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Self-Limited Accumulation of Colloids in Porous Media

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We present local direct imaging of the progressive adsorption of colloidal particles inside a 3D model porous medium. By varying the interparticle electrostatic interactions, we observe a large range of particle deposition regimes, from a single layer of particles at the surface of the medium to multiple layers and eventually clogging of the system. We derive the complete deposition dynamics and show that colloid accumulation is a self-limited mechanism towards a deposited fraction associated with a balance between the particle interactions and the imposed flow rate. These trends are explained and predicted using a simple probability model considering the particle adsorption energy and the variation of the drag energy with evolving porosity. This constitutes a direct validation of speculated particle transport mechanisms, and a further understanding of accumulation mechanisms.

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Colloidal particles from industrial or natural sources 18 propagate and alter the environment they flow through. 19 Common problems include the accumulation of particles 20 impacting industrial [1,2] or biological [3-6] processes 21 (filtration, storage, cleaning, sorting, etc.) or the leaking of 22 23 contaminants in ground water [7–9]. Predicting particle transport and stoppage in these porous media is key to solve 24 these problems. All stoppage events have either a geomet-25 26 rical (i.e., straining [10], clogging [11], bridging [12]) or a physicochemical origin (i.e., adsorption). Although the 27 elementary processes are well identified, their development 28 in a porous medium is often described by empirical models. 29 Moreover, because of the complexity of real 3D porous 30 31 structures, the dynamics of particle deposition are usually 32 extrapolated from indirect observations like breakthrough 33 curves [13–15], direct observations in simplified systems (2D and/or at pore scale) [16-19], and qualitative static 34 imaging or indirect imaging in realistic systems [20,21]. 35

36 For attractive interactions between suspended particles 37 and porous matrices, several regimes are speculated but unequally understood. First, a particle following a stream-38 line close enough to a surface can be intercepted and 39 40 adsorbed. Subsequent adsorption is then limited by the 41 presence of previously adsorbed particles, and by interparticle repulsion [22]. This leads to the formation of a 42 uniform single layer of adsorbed particles well described 43 by a simple deposition model relying on a blocking 44 function [23,24] and the DLVO theory [25,26]. Then, 45 for weak interparticle repulsion, particles can adsorb to 46 each other and form complex deposits not limited to the 47 surfaces. As deposits can then grow to larger sizes they may 48 induce significant changes to the flow and boundary 49 conditions, which makes the system much more difficult 50

to study. These situations are generally ignored or described assuming no coupling between the deposition mechanisms and the state of the flow [27].

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Here, we present a full description—by direct internal observation in a model system—of the successive states of deposition from the formation of a monolayer of particles to multilayer accumulation and potential clogging. We explore how the deposition dynamics are affected by the coupled evolution of the pore space and hydrodynamics. Finally, we show that the accumulation of particles can be a self-limited phenomenon driven by a competition between the drag force applied on the adsorbing particles and the colloidal interaction energy.

As a model porous medium we use monodisperse borosilicate beads (negatively charged, 63 μ m average diameter) randomly packed (porosity $\phi_0 = 0.38$) in a square glass capillary (width w = 1.0 mm), and we invade this medium with positively charged, fluorescent, monodisperse (diameter $d_p = 1.0 \ \mu m$) latex colloids. The suspensions are made at constant dilute concentration $C_0 =$ 20 mg mL^{-1} in a mix of deionized water and DMSO to match the refractive index of the beads [28]. This allows us to visualize the particles inside the porous medium by confocal microscopy. The opposite charges of the beads and the particles fosters particle adsorption at the surface of the beads. Particle-particle bonding may be promoted by screening the interparticle repulsion (i.e., decreasing the Debye length) through an increase of the ionic strength *I* by addition of salt (sodium iodide) [29].

