

Unraveling hydrogel resistance to particle deposition: Adhesion versus elastohydrodynamic lift

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Soft hydrogels are widely used as antifouling coatings because they strongly suppress deposition of particles, cells, and microorganisms from flowing aqueous suspensions, yet the physical origin of this resistance remains unclear, as several near-wall phenomena occur simultaneously over rapidly changing length scales. Here, we combine deposition kinetics, single-particle trajectory tracking, and independent measurements of adhesion dynamics to identify the key kinetic barrier. Using polystyrene microbeads depositing onto a submicron poly(ethylene glycol) hydrogel coating in a parallel-plate flow cell, we examine how the near-wall deceleration of depositing particles during the last few seconds preceding arrest compares with the one expected for particles rolling in contact with the surface and decelerating through adhesive force maturing over time. Estimating the latter using a braking model, derived from the adhesion dynamics extracted from AFM force spectroscopy using identical colloidal probes and surface, predicts immobilization of several orders of magnitude faster than observed. When further combined with attachment rate constants deduced from the variation of the deposition rate along the channel and their ionic-strength dependence, this disagreement rules out rolling-in-contact scenario. This implies that most particles stay hovering above the surface, separated from the hydrogel by a thin fluid gap maintained by a repulsive force. We further rule out as such force both the electric double-layer interactions, attractive in the present system, and the inertial lift, as it is negligible at the present shear rates. This points to elastohydrodynamic lift (EHL)—arising from pressure-deformation coupling in lubrication flow amplified by large compliance of the hydrogel—as the only plausible barrier force. Using mechanical parameters measured by AFM and established EHL scaling, we show that EHL can indeed impose a sufficient kinetic barrier at a few tens of nanometers from the surface, cutting off attractive adhesion forces and suppressing deposition.

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I. INTRODUCTION

Aquatic surfaces are threatened by fouling with nonliving particles, colloids, salts, and organics [1,2] as well as microorganisms such as bacteria and algae [3,4]. Such deposits compromise pipes [5], membranes [6,7], porous media [8–11], and marine constructions [12]. Deposition and fouling depend on surface properties and near-surface hydrodynamics [13–15], yet an in-depth understanding that would allow rational surface design is still lacking [2,12,14]. In this respect, soft hydrogels are known to behave as antifouling surfaces, but the exact mechanism is unclear and could be related to their distinctive mechanics [16–18], hydration-mediated weak adhesion [19,20], and minimal transport resistance [21,22].

The deposition process involves two coupled phenomena: (1) transport from the bulk to the near-wall region [14,23,24]

and (2) transition from a suspended state to an attached one in the near-wall region [13–15,25]. Though the deposition kinetics is commonly represented by its overall rate [2,14,26,27], it involves many physical processes with distinct rates, which are differently affected by the shear rate [15,28,29], particle size and density [26,30–34], and surface material characteristics [12,19,35]. The mechanisms involved in the bulk transport such as convection [23,24], Brownian diffusion [14,28,31], gravitational settling [32,33], and inertial lift-driven migration (at sufficiently high shear rates) [36] are relatively well understood. The resulting bulk transport resistance may then be reasonably modeled. It is often described in terms of a diffusion boundary layer (BL) [14,23,24], whose thickness for micron-sized objects is on the order of 10 μm , still much beyond the near-wall region, a few tens of nanometers thick, where forces directly responsible for attachment take over [14,23,24]. The latter includes electrostatic and van der Waals interactions (DLVO forces) [13,25], possibly modified by various non-DLVO contributions (hydration, steric, and polymer-mediated forces [19,25,37]) at still smaller distances. For “nonperfect sink” surfaces, particles are captured with a certain probability [13–15,25] and the near-wall concentration becomes finite [2,33], thus particles spend some time in the near-wall region [15,28,29] before they come in direct

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mechanical contact with the substrate. Ultimately, they may adhere irreversibly or dwell on the surface and detach after some time under sufficient shear [29,38,39]. Unfortunately, exact interplay of different phenomena and the dominant rate-controlling mechanism still remains elusive.

The complexity further increases on hydrogels and other soft hydrated surfaces [16–19,35]. The antifouling mechanism of hydrogel surfaces has been attributed to adhesion weakened by distinct mechanics (softness and viscoelasticity) of hydrogels [16–18], high hydration [19,37], or altered rates of transport toward the surface or capture [21,22]. However, much ambiguity remains, as the antifouling behavior may imply various effects such as, for instance, reduced probability of attachment [25,37], slower adhesive bond maturation [20], altered near-contact hydrodynamics [40–43], varied postcontact behavior (rolling, slip, detachment) [29,38,39], or just fewer particles reaching conditions favorable for capture [21,22]. Discriminating possible scenarios and identifying the dominant mechanism on hydrogels is the focus of the present study.

The approach presented in this work addresses this problem in a manner of *reductio ad absurdum*, by systematically hypothesizing and ruling out candidate mechanisms and scenarios based on several complementary measurements. To discriminate scenarios in a sound manner, we focus on near-wall *particle deceleration preceding arrest*, which appears to strongly vary across possible mechanisms. Specifically, we consider diffusion, convection, and migration driven by gravity [28,32,33], precontact adhesion—mainly, electric-double layer Derjaguin–Landau–Verwey–Overbeek (DLVO)—forces [13,25], and slowdown of postcontact particle rolling on the surface. The latter is an important special case, for which we develop here an analytical approach based on parameters measured using AFM force spectroscopy and an adhesive braking model [20,29,38,39]. Notably, particle rolling is well established as postcontact mode of motion for larger living cells, e.g., various blood platelets, adhering to substrates through specific biological interactions [44,45], but is rarely considered in the context of deposition of microparticles [46]. Having ruled out these mechanisms, we ultimately conclude that deceleration prior to arrest is controlled by yet another long-range repulsive force that plays a key role in preventing deposition and generating antifouling effect. We hypothesize that this force is the elasto-hydrodynamic lift (EHL), arising from nonlinear coupling between lubrication pressure in the gap under the particle and elastic deformation of the substrate, enhanced by the hydrogel’s softness [41,42,47]. We reinforce this hypothesis by verifying that, for the present system, EHL magnitude and range fall in the relevant range using experimentally measured parameters and established model relations [41–43,47].

