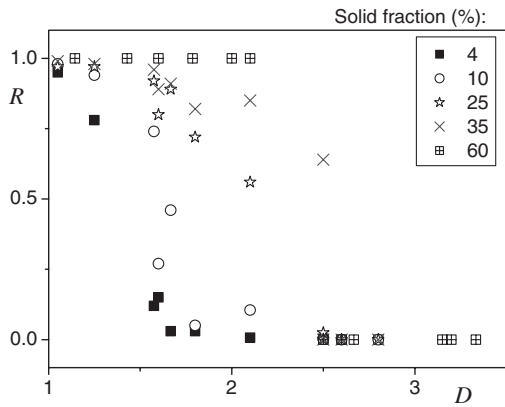


FIG. 1. Residue as a function of the mesh hole to particle diameter ratio for different tests with gel suspensions at different solid fractions (data from Fig. 5) and dry beads at a solid fraction of 60%.

be stopped even when they are significantly smaller than the holes. Such a phenomenon was already observed for flows of granular materials or suspensions through hoppers [8], porous media [9], or microtubes [6] and was shown to result from the formation of particle arches between the solid walls. In our case, the fact that we obtain values for  $R$  intermediate between 0 and 1 means that some particles were able to go through while some others were stopped. This again reflects the probabilistic aspect of the process: The particles dispersed at random in the fluid successively reach the mesh holes, and, for appropriate relative positions of several particles at the approach of one mesh hole, clogging occurs. We can get a direct view of this process by pouring a thin layer of dry grains over the sieve with a special procedure (see the caption of Fig. 2). Although the

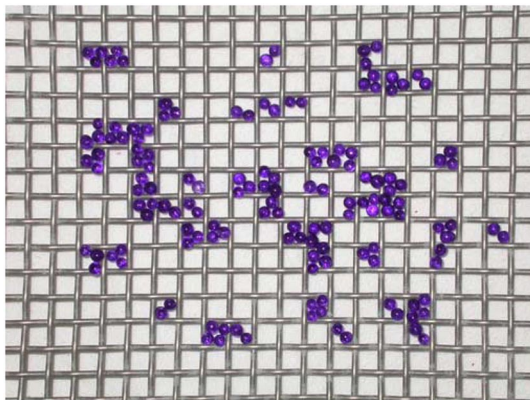


FIG. 2 (color online). View of bridges formed by dry grains ( $d_0 = 3$  mm) over a sieve ( $D = 5$  mm) at the end of a filtration test: The granular sample is initially vibrated so that it goes altogether through the mesh; then, just before the end, the vibrations are stopped so that clogging can occur with the last grain layers. Some particles just lie over a bridge but do not take part in the jamming; they have been stopped by the bridge formed ahead. The average number of particles effectively involved in bridges is  $n = 2.34$ .

conditions of motion of the grains at the approach of each hole are similar, the result looks like a draw with some specific probability: Some holes are empty while others are blocked by a grain bridge (see Fig. 2), and for this sieve there is a wide variety of bridge types involving 2, 3, or 4 particles.

This concept of probability has a critical implication: A clogging event requires that the particles be sufficiently close to each other and, thus, is more probable when the solid fraction is larger, so that, for the same total number of particles arriving at each hole ( $N_e$ ),  $R$  should increase with  $\phi$ . This is effectively what we observe from such a series of tests at different  $\phi$  but with the same  $N_e$  (see Fig. 3). This result, in particular, means that it is possible to improve the efficiency of a filtration process by adjusting the solid fraction or even suppress clogging by a sufficient dilution of the particles. Moreover, the number of clogging events increases with the number of drawings, which implies that, even if the probability of clogging is low, the residue should increase with the sample volume for a given solid fraction. This is effectively what we observed (see Fig. 3).