A test consists of continuously injecting a suspension in the porous medium (initially saturated with liquid) and tracking the particle deposition inside the medium. Except when mentioned, we impose a constant flow rate

 $Q_0 = 5 \ \mu L \min^{-1}$, which implies that the local mean 84 velocity, i.e., $v_0 = Q_0/w^2 \phi_0$, increases when the porosity 85 decreases. The flow is laminar: the Reynolds number 86 at the maximum flow rate used in this study is 87 $\text{Re} = v_0 \rho l / \eta = 1.4 \times 10^{-3} \ll 1$, with $l = 15 \ \mu\text{m}$ the typical 88 pore scale, $\eta = 2.5$ m Pas and $\rho = 1.1$ kg m⁻³ the viscos-89 ity and density of the mix. At some distance from the 90 entrance, the concentration of suspended particles may 91 significantly vary as a function of the history of adsorption 92 all along the porous medium. Here we focus on the 93 processes occurring around the entrance of the medium 94 95 (i.e., over a distance of a few bead diameters), where the impact of adsorption history is expected to be negligible, so 96 that the concentration of suspended particles is considered 97 constant and equal to the injected concentration [29]. 98

Let us first focus on negligible interparticle bonding 99 due to strong repulsive electrostatic interactions (i.e., 100 $I = 10^{-6}$ M). Over time, the beads are progressively 101 covered by particles adsorbed to their surface (see 102 103 Fig. 1, I_1 - t_2). A steady state is reached after 1 h of injection and characterized by a single layer of particles, outlining 104 105 the surface of the beads (see Fig. 1, I_1 - t_3 and t_4). The coverage is uneven, with a fairly uniform, significant 106 107 coverage on the upstream side of the beads but a negligible 108 one on the downstream side. For a single bead in a uniform, laminar, fluid flow (far from the bead), the streamlines are 109 110 symmetrical [30] with regards to a cross section. Extrapolating this situation to the flow around packed 111 beads on average we deduce that a particle following a 112 streamline getting closer from the surface than its own 113 radius will theoretically first intercept the surface of 114



F1:1 FIG. 1. Confocal microscopy images of particles (red) deposited F1:2 in the pore space between glass beads (black) in a $150 \times 150 \ \mu m^2$ F1:3 window at the entrance of the porous media [see Fig. 2(c)], with the F1:4 suspension flowing upwards. 4 conditions of ionic strength are F1:5 presented at 4 times.

the upstream bead face. The collision leads to a surface adsorption event, or a bouncing of the particle to a nearby streamline. This means than the observed preferential adsorption on the elements of surface facing the flow is related to the finite size of the particles.

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We also observe that the trajectories of particles are 120 rather smooth at relatively large distance from the walls but 121 become more fluctuating at the approach of some wall [see 122 Fig. 2(a)]. It is worth emphasizing that such effects do not 123 result from inertia effects (small Re) or from diffusion 124 due to thermal agitation, as the Peclet number (i.e., 125 $Pe = d_p v_0/D$, with $D = 10^{-13} \text{ m}^2 \text{ s}^{-1}$ the diffusion coef-126 ficient of the particles) is over 100. Smooth trajectories 127 would be obtained for a single particle moving through a 128 simple bead packing [29]. This effect therefore likely 129 results from boundary conditions continuously evolving 130 due to other particles in suspension and depositions at the 131 wall, which in particular rapidly changes the apparent bead 132 roughness. A particle will be even more sensitive to these 133 effects as it is closer to the wall, which explains these 134 apparent fluctuations in the trajectories growing as the 135 particles approach the wall. This likely enhances the 136 adsorption of particles at the bead surfaces, as it tempo-137 rarily places them closer to the walls. These fluctuations 138 significantly affect the frequency of attempt of adsorption, 139 which depends in a complex way on the particle concen-140 tration, the flow rate, and the current structure of the 141 deposit. Here, as a critical aspect of our approach, we 142 will consider this frequency as a constant factor for a 143



FIG. 2. Local transport and adsorption mechanisms. All scale F2:1 bars are 10 μ m. (a) Time projection of confocal imaging, with the F2:2 suspension flowing upwards. $I = 10^{-6}$ M. Steady particles F2:3 appear as white dots, while moving particles are represented F2:4 by their trajectories. Selected trajectories: particle 1 along wall, 2 F2:5 close to wall, 3 far from wall. (b) Schematic velocity profile in a F2:6 pore of initial and current radii r_0 and r. Dashed area shows F2:7 positions favorable to adsorption: $[r_c: r - r_p]$. (c) Schematics F2:8 of the three scales used in Figs. 1, 2 and 3(a). (d) Selected area F2:9 under steady state for different flow rates $[Q_0; 8Q_0; 16Q_0]$, at F2:10 $I = 10^{-1}$ M. (e) Cluster formed on a surface at $I = 10^{-1}$ M (top), F2:11 detaching and moving to a more stable position (bottom). Dashed F2:12 lines highlight bead surfaces. F2:13

given system, while, as described below, the probability of
adsorption will significantly depend on the characteristics
and evolution of the system. Such an approach relies on the
same fundamental assumptions as the basic approach of
adsorption [31] or the "Eyring model" for describing the
viscosity of a simple liquid [32].