II. MATERIALS AND METHODS

A. Trajectory tracking and deposition experiments

To probe the near-wall approach dynamics and quantify the deceleration immediately preceding arrest, we performed particle deposition experiments in a parallel-plate flow cell (PPFC) that yielded two complementary datasets: (1) trajectories of individual particles prior to immobilization, which yielded variation of their velocity with time [Fig. 1(a)], and

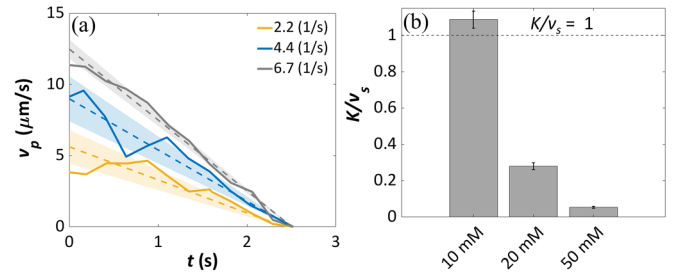


FIG. 1. (a) Variation of particle velocity v_p for last 2.5 s before arrest in PPFC at indicated wall shear rates in 10 mM NaCl for wall shear rates $\sigma = 2.2, 4.4$, and 6.7 s^{-1} . Each trace is the average of four particles. The dashed lines and colored sectors represent average slopes and its 90% confidence level interval, respectively. (b) Attachment rate constants K normalized to the sedimentation velocity v_s for different ionic strengths of NaCl solutions [33]. Displayed values represent average model fits of K to four separate deposition experiments for each ionic strength. Error bars are 90% confidence intervals. Experiments used $1 \mu\text{m}$ polystyrene beads and 250 nm thick PEG hydrogel coating synthesized from amine/epoxy star-PEG precursors and anchored to a glass slide.

(2) attachment rate constants K extracted from overall deposition kinetics [Fig. 1(b)] [33]. Experiments employed $1 \mu\text{m}$ polystyrene particles suspended in 10 mM NaCl aqueous solution. The suspension was fed as a laminar flow by a low-speed digital peristaltic pump (Ismatec IPC-N ISM935C) at several shear rates to a 300- μm -thick, 0.5-cm-wide, and 5.3-cm-long PPFC whose bottom wall was a hydrogel-coated glass slide [Fig. 1(a)].

For the trajectory-tracking measurements [dataset (1)], particles in the near-surface region of thickness defined by the microscope’s depth of focus $\sim 5 \mu\text{m}$ were monitored by phase-contrast microscopy at 20 frames/s and then counted and tracked from contrast-enhanced, median-filtered images using normalized cross-correlation with a 30×30 -pixel template within 60×60 -pixel search windows [48,49]. After calibration, the instantaneous lateral particle velocity along the flow direction was computed as $v_p = \Delta x_i / \Delta t$, with $\Delta t = 1/20 \text{ s}$, yielding velocity-time traces for individual particles during the seconds immediately preceding immobilization [Fig. 1(a)].

For the deposition-kinetics measurements [dataset (2)], the suspension was delivered to a flow cell by a peristaltic pump at 11 s^{-1} wall shear rate, and the accumulation of deposited particles along the channel was quantified by automated fluorescence microscopy (upright microscope Eclipse Ni, Nikon; digital camera; programmable motorized stage): Five locations along the stream were imaged every 5 min for 40 min (first location 12 mm downstream of the inlet, subsequent locations spaced by 5 mm), using a $400\times$ magnification with an imaged area of 0.0713 mm^2 per location [33]. The local deposition rate at each location was obtained from the linear slope of particle counts versus time, and, together with a separately measured sedimentation velocity in a zero-shear control experiment, these spatially resolved kinetics were converted into the attachment rate constants K reported in Fig. 1(b) using the transport model that describes spatial variation of deposition rates along the channels (see Ref. [33] for more detail).

The compliant hydrogel coating of thickness ~ 250 nm was synthesized from star-shaped poly(ethylene glycol) (PEG) precursors and anchored with amine end groups to the glass substrate, as described previously [20,35]. The particles had a negative ζ potential -30 mV at pH 7 in NaCl [35]. ζ potential of the hydrogel could not be measured due to its significant ionic conductivity that short-circuited streaming-potential and the total charge was too small for chemical analysis, but the amine end groups must impart it with a weak positive charge at neutral pH [35]. As the particles and coating were oppositely charged, the DLVO force between them was attractive, which was also consistent with the ionic-strength dependence of the attachment rate that dropped with ionic strength, as discussed below [see Fig. 1(b)].

B. AFM force spectroscopy: adhesion maturation and contact parameters

To model and predict the deceleration associated with post-contact rolling/braking scenario, we independently measured adhesion maturation and contact-mechanical parameters by AFM force spectroscopy. Measurements were performed on JPK Nanowizard II and Cypher ES (Asylum Research, Oxford Instruments, USA) AFMs using preglued polystyrene colloidal probes (Novascan Technologies, Ames, USA) of radius $0.5 \mu\text{m}$ and nominal spring constant 5.4 N/m . The true spring constants and detector sensitivities were calibrated using the thermal tune method. PEG hydrogel-coated samples were immersed in 10 mM NaCl for 1 h prior to measurements. Force-distance curves, yielding the variation of the measured force $F(\delta)$ with indentation δ upon loading and unloading, were collected at four distinct positions on each coating, with dwell times t_d spanning $0\text{--}204 \text{ s}$ to quantify the time dependence of adhesion. To probe different indentation regimes and obtain contact parameters under controlled loading force, setpoint (maximal) forces F_m of $35, 75, 85, 150, 300$, and 600 nN were applied, while both loading and unloading velocities were held constant at 450 nm/s . For each value of F_m and t_d , the resulting loading and unloading curves, i.e., $F(\delta)$, were fitted to an adhesive viscoelastic model reported previously to deduce the relevant constant parameters, namely, elastic modulus, mechanical relaxation time, and adhesion energy [20].

