

Getting out of a tight spot: Cooperative unclogging of hydrogel particles in disordered porous media

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(Received 5 May 2025; accepted 25 June 2025; published 11 July 2025)

We use event-driven pore network modeling to study the transport of hydrogel particles through disordered porous media—a process that underlies diverse applications. By simulating particle advection, deformation, and clogging at the pore scale, we identify a dimensionless “squeezing parameter” that quantitatively predicts the depth to which particles penetrate into a given medium across diverse conditions. Our simulations also uncover a surprising cooperative effect: Adding more particles enables them to penetrate deeper into the medium. This phenomenon arises because individual particles redirect fluid to adjacent throats, forcing nearby particles through tight pores that they would otherwise clog. Altogether, these results help to establish a quantitative framework that connects microscopic particle mechanics to macroscopic transport behavior.

DOI: [10.1103/j3jz-x97q](https://doi.org/10.1103/j3jz-x97q)

Introduction. Hydrogels are crosslinked networks of hydrophilic polymers that can absorb up to $\sim 10^3$ times their dry weight in water while maintaining structural integrity. The transport of hydrogel particles through porous media underlies a broad range of processes in energy, industry, medicine, and sustainability. For example, hydrogel particle suspensions are used to clog high-permeability zones in oil reservoirs, promoting fluid redirection and oil recovery from bypassed lower-permeability zones [1–8]. A similar approach can improve the efficiency of geothermal energy extraction, which is similarly plagued by reservoir heterogeneity [9,10]. In other applications such as groundwater remediation [11–13] and drug delivery [14], hydrogel particles are used to carry useful compounds, such as treatment chemicals and therapeutic agents, through porous groundwater aquifers and biological tissues/gels, respectively. All these uses of hydrogel particles require their spatial distribution through a disordered porous medium to be predictable and precisely controlled. However, achieving such predictability is still an open challenge; as a result, applications often proceed by trial and error, with highly variable results. Here, we address this challenge.

The opacity of porous media typically precludes direct visualization of particle transport *in situ*. Therefore, recent work has employed numerical simulations to characterize the transport of hydrogel particles in simplified settings. For

example, studies have examined how hydrodynamic forces can enable a *single* hydrogel particle to squeeze through an individual pore constriction (a “throat”) [15–19]; more recent work extended these studies to the case of a deformable particle moving through a network of multiple interconnected pores [20]. Other work hints that interactions between *multiple* particles—both due to short-ranged collisions [21,22] and longer-ranged alterations in the flow of the suspending fluid as particles squeeze into/clog pores [23]—can strongly influence particle transport. However, these studies focused on ordered networks of identical pores, thus neglecting an inherent feature of most porous media: disorder arising from variations in the pore sizes, which can greatly alter flow and transport [24,25]. As a result, for a suspension of hydrogel particles of a given size, internal permeability, and stiffness, injected into a disordered porous medium of a given geometry, it is still not possible to predict what the spatial distribution of particles and resulting pore-scale changes in fluid flow will be.

Here, we address this gap in knowledge using event-driven pore network modeling of hydrogel particle suspensions injected into porous media of tunable degrees of disorder. We use simulations to characterize how the distance that particles penetrate into a medium varies with the imposed flow rate, degree of disorder in the medium, and hydrogel properties. Across the broad range of conditions tested, our results can all be quantitatively rationalized by considering the competition between the flow-induced viscous stress on a particle to the mechanical stress required to squeeze it through a pore throat. Our simulations also reveal a surprising phenomenon: Increasing the concentration of injected particles *promotes* their penetration into a disordered medium, overturning the common assumption that more concentrated suspensions are more likely to cause clogging. We trace the origin of this cooperativity to dipolarlike alterations in the flow through

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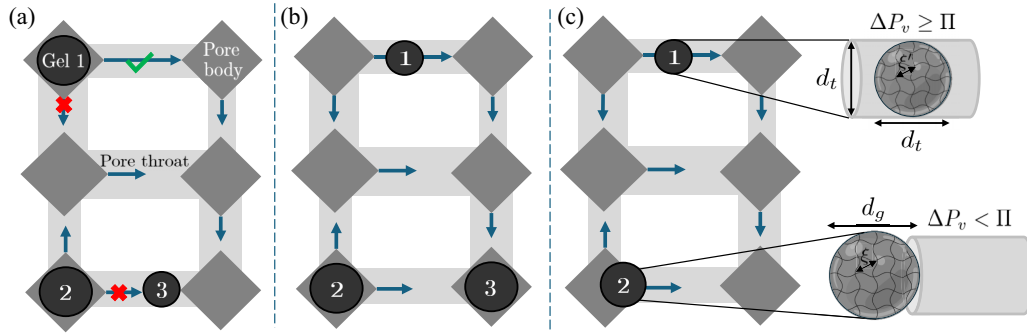


