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## Particle-Size-Exclusion Clogging Regimes in Porous Media

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From observations of the progressive deposition of noncolloidal particles by geometrical exclusion effects inside a 3D model porous medium, we get a complete dynamic view of particle deposits over a full range of regimes from transport over a long distance to clogging and caking. We show that clogging essentially occurs in the form of an accumulation of elements in pore size clusters, which ultimately constitute regions avoided by the flow. The clusters are dispersed in the medium, and their concentration (number per volume) decreases with the distance from the entrance; caking is associated with the final stage of this effect (for a critical cluster concentration at the entrance). A simple probabilistic model, taking into account the impact of clogging on particle transport, allows us to quantitatively predict all these trends up to a large cluster concentration, based on a single parameter: the clogging probability, which is a function of the confinement ratio. This opens the route towards a unification of the different fields of particle transport, clogging, caking, and filtration.

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A multitude of situations involve the passage or stoppage of elements suspended in a liquid through a porous medium: separation of species in chromatography or microfluidics [1], filtration in biological processes [2], water and wastewater treatment [3], drilling and oil recuperation processes [4], pollutant transport or storage in soils [5], and sediment transport [6]. Depending on the characteristics of the elements or of the porous medium, different situations may be encountered: The elements may be transported without being stopped, be transported at some depth and finally stopped—this is *depth filtration*—or be blocked at the entrance—this is *caking* (outside the filter). Various crucial questions of practical importance emerge: the efficiency of the filter (generally a porous medium), conditions leading to caking, depth reached by stopped elements, induced evolution of flow characteristics and filtration properties, etc. So far, theories or experiments essentially focused separately on caking [7], clogging at the entrance of a filter or a model 2D porous medium [8], deep-bed filtration [9], permeability evolutions [10], or colloid transport [11], and recent studies showed the possibility of observing indirectly [12] or directly [13] particle depositions in depth in porous media. These studies provided insights into the physical mechanisms of these different processes under specific conditions, but there is no unifying physical approach allowing us to deal simply, at first order, with the different regimes (transport, depth filtration, and caking).

Here we focus on the case for which, whatever the different possible effects at work (Brownian motion, colloidal interactions with walls, sedimentation, aggregation between elements, drag force due to flow, etc.), the porous medium

clogs after a more or less long time of flow. In that aim, we use a model system for which clogging can result only from size exclusion (particle jamming in a path smaller than its diameter). We show that clogging occurs as an accumulation of elements in pore size clusters, which ultimately constitute regions avoided by the flow. The clusters are dispersed in the medium, and their concentration (number per volume) decreases with the distance from the entrance, a process ultimately leading to caking. A model involving a single parameter, i.e., the probability to form a cluster, but taking into account the impact of the existence of clogged paths, allows us to predict all the trends observed inside the sample. Systems with additional, more complex interactions between the elements and the wall might be considered within a similar frame, through a probability of blockage (or attachment) now depending also on these effects.

Our experiment consists in making a dilute suspension of noncolloidal particles flow through a model porous medium and then following the clogging dynamics. The particles are monodisperse, spherical, polystyrene beads with a diameter of  $41 \pm 9 \mu\text{m}$ . The carrying fluid is a glycerol-water mixture chosen to match the density of the particles and avoid sedimentation effects (see [14]). The flow is imposed at a constant flow rate, and all our tests are carried out under laminar flow conditions (maximum Reynolds number at the pore scale  $< 1$ ). The porous medium is composed of almost monodisperse spherical glass grains stacked at random close packing in a glass tube of a diameter much larger than that of the grains ( $d_g$ ). Various particle to grain ratios ( $r$ ) are tested in the range [0.054–0.13], which corresponds to the range for which