III. RESULTS AND DISCUSSION

A. Observed prearrest deceleration versus hovering scenario

Figure 1(a) shows velocity variation for particles that ended up attached (immobilized) within the field of view for the last 2.5 s before arrest, averaged over several such particles, for different shear rates. Despite scattered individual points, the negative linear slopes are robustly determined for each shear rate, with a statistical error under 15% (for confidence level 0.9). Thus, the deducted absolute value of the deceleration $\dot{v}_p = \frac{dv_p}{dt}$ varies between 2 and $6 \mu\text{m/s}^2$ for shear rates from 2.2 to 6.7 s^{-1} , respectively, showing a reasonable correlation between the two.

Due to the optical microscope's finite depth of view, the observed v_p may reflect two different modes of motion, depending on whether the particle is *still hovering* above the substrate or *already in contact* with it. In the former scenario,

the deceleration should reflect drag by the moving fluid superimposed either on horizontal Brownian motion or gravitation settling. If R is the particle radius and $d > 0$ is the gap (separation) between the particle and the wall, the center of the particle is at distance $L = R + d$ from the wall. Unless $d \ll R$, the drag velocity should decrease toward the wall as about σL , i.e., in direct proportion to L and wall shear rate σ . At long distances, gravitational settling will result in L varying with t , the time *remaining until arrest*, as $L \sim tv_s$, where v_s is the sedimentation velocity. Conversely, diffusion generates—on average—approach to the surface, as $L^2 \sim 2Dt$, which becomes faster than sedimentation within BL. The observed average $\dot{v}_p$ will then be on the order of σv_s and $\sigma D/L$ for sedimentation and diffusion, respectively. Both agree with about linear correlation between the deceleration and σ observed in Fig. 1(a). Yet, based on previously reported $v_s \approx 3.85 \times 10^{-8} \text{ m/s}$ [33] and the present wall shear rates $\sigma \approx 2.2\text{--}6.7 \text{ s}^{-1}$, the observed deceleration $\dot{v}_p$ on the order of a few $\mu\text{m/s}^2$ is much faster than $\sigma v_s \sim 0.1 \mu\text{m/s}^2$ expected for convection coupled with sedimentation. We then conclude that, within the non-contact (hovering) scenario, the observed deceleration may only be diffusion controlled, which is in line with our transport modeling [33] and the noisy traces in Fig. 1(a), consistent with random diffusive motion. Indeed, given the previously reported diffusivity $D = 4.8 \times 10^{-13} \text{ m}^2/\text{s}$, over the last 2.5 s , a particle should be on average at a distance $L = \sqrt{2Dt}$ from the wall, which is on the order of $1 \mu\text{m}$, well within the fully developed diffusive boundary layer thickness $D/v_s \sim 10 \mu\text{m}$ [33].

B. Postcontact scenario: adhesive braking model

The observed deceleration rate matching the hovering scenario does not exclude the alternative possibility that particles slow down while already in contact with the substrate ($d = 0$). In that case, v_p would still be dragged by the flow and correlate with σ , but it could be reduced progressively by time-dependent adhesion that ultimately brings the particle to a halt. To the best of our knowledge, such *adhesive braking* scenario was not quantitatively modeled and analyzed for microparticles such as the present ones and in what follows we develop such analyses.

For $d = 0$, particle motion along the surface should occur mainly as rolling rather than sliding, as rolling torque more readily overcomes the dynamic adhesion force, compared with translation [44,45,50]. Braking of the rolling motion must obey Newton's second law for rotation:

$$I \frac{\dot{v}_p}{R} = M = M_H - M_B, \quad (1)$$

where M is the total torque acting on the particle, and $I = \frac{2}{5}mR^2$ is the rotational moment of inertia of the particle viewed as a solid sphere, $m = 4/3\pi R^3 \rho$ and ρ being the particle mass and density. The net torque M sums up the hydrodynamic torque M_H , exerted by the moving fluid on the rolling particle, and the braking torque M_B of opposite sign, resulting from the difference in adhesion force between the leading and trailing edges, as schematically depicted in Fig. 2. This difference stems from the time dependence (maturation) of the adhesion energy per contact area (W), whose value W_T at the trailing edge has had a finite time $t_T > 0$ to mature,

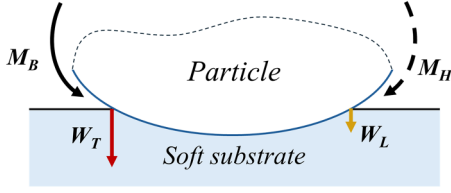


FIG. 2. A sketch of the adhesive particle braking on the hydrogel surface: M_B and M_H are the braking and hydrodynamic torques, and W_T and W_L have the meaning of the force per unit length of the contact area boundary at the trailing and leading edges.

compared with its value W_L at the just formed leading edge ($t_L = 0$). Here, adhesion dynamics resulting in maturation is a critical effect, as without such dynamic symmetry breaking, commonly considered conservative adhesion forces would not generate a net braking torque. At the final braking stage, as v_p decreases, the time of maturation diverges as $t_T \sim 1/v_p$. As a result, M_B increases steeply, while M_H approaches a maximum value for an immobilized particle that eventually becomes negligible compared with M_B ; i.e., the approximation $I \frac{\dot{v}_p}{R} \approx M_B$ becomes reasonable.