FIG. 1. Schematic of the event-driven pore network model. (a) Hydrogel particles only move into vacant throats with the fastest local flow speed. (b) The particle with the shortest transit time across its throat (here, No. 3) moves into the next pore body while other particles move partially through their throats based on this transit time. (c) Hydrogel particles can only compress and squeeze into a pore throat if the viscous stress ΔP_v exceeds the compressive stress Π (No. 1); otherwise, they continue to clog the throat (No. 2). Here, d_g and ξ represent the diameter and internal mesh size of the unconfined particle, while ξ' represents the smaller mesh size of a compressed particle.

the medium induced by individual particles. Altogether, our work establishes a quantitative framework to help predict and guide ways to control the distribution of hydrogel particles in a porous medium, with implications for diverse applications.

Event-driven pore network model. We follow the established approach of pore network modeling [26–28] by representing each porous medium as a one- or two-dimensional (1D or 2D) square lattice of pore throats (the edges of the network) connecting larger pore “bodies” (the nodes). This approach greatly simplifies the complexity of typical porous media while incorporating the essential physics of fluid flow through the different pores, flow-induced particle advection and squeezing into/clogging of individual pore throats, and the resulting changes in flow behavior and the distribution of particles over larger scales (Fig. 1).

In particular, we represent the pore throats as cylinders with diameters d_t chosen randomly from a normal distribution with a prescribed standard deviation σ , using the absolute value of any nonphysical negative values of d_t if they are chosen. This choice provides a straightforward way to tune the ratio $\Sigma \equiv \sigma/\bar{d}_t$, where $\bar{d}_t$ is the mean throat diameter, in the model; we refer to Σ as the degree of disorder. The length l_t of each cylinder is then set based on the lattice spacing and diameters of the adjacent throats. We consider slow, viscous, incompressible flows with fluid injected into the throats spanning one side of the network (the “inlet”) with a prescribed total flow rate Q . The pressure drop across a pristine throat is given by Poiseuille’s law [20,22,23], $\Delta P_t = \frac{8\mu l_t}{\pi(d_t/2)^4} Q_t$, where μ is the fluid viscosity and Q_t is the throat flow rate. In this representation, the advection, squeezing, and clogging of hydrogel particles is dictated solely by the geometry of the pore throats, which can only contain one compressed particle at a time; the bodies simply act as nodes in this network that can also only contain a single fully swollen particle at a time, and their sizes are otherwise not explicitly considered. Future extensions of our work could relax this assumption by, e.g., allowing multiple particles to enter the pores, thereby enabling one to explore more concentrated particle suspensions. Applying conservation of mass at each body then results in a linear system of equations that provides the quasi-steady-state flow profile of the network for each configuration of hydrogel particles, where the equilibration timescales for pressure and

viscous flows are much shorter than the timescale of one particle moving through a throat.

In our simulations, we combine this pore network flow model with event-driven hydrogel particle dynamics based on experimental observations [19] (Fig. 1)—numerically solving this system of equations successively as the distribution of particles throughout the network changes step by step. We initialize the particles in pore bodies along the inlet of the medium [Fig. 1(a)]. The particles have a fully swollen diameter $d_g > \bar{d}_t$ and internal swollen mesh size ξ [magnified schematic in the lower right of Fig. 1(c)], which then sets the internal permeability $\sim \xi^2$ and bulk modulus $K \sim k_B T / \xi^3$ [29]. At each step of the simulation, we determine which hydrogel particles have vacant throats to potentially move into, advected by the flow out of the pore bodies they occupy. We then specify that each such particle will either squeeze into the accessible vacant throat that has the largest pristine flow speed [arrows in Figs. 1(a) and 1(b), upper right of Fig. 1(c)] or become stuck and clog the throat entrance [lower right of Fig. 1(c)], depending on the local flow speed.