Then, if a particle encounters a free site, it can adsorb to it 150 with some probability resulting from the surface-particle 151 152 interactions and its motion characteristics. Otherwise, if it encounters an adsorbed colloid, it will be repulsed and will 153 progress further along its streamline. A fundamental obser-154 vation is our experiments is that no particle deposited on the 155 beads detaches later. Thus, neglecting in first approximation 156 the variation of boundary conditions due to progressive 157 particle adsorption, the probability of adsorption depends on 158 the flow rate, and is proportional to the particle concentration 159 and to the fraction of bead surface still available for 160 161 adsorption. Let us call s the surface coverage, i.e., the ratio of the current number of particles adsorbed at the surface S to 162 the maximum possible value under our flow conditions S_0 . 163 Then, the rate of variation of s writes as $\partial s / \partial t = k(1 - s)$. 164 In this expression 1 - s is the available surface coverage, 165 and k is a factor including the frequency of attempt of 166 adsorption and the probability of adsorption for a particle 167 approaching a free wall. This model is a specific case 168 169 of the Langmuir approach initially developed for molecular adsorption, but here without detachment. It solves as 170 $s = 1 - \exp(-kt).$ 171

From the images, we compute the fluorescence F over 172 time and over a large number of beads, i.e., the whole 173 capillary entrance [see Fig. 2(c) and [29]]. With regards to 174 a saturated surface deposition S_0 , for $I = 10^{-6}$ M, the 175 deposition F/S_0 increases gradually before reaching a 176 177 plateau value around 1 [see Fig. 3(a)] associated with a single layer of particles (see Fig. 1. I_1 - t_4). This dynamics is 178 well described by the above model for s(t) [see Fig. 3(a)] 179 with $k = 5.0 \times 10^{-4} \text{ s}^{-1}$, which confirms our understand-180 ing of the whole process of monolayer deposition. 181

Let us now see how the process evolves when interparticle 182 bonding is allowed, by increasing the ionic strength. For a 183 relatively low ionic strength (i.e., $I = 10^{-4}$ M) we observe a 184 change in the distribution of the deposits (see Fig. 1, I_2 - t_3): 185 the process again starts by particle adsorption on the bead 186 surface but it is soon followed by particle adsorption to other 187 particles, which leads to the formation of deposits thicker 188 than one particle size. 189

Under these conditions, since the particles keep the same 190 properties all along the process, we would expect a 191 continuous adsorption until filling the whole pore space 192 193 and thus clogging of the system. On the contrary, we observe (see Fig. 1, I_2 - t_4) that the deposition progressively 194 slows down and seems to tend to a maximum, significantly 195 before saturation of the whole pore space. More precisely, 196 197 the deposited amount increases at a decreasing rate and finally tends to a plateau at long times [see Fig. 3(a)]. Thus, 198



FIG. 3. (a) Overall deposition F over time rescaled by surface F3:1 saturation S_0 , at the entrance of the porous media for increasing F3:2 ionic strengths (bottom to top). Plain lines correspond to data. F3:3 Values above the horizontal line are not quantitative due to F3:4 resolution limitations (see Ref. [29]). Dashed lines show the fitted F3:5 model: s(t) for $I = 10^{-6}$ M, with $k = 5.0 \times 10^{-4} \text{ s}^{-1}$; $s(t) + \beta b(t)$ F3:6 for larger values of I, with $\tau = 3.0 \times 10^4$ s. (b) Calculated and F3:7 measured values for the initial bulk deposition probability p_0 F3:8 (dotted line, squares) and the bulk deposition at saturation b_{sat} F3:9 (dashed line, diamonds), as a function of the ionic strength. The F3:10 vertical line shows the limit of application of the model (i.e., F3:11 suspension instability). F3:12

colloid deposition appears limited by some self-equilibrium process.