The AFM measurements [see next section and Fig. 3(a)] show that, over the short residence times relevant to initial contact formation, adhesion energy W grows approximately linearly with time, i.e., $dW/dt = k_W$ is a constant. Integrating the torque produced by W over the circular contact area of radius a for a particle rolling at velocity v_p yields (see Appendix A)

$$M_B = \frac{4\pi a^3 k_W}{3 v_p}. \quad (2)$$

Hence, neglecting M_H in Eq. (1) and integrating it back in time from $v_p(t=0) = 0$, we obtain adhesive deceleration as

$$\dot{v}_p = \frac{10 \pi k_W a^3}{3 m R v_p}, \quad (3)$$

where a is the contact area radius. Finally, to compute the time dependence of the contact area radius $a(t)$, we employ a previously developed dynamic model of viscoelastic contact (Model 3 of Ref. [20]). As elaborated in Appendix B, the resulting deceleration depends on the nature of the indenting force responsible for forming the contact area. If, as one possibility, this force is gravity, F_g , we obtain

$$\dot{v}_p = \sqrt{\frac{10\pi k_W F_g}{3 m G \tau}}, \quad (4)$$

where G is the elastic modulus and τ is the mechanical relaxation time of the substrate. Conversely, if the dominant force is adhesion itself (W), we obtain an alternative relation. These results are compared below with $\dot{v}_p$ observed in Fig. 1(a):

$$\dot{v}_p = \sqrt{\frac{20\pi^2 k_W W R}{m G \tau}}. \quad (5)$$

C. Adhesive braking: parameter evaluation and analysis

The parameters required for modeling the adhesive braking scenario were deduced from AFM force spectroscopy (FS) measurements using a 1 μm polystyrene colloidal probe on the

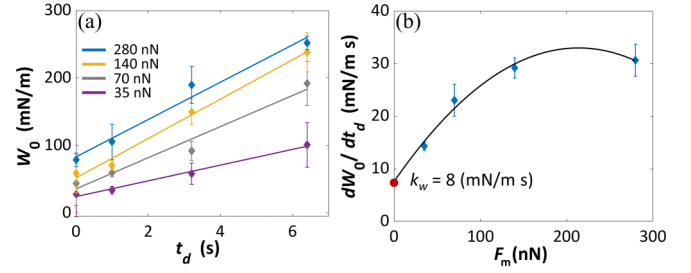


FIG. 3. (a) The adhesion energy W_0 as a function of dwell time under different maximum indentation forces. Magenta, gray, orange, and blue colors correspond to maximum indentation force (F_m) 35, 70, 140, and 280 nN, respectively. Symbols are deduced from FS measurements using the model of Ref. [20] and lines are linear fits. (b) The slope dW_0/dt_d of linear fits in panel (a) vs the applied indentation force and its extrapolation to zero force is marked with a red circle. All AFM-FS measurements were performed under water, using glass slides coated with 250-nm-thick anchored layer of PEG hydrogel and a 1 μm polystyrene spherical probe.

same PEG hydrogel in water using several setpoint forces F_m . To evaluate the maturation rate k_W , the adhesion energy at the centre of the contact area W_0 for different dwell (maturation) times t_d , as well as the elastic modulus G and mechanical relaxation time τ , were deduced using the model of adhesive viscoelastic contact presented previously [20]. This analysis presumes that (1) the local normal stress within the contact area is distributed according to the Hertz contact model and (2) the adhesion force increased both with the dwell time t_d and applied load F_m . Figure 3(a) presents thus deduced time dependence of W_0 and Fig. 3(b) plots the resulting $k_W = \frac{dW_0}{dt_d}$ for different F_m , showing a moderate dependence on F_m . Since the gravitation or adhesion forces acting on particles during deposition are much weaker than loads used in AFM-FS measurements, the value $k_W = 8 \text{ mN m}^{-1} \text{ s}^{-1}$ obtained by extrapolation to the low-force limit [$F_m \rightarrow 0$; see Fig. 3(b)] was used below.

Figure 4 shows parameters G , τ , and W deduced in an equivalent manner from the unloading curves obtained in AFM-FS measurements with zero dwell time at different F_m using the same dynamic contact model [20]. Again, the fitted parameters were extrapolated to $F_m \rightarrow 0$ to obtain estimates representative of the much weaker loading force acting on particles depositing from the flowing suspension (see Fig. 4 and further discussion below on the EHL hypothesis).

Using the AFM-derived values of k_W , G , τ , and W (Figs. 3 and 4), we estimated $\dot{v}_p \sim 10^{-2} \text{ m/s}^2$ for the gravity force and $\dot{v}_p \sim 10^2 \text{ m/s}^2$ for adhesion. Gravity generates a much weaker indenting force, compared with adhesion, yet both estimates exceed the observed deceleration $\sim 10^{-6} \text{ m/s}^2$ by many orders of magnitude. This indicates that, regardless of the nature of the loading force, once a particle touches the substrate and forms an incipient contact, adhesive braking would immobilize it almost instantly. Therefore, we may confidently rule out the postcontact adhesive braking as the mechanism behind particle deceleration in Fig. 1(a). Essentially, great mismatch between observed deceleration and adhesive braking rules out any contact up until the moment of arrest.