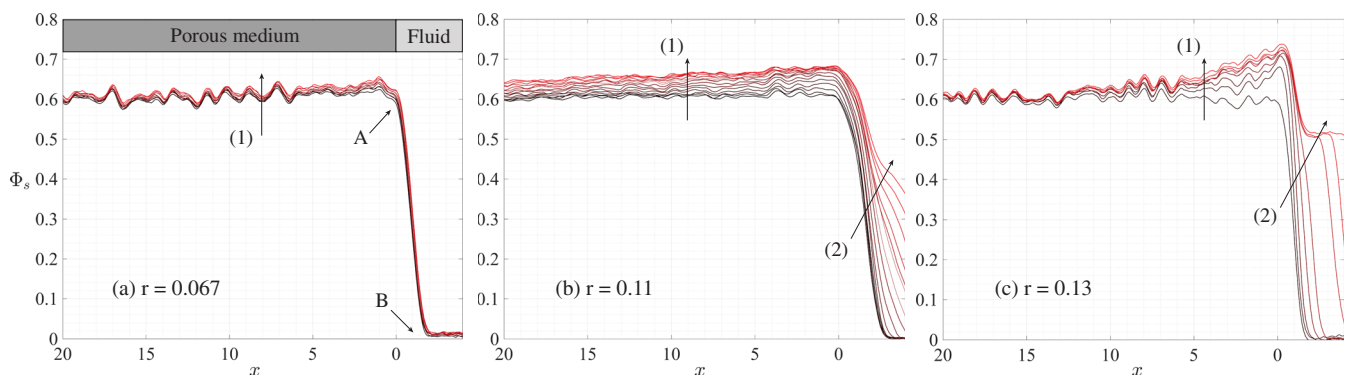
deposits are expected [15]. In the absence of significant physical effects varying with the grain or particle size, filtration properties are expected to depend solely on  $r$ .

In order to measure the distribution of particle concentration in the sample, a benchtop NMR spectrometer is used with an additional gradient field over the vertical axis (i.e., that of the glass tube), which makes it possible to perform proton density unidirectional profiles [14]. The measured NMR signal corresponds only to the protons of the liquid phase, from which we deduce the local solid fraction ( $\Phi_S$ ) in each cross section. NMR measurements are carried out after successive periods of time, corresponding to a constant additional number of injected particles. After each of these periods, we maintain the flow with liquid only, so as to remove unblocked particles, and then a 1D profile imaging starts. The corresponding successive profiles along the depth ( $x$ ) axis (in grain diameter unit) thus provide a clear view of the clogging dynamics.

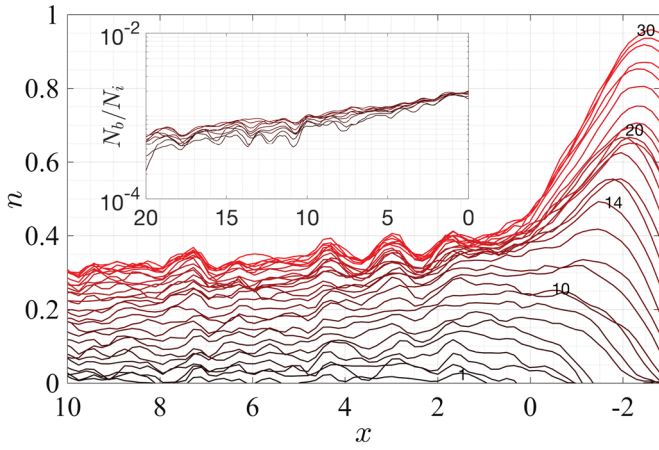
Two mechanisms appear: (1) in-depth deposition, where particles are clogged at various depths in the medium, inducing a local increase of  $\Phi_S$ ; and (2) surface deposition or caking, where particles tend to accumulate at the surface and create a packing of particles on top of the sample. Different clogging regimes are observed when increasing  $r$ . For  $r < 0.05$ , no particle clogged in the porous medium is detected by our technique. For a slightly larger  $r$  value, only a slight in-depth deposition is visible after our series of injections [see Fig. 1(a)]: The height of the profiles increases slowly close to the sample top. For larger  $r$  [e.g., 0.11 in Fig. 1(b)], in-depth deposition becomes significant, and, beyond a critical number of injected particles, surface deposition begins. However, at the same time, for increasing  $r$ , the in-depth deposition tends to be more concentrated close to the top surface, and surface deposition begins earlier, i.e., for a smaller number of injected particles [e.g.,  $r = 0.13$  in Fig. 1(c)]. At the beginning of surface deposition, there may still be some

residual in-depth deposition due to some unevenness of the process, but soon this effect completely disappears [see Fig. 1(c)]: A few-particle-high cake forms, which obviously does not allow the passage of other particles. The solid fraction in this cake is 52%, which is consistent with a loose nonvibrated random packing [16].