We can describe this process on the basis of a simple probabilistic model giving the residue as a function of the basic variables of the problem, namely,  $D$ ,  $\phi$ , and  $N_e$ , by taking into account the main effects occurring in the flow. In this context, this model is expected to be applicable to various situations (in particular, other particle and mesh shapes). At any step of this theory, we assume that  $N_0$  and  $N_e$  are sufficiently large so that we can make relevant statistics. Thus, these are the average quantities over a great number of events which are computed. Far from the sieve, the particles rain; i.e., they advance at the same velocity  $V$  (the average velocity of the fluid) in the direction perpendicular to the sieve (we neglect side edge effects leading to a nonuniform velocity profile). At the approach

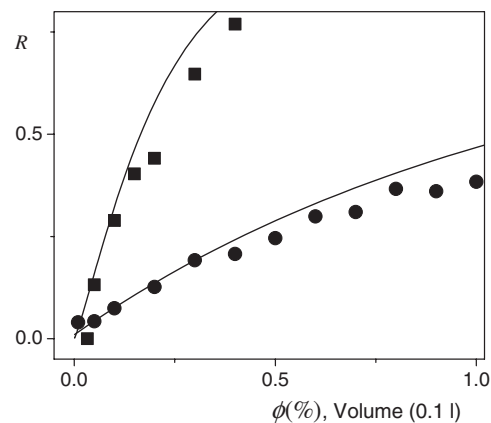


FIG. 3. Residue as a function of the volume fraction (squares) measured for different sample volumes containing a constant mass of particles (the sample volume was 0.7 l at  $\phi = 10\%$ ). Residue as a function of the sample volume (circles) for a given volume fraction (4%) of particles. The particles diameter was 5 mm, and the mesh hole size was 8 mm. The continuous lines correspond to the predictions of the model (see text).

of the sieve, a gradient of velocity develops: Roughly speaking, the fluid regions above the mesh holes flow faster, while those above the solid surface are almost stopped. Various types of particle bridges with various shapes involving different numbers of particles may form. For the sake of simplicity, we consider that the number of grains involved in each bridge is constant and equal to  $n$  (which, in fact, corresponds to the average value). Since the number of grains in a bridge (which has basically a pyramidal structure) is proportional to the surface to cover, all other things being equal, we can write  $n = \gamma D^2$ , in which  $\gamma$  is a factor independent of the particle and mesh sizes (but depending on mesh and particle shapes). From the results of Fig. 2, we deduce  $\gamma \approx 0.85$ .

A bridge forms when  $n$  particles get in contact, thanks to the gradient of velocity at the approach of the mesh. To start with, one particle must touch the sieve, which ensures that the future assembly of particles will effectively lie over a solid region. The probability of such an event is that the particle center be above the solid surface of the sieve:  $u = 1 - [(d - d_0)/(d + l)]^2$ . We assume that, on average, such a particle then moves over a distance  $Kd_0$  with a net vertical velocity  $V_0$ . This velocity is smaller than  $V$ , because of the proximity of the solid surface (either more intense viscous flow or frictional process) and because the particle has to turn around the solid region to fall through the neighboring hole. Thus, the contact between the particle and the solid surface lasts a time  $\theta = Kd_0/V_0$ . In order to jam this particle before it definitively leaves the sieve, another particle has to get in contact with it over this time. The probability for such an event is that to have initially another particle over the distance ( $e$ ) of fluid passing over the (moving) particle during a time  $\theta$  within some vertical conduit above the first particle, i.e.,  $e = (V - V_0)\theta$ . Using the above expression for  $\theta$ , we can rewrite more simply  $e = K(V - V_0)d_0/V_0 = kd_0$ , in which  $k$  is a parameter depending only on flow characteristics. This second particle will then remain in contact with the first one approximately during a time  $\theta$ , and, in order to jam the two other particles and go on building the bridge, there must be a third particle arriving during that time. The probability for such an event is again the probability to have a particle over the distance  $e$  in a conduit of appropriate shape around the second particle. A bridge has approximately a pyramidal shape, so that there are several chains geometries and one or several top particles, but we assume that the above process describes the conditions of occurrence of any contact between two particles of the chain, while the other possible contacts of the pyramid with the sieve are naturally formed by such a construction. Finally, a bridge is formed by one initial contact with the solid surface and  $n - 1$  intergrain contacts occurring in an appropriate timing. The shape of the conduit to take into account depends on the way the hole is covered and, thus, varies for each new contact. We simply assume that its characteristic size (in the plane perpendicular to its axis) is constant and proportional to  $d_0$ .