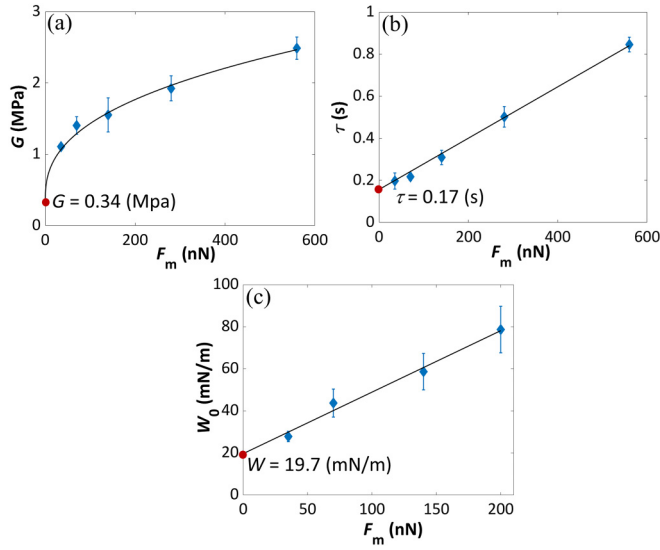


FIG. 4. Variation in the elastic modulus [panel (a)], relaxation time [panel (b)], and adhesion energy W_0 [panel (c)] deduced using Model 3 of Ref. [20] from unloading curves obtained by AFM-FS measurements with zero dwell time as a function of the maximum indentation force F_m . Red dots indicate G , τ , and W_0 values extrapolated to zero force. The measurements were performed under water, using glass slides coated with 250 nm thick anchored layer of PEG hydrogel and a 1 μm polystyrene spherical probe.

This conclusion, however, immediately poses another question. With nearly instant, strong and irreversible immobilization upon contact, the deposition kinetics should be close to the perfect-sink regime, described by the well-known Levich-Smoluchowski solution representing the lower limit in the presence of sedimentation [15,33]. Yet, the observed rates were far lower. While the traces in Fig. 1(a) correspond to the few particles that ended up attached within the observed region, vast majority of particles observed near the surface, within the field of view, apparently stayed out of contact.

To demonstrate this quantitatively, we examined particle deposition rate along the channel for the same conditions. Using a transport model reported previously that separates attachment kinetics from bulk transport, we could deduce the ratio of the attachment rate constant K to sedimentation velocity v_s [33]. While perfect sink corresponds to $K \gg v_s$, Fig. 1(b) shows that, in 10 mM NaCl, K was only slightly above v_s and sharply dropped far below the perfect-sink rate in 20 mM and further in 50 mM solutions. This finding strongly suggests that most particles are effectively prevented from coming in direct contact and remain separated from the substrate by a fluid gap. This gap may be sustained only in the presence of a repulsive force opposing adhesion. We rule out electrostatic (DLVO) interactions as such repulsive barrier, since negatively charged polystyrene beads and the positively charged PEG hydrogel at neutral pH must interact attractively at all separations [35]. This is also consistent with the results in Fig. 1(b). Indeed, in contrast to DLVO repulsion progressively screened with increasing ionic strength for similarly charged surfaces or colloids, here screening progressively reduces *attraction* between the particles and substrate; thus, K steadily decreases. Likewise, we also rule out the

inertial lift, as it is negligible at present shear rates, channel dimensions, and particle sizes, and becomes substantial only at $\sigma > 100 \text{ s}^{-1}$ [15,33].

D. EHL as repulsive barrier: hypothesis and its test

Given all other repulsive forces must be ruled out, we hypothesize here—as the only remaining plausible explanation—that the barrier that allows the particle-surface gap to sustain in the present system is the EHL force. According to the current theory, EHL magnitude is given by the following general expression [47,51,52]:

$$F_{\text{EHL}} \cong \frac{\mu^2 v_p^2}{G} \left(\frac{R^*}{d} \right)^n, \quad (6)$$

where μ is the dynamic viscosity and n is an “elastic-transfer” exponent that encodes how efficiently the hydrodynamic pressure field couples to normal substrate deformation [47,51]. The parameter R^* is a regime-specific length scale that bundles the particle radius R , the soft layer’s thickness l (when it is finite), and Poisson ratio ν . The value of n depends on the solid surface distortion by the fluid pressure p in the gap relative to the characteristic lateral hydrodynamic footprint dimension $r_c \sim (Rd)^{1/2}$ [47,51]. For a *semiinfinite* half-space, $R^* \cong R$ and the stress Green’s function distributes the load through the bulk, thus the compliance decays as $\sim d^{-2}$ and $n = 5/2$ [47,51], which was confirmed by AFM measurements on polydimethylsiloxane substrate [47]. For a *thin compressible* substrate (Winkler limit) the deformation is essentially local, thus the substrate compliance scales as $\sim d^{-5/2}$ [47], which gives $n = 3$ predicted analytically by Urzay [42,47,51–53] and verified on dense hyaluronan brushes mimicking endothelial glycocalyx [42]. On the other hand, for a *thin incompressible* layer ($\nu \approx 0.5$), the need to conserve volume enforces additional lateral spreading of solid stress, steepening the compliance scaling to $\sim d^{-7/2}$ and yielding $n = 4$ [51]. Larger n values produce a faster divergence of F_{EHL} as the particle approaches the surface, extending its range. Presumably, $n = 4$ applies to the present case of thin and swollen hydrogel layer, which is thin and, similar to regular liquids, must be nearly incompressible mechanically (large bulk modulus hence $\nu \approx 0.5$) despite easy shearing (small G) [54].

To form an effective barrier for diffusion, the associated energy must exceed the thermal energy $k_B T = 4 \times 10^{-21} \text{ J}$ at distances above those when adhesion becomes substantial. That means an energy exceeding $k_B T$ must be expended to perform work against F_{EHL} or, equivalently, overcome a quasipotential energy barrier $U_{\text{EHL}}(d) = \int_{\infty}^d F_{\text{EHL}}(x) dx$. This criterion may be alternatively justified by noting that, to suppress deposition substantially, the factor $e^{U/k_B T}$ in the theoretical expression for the attachment rate constant K [33] must become large beyond the range of attractive adhesion forces. Since in our experiments, $l = 250 \text{ nm}$ and $R = 500 \text{ nm}$ are of the same order of magnitude, we have $l \cong R \cong R^*$ and $v_p \cong \sigma R$. Along with Eq. (6), these estimate the energy barrier as

$$U_{\text{EHL}}(d) \cong \frac{\mu^2 v_p^2 R^{*n}}{G(n-1)} d^{1-n} \cong \frac{\mu^2 \sigma^2 R^{n+2}}{G_0 d^{n-1}}, \quad (7)$$

where G_0 is understood as the elastic modulus of the hydrogel in its outmost part or, equivalently, at small deforming force.