For a better appreciation of the physical situation, the deposited solid volume may be represented in terms of the density ( $n$ ) of blocked particles, i.e., the number  $N_b$  of blocked particles in a slice of elementary thickness  $e$  divided by the (maximum) number  $N$  of particles in this slice at a concentration in the pore space equal to that in the cake. Considering the pore structure, we can reasonably take  $e = d_g/2$ . Then we see that, during a first stage, the particles are regularly deposited in depth (see Fig. 2). This process goes on up to a critical density ( $n = 45 \pm 5\%$ ) significantly smaller than the maximum one (i.e.,  $n = 1$ ), for which the particles start to be significantly deposited above the sample surface [see Figs. 1(b), 1(c), and 2]. This cake growth coincides with the end of the in-depth deposition (see Fig. 2), which provides a clear criterion for a cake beginning (no particle can be transported through the particle-packed cake). Furthermore, during the in-depth deposition regime, for each depth,  $n$  increases linearly with time: We get a master curve when rescaling  $N_b$  by the number  $N_i$  of injected particles since the beginning of the test (see the inset in Fig. 2). Thus, during the first injections, in-depth deposition is predominant and the evolution of the particle distribution in depth remains unchanged (it varies linearly with  $N_i$ ). This unexpected result means that the particles go on discovering and populating new sites regardless of the particles already blocked, even if they occupy up to about 50% of the available volume. Then the situation suddenly changes: The in-depth deposition becomes negligible, and caking starts, as if some percolation network formed, precluding a further penetration of particles in the sample.



F1:1 FIG. 1. Successive injections of the same suspended particle number ( $5.0 \times 10^5$ ) for different  $r$  values through bead packing: solid  
 F1:2 volume concentration profile after each injection (solid lines, from bottom to top). The lowest curve (darkest) accounts for the initial  
 F1:3 state, where  $\Phi_S \approx 60\%$  in the medium. The oscillations around this value result from a specific local arrangement of grains, which is  
 F1:4 stable, as proved by the stability of these oscillations during flow. Note that, due to the nonperfect flatness of the free surface, the initial  
 F1:5 profile collapses to zero at the sample top, with some slope (from A to B).



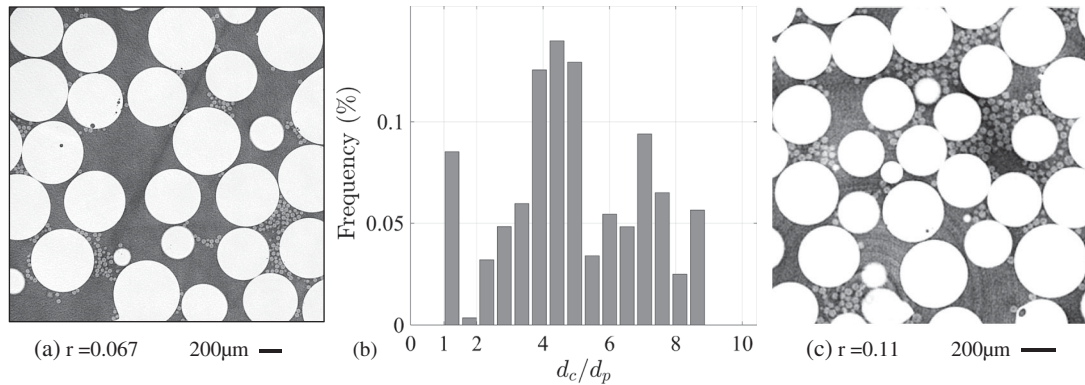
F2:1 FIG. 2. Successive (numbered from bottom to top) injections of  
 F2:2 the same suspended particle number for  $r = 0.11$  density vs  
 F2:3 depth. Note that there is some apparent accumulation of particles  
 F2:4 just above the free surface, which is due to the nonperfect flatness  
 F2:5 of the sample (see Fig. 1). The inset shows the blocked to injected  
 F2:6 particle ratio for injections 1–14.