In particular, the pressure drop across the throat is then given by $\Delta P_t + RQ_t$, where R is known as the hydraulic resistance of the hydrogel particle and Q_t is the reduced flow rate through both the particle and throat. For a fully swollen particle, $R \sim \mu/(d_g \xi^2)$ [19]. However, the fluid flow through the particle compresses it [19,30–34]. Assuming isotropic compression for simplicity, the compressed diameter is $d'_g < d_g$, the mesh size is then $\xi' = \xi(d'_g/d_g)^{9/4}$ [35,36], the hydrogel resistance is $R \sim \mu d'_g/(d_t \xi')^2$ [19], and the bulk modulus is $K \sim k_B T / \xi'^3$ [29]; investigating the influence of anisotropic deformation mechanics will be an interesting extension of our work. So, in this step of the simulation, we determine the diameter of the compressed particle by balancing the viscous pressure drop across the particle, RQ_t , with the mechanical stress required to compress the hydrogel, $\Pi = K\varepsilon$, where $\varepsilon \equiv (d_g^3 - d'^3_g)/d_g^3$ [19], and then update all flow rates throughout the network accordingly. If $d'_g > d_t$, then the particle cannot fully squeeze into the throat and remains stuck in the upstream pore body, clogging the throat entrance. Once the flow is sufficiently fast to reach the threshold $d'_g = d_t$, however,

the particle is advected through the throat [upper right of Fig. 1(c)], as validated experimentally [19].

Having thereby identified which hydrogel particles will squeeze through throats, we then determine how far they will be advected during this step of the simulation. To do so, we determine the total time required for each particle with $d'_g = d_t$ to transit through its pore throat, based on the local flow speed. The particle with the shortest transit time—which sets the duration of this step of the simulation—is then advected to its downstream pore body, while the others move only partially through their respective throats as dictated by the local flow speed [Fig. 1(b)]. Our numerical simulation then repeats this series of steps—which we refer to as “event driven” because the temporal duration of each step is not fixed, but is determined by the dynamics of the fastest-moving particle in the network—until all particles are stuck ($d'_g > d_t$) or have exited the medium.

Thus, our approach is similar to classical percolation approaches [37], such as the random fuse model [38], in representing a porous medium as a network and studying how transport properties change as individual network elements become nonconducting. However, our model differs in that it captures continuous particle deformation rather than discrete bond failure at critical thresholds. Exploring the connections between the results generated by our event-driven network model and classical percolation models will be an interesting direction for future work.

Single-particle dynamics. To establish a simple baseline, we first examine the transport of a single hydrogel particle across a range of inlet flow rates Q through a 1D medium. For each value of Q tested, we simulate 10^3 different but statistically identical media with the same mean diameter $\bar{d}_t = 0.8d_g$ and prescribed degree of disorder Σ . In each simulation, we measure the number of throats x that the particle penetrates along the length of the medium before it can no longer move.

The probability density function (PDF) of x determined from all simulations of the exemplary case of $\Sigma = 0.5$ and $Q = 3 \times 10^{-12}$ m³/s is given in the inset of Fig. 2. As shown by the curve, the PDF $\sim e^{-x/\Lambda}$, where we term Λ the mean penetration length (normalized by $\bar{d}_t$). As intuitively expected, our simulations indicate that Λ increases monotonically with increasing Q : Hydrogel particles penetrate deeper into the medium at higher flow rates. This intuition can be quantified by comparing the characteristic fluid viscous stress on a compressed particle $\Delta P_v \sim \mu Q_t / (d'_g \xi'^2)$ and the characteristic compressive stress $\Pi \sim K\varepsilon$, with $d'_g = d_t$ and $Q_t = Q$ in 1D; the ratio between these stresses yields a dimensionless group that we term the “squeezing parameter,” $S \equiv \Delta P_v / \Pi$. As shown by the light blue points in Fig. 2, Λ increases as S increases above a critical value ~ 1 , as expected. Moreover, repeating our simulations for a given degree of disorder Σ but independently varying the other input parameters $\bar{d}_t$, ξ , and μ instead of Q yields values of Λ that collapse when presented as a function of S [39], confirming that this dimensionless parameter controls the extent of particle penetration.