Equation (7) shows that $U_{\text{EHL}}(d)$ critically depends on G_0 and n . The use of small-force limit G_0 in Eq. (7) is justified by the fact that the gravity or adhesion forces acting on a particle during deposition are far smaller than the indenting force F_m in AFM measurements and generate far smaller deformations. A depositing particle then deforms mainly the outermost layer, whose limiting small-strain modulus we identify as G_0 , and transmits only a small load to the underlying substrate. Typically, G of gels varies with depth, since the upper tens of nanometers often comprise loose dangling chains and brush-like structures having a larger conformational freedom than the bulk beneath, which may also exhibit a cross-link-density gradient [54]. In addition, and apparently more important in the present case, the sensitivity of compliance to δ is further enhanced by the fact that finite thickness of the soft substrate layer makes the effective compliance decrease when indentation δ approaches l . This poses the challenge of accurately determining G_0 of hydrogels in indentation experiments (AFM-FS), when it varies with the depth of indentation δ .

Although nonlinear extrapolation of G to $F_m \rightarrow 0$ in Fig. 4(a) could supply a rough estimate of G_0 , it is highly uncertain and likely an overestimate. First, these G moduli reflect the mechanical response to relatively large mechanical force, hence substantial indentations much beyond the soft outermost part of the hydrogel. Second, these moduli were deduced from unloading (retraction) curves, thus the adhesive force may add to indentation δ , taking the system far from the $\delta \rightarrow 0$ limit, even after short dwell times. Therefore, an alternative estimate for smallest indentations was obtained from the loading (approach) part of measured AFM-FS curves. Since during loading the adhesive contact has a minimal time to mature, $F(\delta)$ should be close to the Hertz relation for incompressible nonadhesive substrate:

$$\delta - \delta_0 = \left(\frac{9}{16RG^2} \right)^{1/3} F^{2/3}, \quad (8)$$

where δ_0 corresponds to the hydrogel-fluid boundary. This relation was applied to the loading curves, and the slope of $F^{2/3}$ versus δ , displayed in Fig. 5(a), was converted to the apparent Hertz modulus G_H [Fig. 5(b)]. Its dependence on δ was then extrapolated to $\delta = 0$ using a second-order polynomial fit. This yielded a much smaller $G_0 \approx 870$ Pa [Fig. 5(b)], but, again, due to uncertainty of nonlinear extrapolation, we use below both $G_0 = 870$ Pa and a still smaller $G_0 = 400$ Pa.

Figures 5(c) and 5(d) illustrate how $U_{\text{EHL}}/k_B T$ computed using Eq. (7) varies with the normalized gap thickness d/R for a $1 \mu\text{m}$ particle. Figure 5(c) displays its variation for shear rate $\sigma = 4.4 \text{ s}^{-1}$, two representative elastic moduli, $G_0 = 870$ Pa (solid curves) and 400 Pa (dashed), and different n values. For $n = 5/2$ (bulk substrate), U_{EHL} exceeds $k_B T$ only for unphysically thin gap $d/R \sim 0.001$ or $d < 1$ nm that is unlikely to sustain even under weakest adhesive forces. Yet, larger n values shift this threshold to more realistic separations above $d/R \sim 0.01$ or $d \sim 10$ nm for a thin and incompressible layer ($n = 4$) and even for the Winkler layer ($n = 3$) [51]. Moreover, the modulus extrapolated from AFM data could still overestimate G_0 and nontrivial effects such as formation

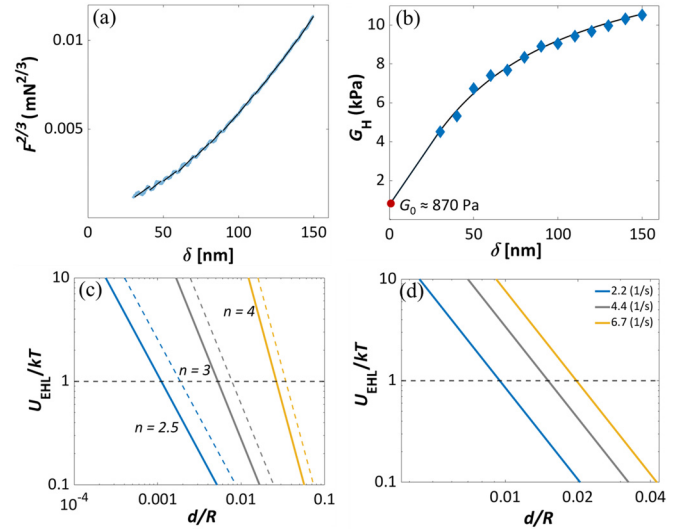


FIG. 5. Substrate mechanical characteristics and predicted repulsive EHL pseudopotential. (a) Load-indentation curve measured by AFM-FS for a $1 \mu\text{m}$ polystyrene sphere on a 250 nm hydrogel in 10 mM NaCl. (b) Hertz model-based apparent moduli: Symbols are derived from the curve slope in panel (a), and line is extrapolation to zero indentation. (c) EHL pseudopotential $U_{\text{EHL}}/k_B T$ vs normalized gap d/R for shear rate $\sigma = 4.4 \text{ s}^{-1}$ and $G_0 = 870$ Pa (solid line) and 400 Pa (dashed), for different exponents n . (d) $U_{\text{EHL}}/k_B T$ vs d/R for shear rates $\sigma = 2.2, 4.4, 6.7 \text{ s}^{-1}$, $n = 4$, and $G_0 = 870$ Pa.

of a “dimple”-like surface profile under the particle [51,53] might further lower the local stiffness. An extra lift, equivalent to softening, may also arise from particle rotation [53].