156 In order to understand these trends, we directly look at  
 157 the distribution of particles at the local scale by x-ray  
 158  $\mu$ tomography, performed after a series of injections (see  
 159 Fig. 3). This shows an original arrangement of the particles:  
 160 They are essentially distributed in clusters, whose apparent  
 161 volume ( $\propto d_c^3$ ) contains from a few tens to a few hundreds of  
 162 particles, plus some isolated particles (i.e.,  $d_c/d_p \approx 1$ ) [see  
 163 Fig. 3(b)]. Surprisingly, this observation is valid even when  
 164 these clusters and the isolated particles occupy a small  
 165 fraction of void in the porous medium [see Fig. 3(a)]. That  
 166 means that during all the filtration process the particles  
 167 are preferentially blocked in dispersed clusters. These  
 168 characteristics of cluster distribution are valid even around  
 169 the critical concentration for a transition to caking [see  
 170 Fig. 3(c)]: The particles are still distributed in large clusters  
 171 separated by void regions, but now the clusters seem  
 172 connected.

173 These observations allow us to deduce key elements  
 174 concerning the process of particle deposition and cluster  
 175 formation. The local width of pathways experienced by  
 176 elements generally ranges from zero to a maximum value  $d$ ,  
 177 so that blockage is a matter of probability: An element of a  
 178 size smaller than  $d$  will be blocked if it is draught by the  
 179 liquid through a sufficiently narrow area. Here, since the  
 180 clusters are essentially independent, have a size of the order  
 181 of the voids between neighboring grains, and include  
 182 almost all the clogged particles, the variations of  $n$  in time  
 183 may be considered to be essentially due to the formation of  
 184 new clusters. They likely nucleate from some initial local  
 185 clogging of one particle and then grow more rapidly than  
 186 our time resolution (i.e., changes between two successive  
 187 injections).

188 Moreover, the first injected particles partially block the  
 189 most probable intergrain sites, but those events likely do  
 190 not significantly affect the flow through the porous  
 191 medium. Other particles can soon follow the same paths  
 192 and arrive ahead of the blocked particles with a probability  
 193 of blocking close to unity. This process goes on until the  
 194 cluster size is sufficient to affect the flow (loss of per-  
 195 meability) at the pore scale, typically by diverting most of  
 196 the flux towards other pores, which also explains that the  
 197 cluster size then stops growing, as it has reached a size of  
 198 the order of the pore size.

199 On this basis, we can build a simple model to describe  
 200 the evolution of the number of blocked particles as a  
 201 function of the depth and the number of injected particles  
 202  $N_i$ . We represent the porous medium as successive identical  
 203 layers of thickness  $e$ , whose value (i.e.,  $d_g/2$ ) appears  
 204 consistent with the process of dispersed clusters filling  
 205 pores. Because of the percolation effect above described,  
 206 the maximum achievable number of particles in a given  
 207 layer during the experiment is not  $N$  but a fraction of this  
 208 number ( $\approx 0.45N$ ). The particles are dispersed homoge-  
 209 nously at random in the flowing liquid and can reach  
 210 various positions in a layer, possibly leading to clogging.



F3:1 FIG. 3. View ( $\mu$ CT image,  $5 \mu\text{m}$  voxels) of the internal structure after particle injection and drying at  $x = 4d_g$  for  $r = 0.067$  (a) and  
 F3:2  $r = 0.11$  (c). White regions correspond to grains, light gray to blocked particles, and dark gray to air. (b) shows an estimation of the size  
 F3:3 distribution of clusters ( $d_c$ ), averaged over depth, for sample (a).