Let us now compute the probability of having a grain over the distance  $e = kd_0$  in a conduit of any shape but sufficiently thin to avoid any two particles to be situated in a cross section of the conduit axis at the same time. We can start by a discrete approach, considering a system made of empty or full squares of size  $B$  aligned along one direction and with, on average, a concentration  $\psi$  of full squares. The probability to have no grain over a number of  $m$  squares side by side, and thus over a distance  $mB$ , is  $(1 - \psi)^m$ . We can extrapolate this result (independent of the element size) to a more complex distribution in a conduit in which the minimum distance between two successive grains would be  $B = k'd_0$ , in which  $k' \approx 1$  is a function of the exact shape of the conduit. In this frame, the probability to have at least one grain over a distance  $e = kd_0 = \alpha B$  (where  $\alpha = k/k'$ ) is  $P = 1 - (1 - \psi)^\alpha$ . Obviously, the concentration of grains is proportional to the solid fraction of grains in the bulk via a parameter  $\beta$ , which depends on the exact shape of the conduit:  $\psi = \beta\phi$ . Since, in principle,  $P \rightarrow 1$  when  $\phi$  tends to the maximum packing fraction ( $\phi_m \approx 74\%$ ), we find  $\beta \approx 1/\phi_m = 1.35$ . We finally obtain the probability of the formation of a bridge over a particle arriving at the sieve:  $P_0 = u[1 - (1 - \beta\phi)^\alpha]^{n-1}$ .

Now when a mesh hole is jammed, the number of particles reaching each empty hole increases, but the solid surface over which particles may lean also increases. Moreover, some migration effects can occur towards slowly flowing regions. In order to take into account these various effects, we simply consider that all of the particles above jammed mesh holes are now stopped, while all of the particles situated above empty mesh holes are not affected. This approximation is no doubt valid for small  $N_e$ . Thus, the first grain arriving at the sieve jams and stops the  $N_e$  grains behind it with a probability  $P_0$ . The average number of grains stopped by the second grain is then  $P_0(1 - P_0) \times (N_e - 1)$ , and so on with the other  $(N_e - 2)$  grains. Adding all of these numbers, we find the average total number of stopped grains:  $RN_e = \sum_{k=0}^{N_e-1} (1 - P_0)^k P_0 (N_e - k)$ , from which we deduce:  $R = 1 + (1 - P_0/N_e P_0)[(1 - P_0)^{N_e} - 1]$ . Thus, we get an expression of the residue as a function of the main variables  $D$ ,  $\phi$ , and  $N_e$  of the problem with only one unknown parameter, i.e.,  $\alpha$ , related to flow characteristics. Our model effectively predicts the rapid evolution from 0 to 1 as  $D$  goes through a critical value and the fact that, for a given volume of grains (i.e., for a given  $N_e$  value), we can lower the value of  $D_c$  by diluting the suspension (i.e., by decreasing  $\phi$ ) (see Fig. 4). It also predicts that, when  $N_e \rightarrow \infty$ ,  $R$  tends to 1 (see Fig. 5), which means that, in theory, any filtration process should be perfectly efficient after an infinite time of flow.

The predictions of this model are in good agreement with our data with different mesh and particle diameters, different solid fractions, and different sample volumes (see Figs. 3 and 5): There is an accumulation of points in 0 and 1, which means that the theory effectively predicts full filtration or no filtration at all as observed in experiments;

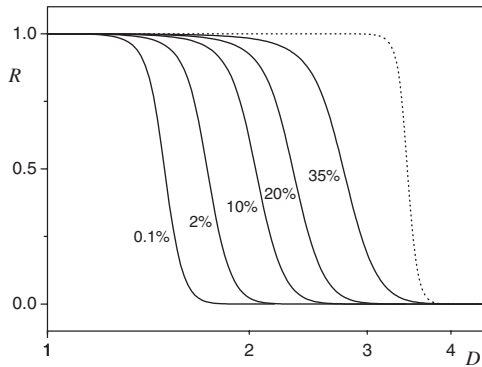


FIG. 4. Residue as a function of the ratio of mesh hole to particle size for different solid fractions, according to the model (see text) with  $\alpha = 0.65$ . The continuous curves correspond to  $N_e = 1000$  and the dotted one to  $N_e = 10^{10}$  and  $\phi = 0.1$ .