Figure 5(d) highlights the variation of U_{EHL} with shear rate in the present range according to Eq. (7) for $n = 4$, $R^* = R = 0.5 \mu\text{m}$, and $G_0 = 870$ Pa. For shear rates in Fig. 1(a), the calculated lift energy exceeds $k_B T$ at $d/R = 0.02$ – 0.05 , i.e., separation $d = 10$ – 25 nm . Notably, this range is fully consistent with the sharp drop in the attachment rate constant K between 10 and 50 mM NaCl in Fig. 1(b). Indeed, for these ionic strengths, the Debye length, setting the range of DLVO attraction, drops well under above d values, from 3 nm in 10 mM to 1.4 nm in 50 mM , while other possible attractive and repulsive adhesion forces, such as dispersive, steric, and hydration, have an even shorter range [25]. As the Debye length gets shorter, EHL more efficiently cuts off the DLVO attraction and increasingly suppresses the attachment.

We may also verify that the roughness of hydrogel surface does not alter the conclusions. The rms roughness r_{rms} of the present hydrogel was estimated to be on the order of $\sim 1 \text{ nm}$ [35]. For a thin gap, when d is far below the hydrodynamic footprint size $r_c \sim (Rd)^{1/2} \sim 100 \text{ nm}$ for above EHL range [47,51] and $r_{\text{rms}} \ll d$, the leading correction to the pressure within the gap—hence to any lift force—scales as $(r_{\text{rms}}/d)^2$ [55], which is at most percent level and inessential.

IV. CONCLUSION

In summary, the present analysis, supported by experiments supplying relevant physical quantities, unravels the interplay of dynamic phenomena behind the unique resistance

of soft hydrogels to particle deposition and fouling. Using near-surface particle deceleration as a mechanism-sensitive indicator combined with several dedicated deposition and adhesion experiments and modeling, we discriminate one by one commonly assumed mechanisms and demonstrate that all known adhesion forces, both contact and noncontact, are inconsistent with the observed particle deceleration and deposition rates. Specifically, a few seconds-long deceleration of particles near the hydrogel before attachment is incompatible with the nearly instant immobilization expected for postcontact adhesive braking. Along with the observed very low rate (probability) of attachment, this indicates that most near-surface particles stay hovering above the surface, held by a force that efficiently counters gravity and attractive DLVO interactions. We rule out common repulsive forces such as inertial lift and short-range adhesion interactions, as well as interferences coming from nonideal surface topography, and conclude that EHL is the only force that may efficiently generate such repulsion. Since EHL arises from coupling between lubrication pressure to substrate deformation, it is greatly amplified by the exceptionally soft outer layer of the substrate and may generate a net repulsive force that cuts off adhesive interactions and imposes a barrier that reduces the attachment rate substantially. Using available model relations and experimentally determined parameters, we validate this hypothesis and verify that the EHL force is of sufficient magnitude and extends to the relevant range.

In outlook, we believe present findings may lead to predictive physical models that connect measurable properties to observed deposition and attachment rates for microparticles on soft surfaces. Such models are absent at present; thus, estimation of rate parameters such as K relies on phenomenology rather than underlying physics [33]. Connecting the deposition with independently measured dynamic mechanical and adhesive characteristics of particles and substrate may pave the way to genuine predictive modeling of deposition rates for given flow conditions. This may help identify needs for further insights, e.g., depth heterogeneity of water-gel interfaces, and guide design of optimal antifouling coatings. In this respect, as EHL-controlled mechanism is nonspecific, highly compliant hydrogel coatings may offer a uniquely broad resistance to deposition and fouling, even in the presence of specific chemical or biochemical interactions between particles and substrate.

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

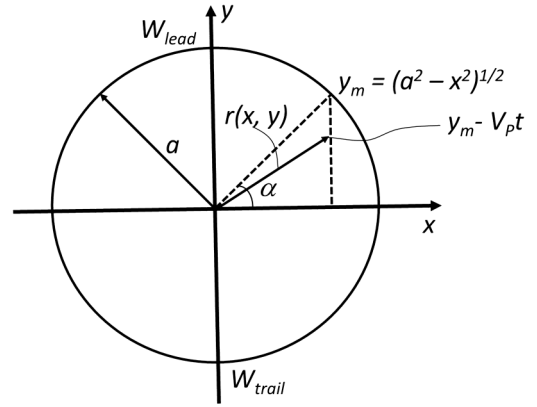


FIG. 6. Contact area of the braking particle during adhesive braking. W_T and W_L are the adhesion energies per area at the trailing and leading edges of the contact, a is the contact radius, x and y are the longitudinal (along the flow) and transversal cartesian coordinates, and r and α are the polar coordinates of a point within the contact area, respectively. y_m is the y coordinate of the leading edge for a given x .

APPENDIX A: DERIVATION OF EQ. (2)

We will define the coordinates within the contact area as illustrated in Fig. 6. The adhesion force is generated by the work of adhesion W at the perimeter of the circular contact area of radius a . A differential arc of the circle of length $a d\alpha$ then generates a force $dF = W(\alpha) a d\alpha$. For a given x , the two such arcs at the trailing and leading edges at (x, y_m) and $(x, -y_m)$ generate a differential torque $dM_B = 2y_m \Delta W(\alpha) a d\alpha$, where ΔW is the difference in W between the two arcs. The total braking torque M_B is found as the following integral:

$$\begin{aligned} M_B &= 2 \int_0^{\pi/2} 2y_m \Delta W(\alpha) a d\alpha \\ &= 4a^2 \int_0^{\pi/2} \Delta W(\alpha) \cos(\alpha) d\alpha, \end{aligned} \quad (A1)$$

where we used $y_m = a \cos(\alpha)$.