211 On average, this amounts to considering that a particle (in  
 212 fact, a potential cluster) arriving at a free site in some layer  
 213 has a specific probability to be blocked by size exclusion or  
 214 otherwise it progresses to the next layer. Moreover, we  
 215 assume that at each step of the process the flow rearranges  
 216 to avoid filled sites, so that when they arrive at some layer  
 217 the particles attempt to go through the free sites only (i.e.,  
 218 we neglect the flow through the clusters). Note that this is  
 219 consistent with the fundamental assumption of previous  
 220 numerical simulations [10], but our experiments show that  
 221 this description become realistic only if it relies on the  
 222 concept of a cluster containing a sufficient number of  
 223 particles and blocking a large pore. This means that any  
 224 new particle arriving in an already partially clogged layer  
 225 will nevertheless have the same probability to meet a free  
 226 site in this layer, the same probability to get stuck on it,  
 227 finally some constant probability  $p$  to get stuck in the layer,  
 228 and a probability  $1 - p$  to progress to the next layer.  
 229 Another way to think about this constant  $p$  is that, while  
 230 clogging goes on, the number of free sites in a layer  
 231 decreases, but for the same global concentration of injected  
 232 particles the number of particles arriving on the free sites  
 233 increases reciprocally, thus keeping constant the probability  
 234 of a clogging event in the layer. Note that the value of  $p$   
 235 obviously depends on the way  $e$  was defined.

236 As a result, the probability  $f$  for a particle to get stuck in  
 237 the layer  $i + 1$  is written as  $f(i + 1) = p[1 - \sum_i^i f(j)]$ . For  
 238  $x \gg e$ , a continuous version of this equation may be written  
 239 as  $eP(x) = [1 - \int_{0 < u < x} P(u) du]p$ , in which  $P$  is the  
 240 probability density function to get stuck at some position.  
 241 We will assume that the solution of this equation, i.e.,  
 242  $P(x) = (p/e) \exp(-px/e)$ , constitutes a good approxima-  
 243 tion of the clogging process in our case. Note that  $\lambda = e/p$   
 244 is an intrinsic characteristic penetration length, i.e., a  
 245 function of the porous structure and particle to grain size  
 246 ratio. It is worth emphasizing that the final model expres-  
 247 sion is similar to the basic conceptual model proposed for  
 248 colloid transport [17] and for filtration [18], where  $\lambda^{-1}$  was  
 249 named the filter coefficient. However, despite its simple  
 250 final form, our model, built on assumptions on the flow  
 251 behavior derived from direct observations, takes into account  
 252 both pore clogging and flow path evolution. Thus, it appears  
 253 to be valid up to large cluster concentrations for which the  
 254 significant part of the medium is clogged (and not only for a  
 255 negligible particle number).

256 Since this distribution is valid for any injected particle no  
 257 matter the clogging stage of the sample, the density of  
 258 deposited particles in the system simply derives from the  
 259 sum of  $P(x)$  over the number of injected particles  $N_i$ :

$$n = \frac{pN_i}{N} \exp\left(-\frac{px}{e}\right). \quad (1)$$

260 The maximum value  $n_c$  of  $n$  at percolation (i.e., 0.45),  
 263 i.e., that reached  $x = 0$  just before caking starts, is

associated with the maximum number of particles that  
 can be injected in the system, i.e.,  $N_c = n_c N/p$ . Thus, the  
 number of injected particles increases when  $p$  decreases  
 and tends to infinity when  $p \rightarrow 0$ , since the penetration  
 length tends to infinity.

From a 3D representation of this solution [see Fig. 4(a)],  
 we see that, for any  $x$  and any  $p$ ,  $n$  increases linearly with  
 $t = N_i/N_c$ , which is in agreement with regime (1) of our  
 experiments [see Figs. 4(b) and 2, inset]. The final  
 distribution of particles in the sample (for  $t = 1$ ) shows  
 that, for higher  $p$  values, the particles tend to accumulate  
 closer to the surface, as observed in our tests, and the  
 critical injection number is reached sooner. Finally, the  
 model predicts that for a given sample length,  $N_c$  tends to  
 its maximum possible value for  $r \rightarrow 0$ . This means, in  
 agreement with the observed trends, that very low values of  
 $r$  allow the best filling of a sample, in the limit of  
 infinite times.