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This implies that the flow evolution plays a major role: as201the deposit thickness increases, the porosity decreases, and202thus the local velocity increases (at constant flow rate); the203drag force on the particles then increases, which decreases204the probability of adsorption. Beyond some critical velocity205the drag force is so large that no adsorption is possible.206

This interpretation is confirmed by additional observa-207 tions. By stepping the flow rate from Q_0 to $16Q_0$, we obtain 208 successive steady stationary deposits [see Fig. 2(d)]. The 209 particles are washed from the large velocity regions and 210 remain stable in the low velocity regions (i.e., around 211 contact points), which provides a straightforward demon-212 stration of the impact of velocity on the deposit thickness at 213 equilibrium. At this point, it is also interesting to note that 214 no detachment of particle is observed as long as deposition 215 is far from its saturation value. This means that the velocity 216 field variations do not affect the cohesion of individual 217 particles. As we approach the stationary conditions though, 218 during injection at a constant flow rate, some clusters of 219 particles can suddenly detach and move slightly further in 220 the medium [see Fig. 2(e)]. This illustrates that the local 221 drag conditions, which initially allowed the formation of 222 the deposit, have then evolved. The detachment of large 223 clusters instead of individual particles likely results from 224 the increase of the drag force on cohesive obstacles 225 inducing a larger torque on the cluster. 226

Finally, with further screening of the repulsion (i.e., 227 increasing *I*), the adsorption rate is larger, and the plateau 228

229 of deposition increases [Fig. 3(a)]. We conclude that the particle deposition is essentially governed by some 230 equilibrium between the flow conditions and the colloidal 231 232 interactions. Such a qualitative conclusion matches standard theoretical analysis in that field [33], but we here have 233 access to a detailed description of the process, from the 234 local to the average level, which emphasizes the main 235 trends of colloidal deposition (plateaux) and provides 236 237 quantitative data for the local rate of adsorption in time. 238 Note that with a strong screening of the interparticle repulsion ($I = 2 \times 10^{-0}$ M), heavy bulk deposition occurs 239 at early times (Fig. 1, I_4 - t_2). Even though this coincides 240 with the limitation of our observation method due to 241 massive diffraction (see Fig. 1, I_4 - t_3 and [29]), particles 242 likely occupy a large fraction of the pore space. In that case, 243 no self-equilibrium prevents the clogging of the system. 244

We can describe this process through a simple model. As 245 246 a particle approaches a previously deposited particle, it interacts with it through Van der Waals forces and electro-247 static forces. This interaction is well represented by a 248 potential well $\Delta \Phi$ increasing with ionic strength, as the 249 screening of the electrostatic repulsion increases (see 250 Ref. [29] for a complete description of $\Delta \Phi$, function of 251 the Hamaker constant A_H , and the particle surface potential 252 253 Φ_0). We can thus expect that while it gets sufficiently close to an already adsorbed particle, the former will tend to fall 254 in this potential well. A particle will then remain attached if 255 the drag force exerted by the liquid flow around it is 256 sufficiently small. In other words, in order to remove this 257 particle from its potential well, the liquid velocity around 258 the particle must be such that the work $W_d = 6\pi d_p^2 \eta v$ due 259 to drag force during the particle displacement out of the 260 well (thus over a typical distance of the order of the particle 261 262 size), is larger than the depth of the well, i.e., if $W_d > \Delta \Phi$. The balance of these energies defines a critical fluid 263 velocity v_c . For a flow through any geometry, the fluid 264 265 velocity increases with the distance from the walls, which is confirmed by our observations (considering that, in general, 266 moving particles have the same velocity as the fluid) [see 267 Fig. 2(a)]. As a consequence, we can define a critical 268 position r_c at which $v(r_c) = v_c$, such that only the particles 269 situated at a distance greater than r_c from the pore axis may 270 271 adsorb [see Fig. 2(b)]. On the other side, particles are 272 sterically constrained at a distance (from particle center to wall) larger than one particle radius r_p . Assuming 273 274 homogeneous particle dispersion in the fluid, the interval $[r_c; r - r_p]$ compared to the full pore volume defines the 275 fraction of particles in a place suitable for adsorption, thus 276 277 proportional to a probability of adsorption p. For a given flow rate the exact critical values and adsorption probability 278 depend on the detailed boundary conditions, i.e., the porous 279 medium structure, which changes with the volume of 280 deposited particles. On average (over a larger number of 281 282 pores), we thus expect a probability p depending on the 283 fraction of pores occupied by deposited particles, i.e., b.