For a given x , ΔW develops as a result of maturation of adhesion force due to osmomechanical relaxation over the time $t_m(x) = 2y_m(x)/v_p$ that it takes the leading edge to become the trailing one [20]. Within our model (see the previous section and Ref. [20]), the adhesion energy W relaxes unevenly across the contact area in proportion to the nonuniform stress field. Specifically, the rate of its increase $\dot{W}$ is zero at the edge and maximal in the centre of the contact area. We simplify by assuming this rate matches the Hertz model's distribution of the normal stress (pressure) and varies with the radial distance r from the contact centre as $\dot{W}(r) = \dot{W}_0 [1 - (r/a)^2]^{1/2}$, where $\dot{W}_0 = k_w$ is the maximal rate at $r = 0$ [42]. Therefore, we have

$$\Delta W(x) = \int_0^{t_m} \dot{W}[x, y(t)] dt = \frac{\dot{W}_0}{a} \int_0^{t_m} [a^2 - r^2]^{1/2} dt. \quad (A2)$$

The radial coordinate r may be expressed as (see Fig. 6)

$$r^2[x, y(t)] = x^2 + (y_m - v_p t)^2 = a^2 - 2v_p y_m t + (v_p t)^2. \quad (A3)$$

Inserting Eq. (A3) into Eq. (A2) and integrating, we get

$$\begin{aligned}\Delta W(x) &= \frac{\dot{W}_0}{a} \int_0^{t_m} [2v_p y_m t - (v_p t)^2]^{1/2} dt \\ &= \frac{\dot{W}_0}{a} \int_0^{t_m} [t_m t - t^2]^{1/2} dt = \frac{\pi \dot{W}_0 v_p t_m^2}{8a}.\end{aligned}\quad (\text{A4})$$

Therefore,

$$\begin{aligned}M_B &= \frac{\pi \dot{W}_0 a v_p}{2} \int_0^{\pi/2} t_m^2(\alpha) \cos(\alpha) d\alpha \\ &= \frac{2\pi \dot{W}_0 a^2}{v_p} \int_0^{\pi/2} \cos^3 \alpha d\alpha = \frac{4\pi k_W a^3}{3v_p}.\end{aligned}\quad (\text{A5})$$

APPENDIX B: DERIVATION OF EQS. (4) AND (5)

Estimating the deceleration slope predicted by Eq. (3) in the main text requires that the variation of the contact radius, a , be determined under two distinct scenarios. In the first scenario, gravity is assumed to be the primary (vertical) force F_g driving the formation of the contact area, with adhesive forces being negligible. In the second scenario, adhesion dominates the contact formation, neglecting gravitational force. Before addressing these individual cases, a general time-dependent expression for a is derived by considering the two force balances within the JKR model modified to allow a viscoelastic rather than purely elastic response [25]. The following equations represent the force balance in the vertical and horizontal direction (see Ref. [20]) integrated over time,

$$\int_0^t \frac{d}{dt} \left(\frac{a^{3/2}}{R} - \frac{\delta}{a^{1/2}} \right)^2 dt = \frac{8\pi}{3} \int_0^t \frac{W}{G\tau} dt, \quad (\text{B1})$$

$$\int_0^t \frac{d}{dt} \left(3a\delta - \frac{a^3}{R} \right)^2 dt = \int_0^t \frac{2F_g}{G\tau} dt. \quad (\text{B2})$$

After integrating Eqs. (B1) and (B2) and rearranging, we obtain

$$a^3 = \frac{3R}{2} \left(\frac{8\pi W}{3G\tau} \right)^{1/2} (at)^{1/2} + \frac{RF_g}{G\tau a}. \quad (\text{B3})$$

The first term essentially represents the effect of adhesion, while the second term expresses the effect of gravity. Keeping

the second term only, we derive the expression for the case when gravity dominates the formation of the contact area,

$$a = \left(\frac{F_g R}{G\tau} t \right)^{1/3}. \quad (\text{B4})$$

Substituting Eq. (B4) into Eq. (3) in the main text, we have

$$\dot{v}_p = \frac{10}{3} \frac{\pi k_W F_g}{m v_p G\tau} t. \quad (\text{B5})$$

Equation (B5) is integrated back in time starting from the full stop, i.e., the moment of complete immobilization ($v_p = 0$), which yields

$$v_p = \left(\frac{10}{3} \frac{\pi k_W F_g}{m G\tau} \right)^{1/2} t \quad (\text{B5a})$$

or

$$\dot{v}_p = b = \left(\frac{10}{3} \frac{\pi k_W F_g}{m G\tau} \right)^{1/2}. \quad (\text{B5b})$$

Conversely, when adhesion (W) dominates the formation of the contact area in Eq. (B3), we obtain

$$a = \left[\frac{3}{2} \left(\frac{8\pi W t}{3G\tau} \right)^{1/2} R \right]^{2/3}. \quad (\text{B6})$$

After inserting Eq. (B6) into Eq. (3) in the main text, we get

$$\dot{v}_p = \frac{20\pi^2 k_W W R}{m G\tau v_p} t, \quad (\text{B7})$$

which may be similarly integrated back in time starting from $v_p = 0$ to yield

$$v_p = \left(\frac{20\pi^2 k_W W R}{m G\tau} \right)^{1/2} t \quad (\text{B8a})$$

or

$$\dot{v}_p = b = \left(\frac{20\pi^2 k_W W R}{m G\tau} \right)^{1/2}. \quad (\text{B8b})$$

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