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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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20 A multitude of situations involve the passage or stoppage 21 of elements suspended in a liquid through a porous medium: separation of species in chromatography or microfluidics 22 23 [1], filtration in biological processes [2], water and wastewater treatment [3], drilling and oil recuperation processes 24 [4], pollutant transport or storage in soils [5], and sediment 25 transport [6]. Depending on the characteristics of the 26 elements or of the porous medium, different situations 27 may be encountered: The elements may be transported 28 without being stopped, be transported at some depth and 29 finally stopped-this is *depth filtration*-or be blocked at 30 the entrance-this is *caking* (outside the filter). Various 31 crucial questions of practical importance emerge: the effi-32 ciency of the filter (generally a porous medium), conditions 33 leading to caking, depth reached by stopped elements, 34 induced evolution of flow characteristics and filtration 35 36 properties, etc. So far, theories or experiments essentially 37 focused separately on caking [7], clogging at the entrance of a filter or a model 2D porous medium [8], deep-bed filtration 38 [9], permeability evolutions [10], or colloid transport [11], 39 and recent studies showed the possibility of observing 40 41 indirectly [12] or directly [13] particle depositions in depth in porous media. These studies provided insights into the 42 physical mechanisms of these different processes under 43 specific conditions, but there is no unifying physical 44 approach allowing us to deal simply, at first order, with 45 the different regimes (transport, depth filtration, and caking). 46 Here we focus on the case for which, whatever the different 47 possible effects at work (Brownian motion, colloidal inter-48 actions with walls, sedimentation, aggregation between 49 elements, drag force due to flow, etc.), the porous medium 50

clogs after a more or less long time of flow. In that aim, we use 51 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, 61 allows us to predict all the trends observed inside the sample. 62 Systems with additional, more complex interactions between 63 the elements and the wall might be considered within a 64 similar frame, through a probability of blockage (or attachment) now depending also on these effects. 66

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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 $41 \pm 9 \,\mu$ 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_a) . 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*.

85 In order to measure the distribution of particle concentration in the sample, a benchtop NMR spectrometer is used 86 with an additional gradient field over the vertical axis (i.e., 87 that of the glass tube), which makes it possible to perform 88 proton density unidirectional profiles [14]. The measured 89 90 NMR signal corresponds only to the protons of the liquid phase, from which we deduce the local solid fraction (Φ_s) 91 in each cross section. NMR measurements are carried out 92 after successive periods of time, corresponding to a con-93 stant additional number of injected particles. After each of 94 95 these periods, we maintain the flow with liquid only, so as to remove unblocked particles, and then a 1D profile 96 imaging starts. The corresponding successive profiles along 97 the depth (x) axis (in grain diameter unit) thus provide a 98 99 clear view of the clogging dynamics.

Two mechanisms appear: (1) in-depth deposition, where 100 particles are clogged at various depths in the medium, 101 inducing a local increase of Φ_S ; and (2) surface deposition 102 or caking, where particles tend to accumulate at the surface 103 and create a packing of particles on top of the sample. 104 Different clogging regimes are observed when increasing r. 105 For r < 0.05, no particle clogged in the porous medium is 106 107 detected by our technique. For a slightly larger r value, only a slight in-depth deposition is visible after our series of 108 injections [see Fig. 1(a)]: The height of the profiles 109 increases slowly close to the sample top. For larger r110 [e.g., 0.11 in Fig. 1(b)], in-depth deposition becomes 111 significant, and, beyond a critical number of injected 112 particles, surface deposition begins. However, at the same 113 time, for increasing r, the in-depth deposition tends to be 114 more concentrated close to the top surface, and surface 115 deposition begins earlier, i.e., for a smaller number of 116 injected particles [e.g., r = 0.13 in Fig. 1(c)]. At the 117 beginning of surface deposition, there may still be some 118