the intermediate points (partial stoppage) are in good agreement with experiments, although there is some scattering which seems consistent with the probabilistic character of the process. Remark that the values of the only adjustable parameter of the model,  $\alpha$ , for the two fluids differ, as expected from the fact that it depends on flow characteristics, but we are unable to further explain these values.

We suggest that this theory can be extrapolated to a porous medium by considering it as a series of successive sieves for which  $d$  is now the typical pore size and  $l$  the typical thickness of the solid skeleton [which may, for example, be computed from the porosity  $\varepsilon = (d/d + l)^3$ ]. The suspension successively flows through the sieves, and, for a grain reaching any of these sieves, the probability to be stopped is  $R$ . Under these conditions, the total residue before the  $Z$ th layer is equal to one minus the probability to be unstopped until that point, which is written:  $R^*(Z) = 1 - (1 - R)^Z$ . For small  $R$  values, which is generally the

case in practice (otherwise, most of the particles are stopped at the top of the sample), we have  $R^* \approx ZR$ , where  $Z \approx H/(d + l)$ , in which  $H$  is the length of the porous medium in the flow direction. This means that we can expect a porous medium to fully filtrate a suspension if the sample thickness is sufficiently large. It is particularly interesting to examine the rather general case of very small solid fractions ( $\phi \ll 1$ ). If  $\alpha$  is not too large, we have  $P_0 \approx (\alpha\beta\phi)^{n-1}$ , and, if  $N_e$  is not too large (i.e.,  $N_e P_0 \ll 1$ ), we have  $R \approx P_0$ , which leads to  $R^* \approx Z(\alpha\beta\phi)^{n-1}$ . This approximation is valid only for small values of  $R^*$  but makes it possible to estimate the typical dimensionless length of full filtration of particles in a porous medium:  $Z_c \approx 1/(\alpha\beta\phi)^{n-1}$ .

The possible attractive forces between the particles and the solid surface of the porous system may be accounted for by this theory by simply using an appropriate value for  $\alpha$ , likely larger than those found in this study, since in that case  $V_0$  is much smaller than  $V$ . Note that assuming  $V_0 = 0$  means that any contact is definitive, which implies that clogging is extremely rapid and corresponds to  $\alpha \rightarrow \infty$  in the model.

As it describes the process as a function of the main variables of the problem, our model can *a priori* be used for describing more complex situations and, in particular, for porous materials and/or attractive particles, but its ability to describe such a process needs to be checked in the near future.

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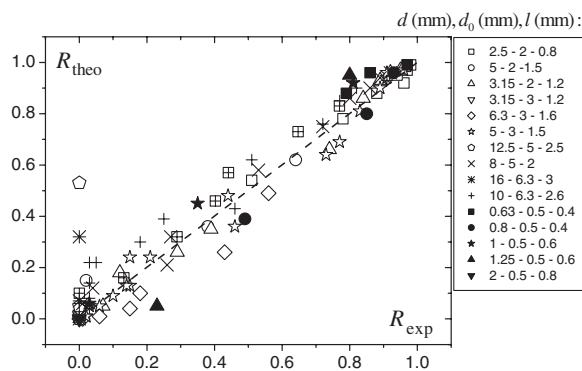


FIG. 5. Theoretical residue as a function of the experimental residue for our different tests: suspensions of particles at different solid fractions in a gel (empty symbols) or a glycerol solution (filled symbols). The coefficient  $\alpha$  in the model is 0.65 for the gel and 2 for the glycerol solution. The cross squares correspond to data in Fig. 3, i.e., the same number of particles (5 mm) through the 8 mm mesh in different gel volumes.

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