More precisely, b is the ratio of the number of particles 284 adsorbed to other particles B to the maximum number of 285 particles that fit in the pore space B_0 , excluding the 286 maximum number of particles in the surface layer S_0 . 287 Note that the ratio $\beta = B_0/S_0$ is a bulk to surface filling 288 capacity ratio. Under these conditions the adsorption rate 289 expresses as $\partial b/\partial t = p(b)$. Consistently with our obser-290 vations this probability will increase with the ionic strength. 291 and decrease when increasing the flow rate (since in 292 laminar conditions, the local velocity increases in the same 293 proportion) or, equivalently, when decreasing the porosity. 294 We can also define b_{sat} as the maximum value reached by b295 under given conditions. 296

For example, for a cylindrical pore throat of radius 297 initially equal to r_0 but reduced to r by deposited particles 298 along its wall, the adsorption probability writes p(r) =299 $1 - r_c^2 / (r - r_p)^2$ [see Fig. 2(b) and Ref. [29]]. In that case 300 we have $b = 1 - r^2/r_0^2$, from which we deduce p(b). 301 Remarkably, the simple expression $p_0(1-b/b_{sat})$ is a 302 good approximation of p(b) for all pore sizes r greater 303 than r_p , i.e., as long as the pore is not fully clogged (see 304 Ref. [29] for details and approximated forms of p_0 and 305 b_{sat}). Looking at the particle deposit as a whole, as a first 306 approximation, it means that the process could still globally 307 be considered as a simple deposition process (see above) 308 with regards to a saturation value, i.e., b_{sat} , depending on *I*. 309

Extrapolating this result to the more complex pore 310 structure in a bead packing suggests writing the adsorption 311 rate as $\partial b/\partial t = k's(1 - b/b_{sat})$, where the factor s appears 312 as the existence of an initial surface layer is required for 313 bulk adsorption. Within our framework, k' is a factor 314 including (i) the frequency of attempt of adsorption and 315 (ii) the probability of adsorption of a particle approaching a 316 deposited particle. Therefore, we can write $k' = p_0/\tau$ with 317 τ a characteristic time of adsorption attempt, considered as 318 constant here, while p_0 strongly depends on the ionic 319 strength conditions. 320

From the above general expression for the bulk adsorption 321 rate $\partial b/\partial t$, and the expression for the surface deposition s, 322 we can analytically solve the bulk deposition dynamic **1** 323 as $(b/b_{sat}) = 1 - \exp\{(k'/k)(1/b_{sat})[1 - \exp(-kt) - kt]\}.$ 324 Besides, after rescaling by the surface layer fluorescence, 325 the total deposition $F = S + B = sS_0 + bB_0$ writes $F/S_0 =$ 326 $s + \beta b$. The predicted trends for F/S_0 agree well with all our 327 observations [see Fig. 3(a)]: initial deposition limited by the 328 formation of a surface layer $(F/S_0 = 1)$, and further 329 deposition (if $k' \neq 0$) decaying to the asymptotic value 330 $1 + \beta b_{sat}$. This is valid up to high ionic strength values 331 $(I \sim 10^0 \text{ M})$, above which the particles may easily form 332 large loose clusters, which enhances the clogging proba-333 bility. To compare our expression for F/S_0 to the exper-334 imental dynamics, β is fixed at 6.7. Comparison for 335 $I \in [10^{-6}; 2 \times 10^{-1}]$ M, by fitting k' (therefore p_0) and 336 $b_{\rm sat}$, shows modeled dynamics in good agreement with 337 the experimental values [see Fig. 3(a)]. Independently, from 338 the model, the values of p_0 and b_{sat} are calculated by slight adjustment of A_H and Φ_0 around their theoretically predicted values [29]. The single value $\tau = 3.0 \times 10^4$ s then allows us to match experimental and calculated p_0 and b_{sat} over 5 decades of ionic strength [see Fig. 3(b)]. We therefore validate our ability to predict the impact of the ionic strength on the bulk deposition probability and saturation.

Overall, the competition between the interparticle inter-346 347 actions and the evolutive hydrodynamics allows us to completely describe the deposition state and rate. The 348 resulting full model can be used to compute the rate of 349 deposition along all regimes, from early single layer surface 350 351 adsorption to limited multilayers or clogging. The additional approximated model can be used to quickly estimate 352 the existence of a critical saturated state of deposition. Such 353 tools can significantly change the accuracy of contaminant 354 transport predictions. More generally, the mechanism of 355 356 controlled limited deposition can pave the way to new filtration, cleaning, or sorting systems, with controlled 357 adaptive pore size distributions. As an important next step, 358 confronting these local mechanisms to deeper sections 359 360 2 (same dataset) will help answer the fundamental question of depth propagation. 361

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