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 125 deposited solid volume may be represented in terms of the 126 density (n) of blocked particles, i.e., the number N_h of 127 blocked particles in a slice of elementary thickness e 128 divided by the (maximum) number N of particles in this 129 slice at a concentration in the pore space equal to that in the 130 cake. Considering the pore structure, we can reasonably 131 take $e = d_q/2$. Then we see that, during a first stage, the 132 particles are regularly deposited in depth (see Fig. 2). This 133 process goes on up to a critical density $(n = 45 \pm 5\%)$ 134 significantly smaller than the maximum one (i.e., n = 1), 135 for which the particles start to be significantly deposited 136 above the sample surface [see Figs. 1(b), 1(c), and 2]. This 137 cake growth coincides with the end of the in-depth 138 deposition (see Fig. 2), which provides a clear criterion 139 for a cake beginning (no particle can be transported through 140 the particle-packed cake). Furthermore, during the in-depth 141 deposition regime, for each depth, n increases linearly with 142 time: We get a master curve when rescaling N_b by the 143 number N_i of injected particles since the beginning of the 144 test (see the inset in Fig. 2). Thus, during the first injections, 145 in-depth deposition is predominant and the evolution of the 146 particle distribution in depth remains unchanged (it varies 147 linearly with N_i). This unexpected result means that the 148 particles go on discovering and populating new sites 149 regardless of the particles already blocked, even if they 150 occupy up to about 50% of the available volume. Then the 151 situation suddenly changes: The in-depth deposition 152 becomes negligible, and caking starts, as if some perco-153 lation network formed, precluding a further penetration of 154 particles in the sample. 155



F1:1 FIG. 1. Successive injections of the same suspended particle number (5.0×10^5) 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.

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

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

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

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



F3:1 FIG. 3. View (μ CT image, 5 μ 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 fact, a potential cluster) arriving at a free site in some layer 212 has a specific probability to be blocked by size exclusion or 213 214 otherwise it progresses to the next layer. Moreover, we assume that at each step of the process the flow rearranges 215 to avoid filled sites, so that when they arrive at some layer 216 the particles attempt to go through the free sites only (i.e., 217 we neglect the flow through the clusters). Note that this is 218 219 consistent with the fundamental assumption of previous numerical simulations [10], but our experiments show that 220 this description become realistic only if it relies on the 221 concept of a cluster containing a sufficient number of 222 particles and blocking a large pore. This means that any 223 224 new particle arriving in an already partially clogged layer will nevertheless have the same probability to meet a free 225 site in this layer, the same probability to get stuck on it, 226 finally some constant probability p to get stuck in the layer, 227 228 and a probability 1 - p to progress to the next layer. Another way to think about this constant p is that, while 229 clogging goes on, the number of free sites in a layer 230 decreases, but for the same global concentration of injected 231 particles the number of particles arriving on the free sites 232 increases reciprocally, thus keeping constant the probability 233 of a clogging event in the layer. Note that the value of p234 obviously depends on the way e was defined. 235

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

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

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

The maximum value n_c of n at percolation (i.e., 0.45), 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. 264

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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 282 results. We fit the model to our data by adjusting p so that, 283 for a given system (fixed r value), all the experimental n 284



FIG. 4. Density of blocked particles as a function of the depth F4:1 and injected particles. (a) Model predictions: surface mapping F4:2 (colored) for p = 0.1, final distributions before caking for F4:3 different p values (continuous lines), and time evolution at F4:4 two specific depths (dashed lines). (b) Experimental data: time F4:5 evolution at $x = 2d_a$ (i) and $x = 20d_a$ (ii), and final distribution F4:6 (i.e., at t = 1) (iii), for r = 0.13 (filled diamonds), r = 0.11F4:7 (circles), and r = 0.093 (crosses). The continuous lines corre-F4:8 spond to model predictions after fitting to all data for a given rF4:9 F4:10 (see the text).

285 profiles in time fall along the theoretical surface mapping. A series of profiles at extreme depths and time illustrate 286 the agreement of the model with the data [see Fig. 4(b)]. 287 288 As expected, the corresponding p value increases with r289 and tends to 1 (i.e., immediate blocking) for a value 290 $(r_c = 0.153)$ close to that corresponding to the typical maximum void size ($\approx d_a/6$) in granular packings. Finally, 291 p(r) may be well represented by the function $p = \exp(r - \frac{1}{2})$ 292 $r_c)/\alpha$ (see Fig. S5 in Ref. [14]), with $\alpha = 0.016$. Following 293 294 the standard approach of capture by a spherical collector [18], we would expect a variation with r^2 . The difference 295 with our result is likely due to the flow distribution in these 296 297 different paths, i.e., the fact that the local flow rate through 298 small paths is smaller than elsewhere. We can finally note that the critical number of particles that can be injected 299 essentially varies as $\exp(-(r-r_c)/\alpha)$. 300

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

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