Let us now compare model predictions with the NMR  
 results. We fit the model to our data by adjusting  $p$  so that,  
 for a given system (fixed  $r$  value), all the experimental  $n$

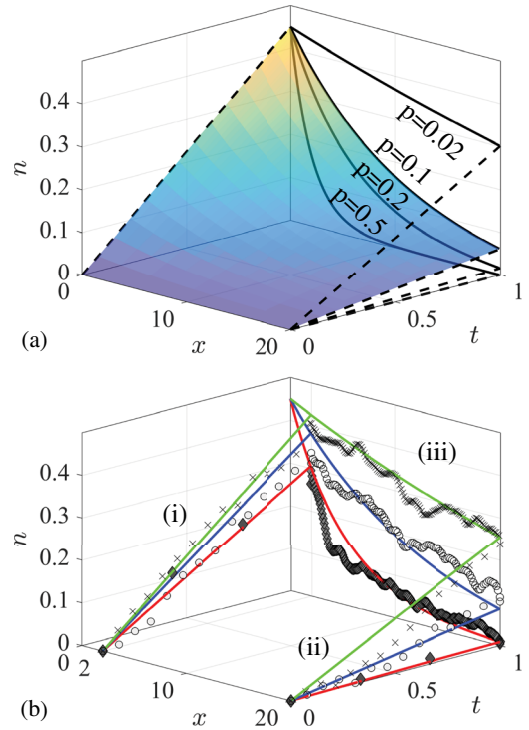


FIG. 4. Density of blocked particles as a function of the depth  
 and injected particles. (a) Model predictions: surface mapping  
 (colored) for  $p = 0.1$ , final distributions before caking for  
 different  $p$  values (continuous lines), and time evolution at  
 two specific depths (dashed lines). (b) Experimental data: time  
 evolution at  $x = 2d_g$  (i) and  $x = 20d_g$  (ii), and final distribution  
 (i.e., at  $t = 1$ ) (iii), for  $r = 0.13$  (filled diamonds),  $r = 0.11$   
 (circles), and  $r = 0.093$  (crosses). The continuous lines corre-  
 spond to model predictions after fitting to all data for a given  $r$

285 profiles in time fall along the theoretical surface mapping.  
 286 A series of profiles at extreme depths and time illustrate  
 287 the agreement of the model with the data [see Fig. 4(b)].  
 288 As expected, the corresponding  $p$  value increases with  $r$   
 289 and tends to 1 (i.e., immediate blocking) for a value  
 290 ( $r_c = 0.153$ ) close to that corresponding to the typical  
 291 maximum void size ( $\approx d_g/6$ ) in granular packings. Finally,  
 292  $p(r)$  may be well represented by the function  $p = \exp(r -$   
 293  $r_c)/\alpha$  (see Fig. S5 in Ref. [14]), with  $\alpha = 0.016$ . Following  
 294 the standard approach of capture by a spherical collector  
 295 [18], we would expect a variation with  $r^2$ . The difference  
 296 with our result is likely due to the flow distribution in these  
 297 different paths, i.e., the fact that the local flow rate through  
 298 small paths is smaller than elsewhere. We can finally note  
 299 that the critical number of particles that can be injected  
 300 essentially varies as  $\exp-(r - r_c)/\alpha$ .

301 The resulting relationship between  $r$  and  $p$  provides a  
 302 means to adapt filter characteristics (pore size and thickness)  
 303 to get expected filtration properties (e.g., distance of depo-  
 304 sition). These results might serve to refine the numerical  
 305 simulation of permeability evolution [10]. For example, they  
 306 suggest that, for a homogeneous porous medium with  
 307 realistic pore size distribution, there always exist, close to  
 308 any large pore, some small pores which can be the source of  
 309 clogging through the formation of a large “cluster” finally  
 310 equal to the large pore. The principles of this model may be  
 311 extended to clogging in porous media under more complex  
 312 conditions, by considering that the different possible addi-  
 313 **2** tional effects essentially affect the clogging probability value.

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