This picture suggests that the probability of a hydrogel particle getting stuck in a 1D medium is independent of space and time [40]—as further supported by our finding that the PDF of penetration lengths decays exponentially, indicative of a Poisson process. Specifically, for a given set of input

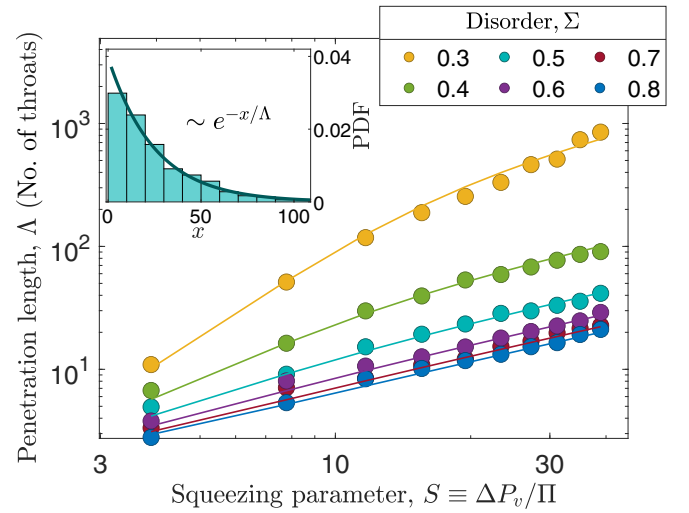


FIG. 2. Penetration of hydrogel particles into 1D porous media. The inset shows the probability density function of the number of throats x that the particle penetrates into the medium for 1000 simulations of a medium with $\Sigma = 0.5$ and $S \approx 12$, following the exponential distribution with mean value Λ as shown by the curve. The main panel shows Λ (each point derived from 10^3 realizations) increasing with increasing squeezing parameter S and decreasing degree of disorder Σ ; curves show the theoretical prediction of Eq. (1). All simulations use $\mu = 10^{-3}$ Pa-s, $\bar{d}_t = 1$ mm, $d_g = 1.2\bar{d}_t$, and $\xi = 10^{-8}$ m in media composed of 200 successive throats, with the exception of $\Sigma = 0.3$ that is instead composed of 1000 throats to accommodate for deeper penetration at low disorder.

conditions parametrized by S , the critical throat diameter $d_t^*(S)$ for particle squeezing is given by equating ΔP_v to Π . As detailed in the Supplemental Material [39], the probability that the particle gets stuck is then equal to the probability that it encounters a throat with $d_t < d_t^*$, and Λ is simply the inverse of this probability:

$$\Lambda(S) = \ln \left[1 - \frac{1}{2} \operatorname{erf} \left(\frac{d_t^*(S) - \bar{d}_t}{\sigma \sqrt{2}} \right) + \operatorname{erf} \left(\frac{d_t^*(S) + \bar{d}_t}{\sigma \sqrt{2}} \right) \right]^{-1}. \quad (1)$$

As shown by the light blue curve in Fig. 2, this prediction agrees well with the simulation data, confirming our intuitive picture. As a final test of this picture, we repeat our simulations for media with varying degrees of disorder Σ . In all cases, Λ increases monotonically with S in excellent agreement with Eq. (1), as shown by the different colors in Fig. 2. These results demonstrate that in 1D, disorder results in a higher likelihood of the particle encountering a throat that is too small for it to pass through, hindering penetration into the medium.

Intriguingly, this trend is reversed in 2D: As shown in Fig. 3, for a given value of S , increasing the degree of disorder Σ increases the hydrogel particle penetration length Λ . Moreover, while Λ again increases monotonically with S , it does so exponentially—not as predicted by Eq. (1) for the 1D case, nor as a power law as expected from percolation theory [20]—as shown by the lines of $\Lambda = \Lambda_0 e^{S/S_c}$ in Fig. 3 with fitting parameters Λ_0 and S_c . The lower right inset shows how Λ_0 monotonically increases with Σ , again reflecting that

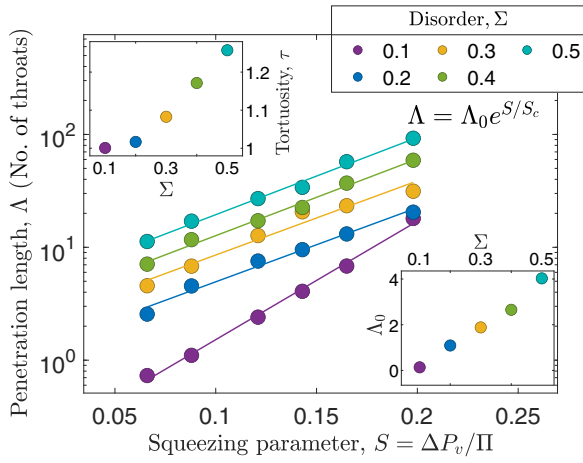


FIG. 3. Penetration of hydrogel particles into 2D porous media. The main panel shows Λ (each point derived from 10^3 realizations) increasing with increasing squeezing parameter S and degree of disorder Σ ; curves show exponential fits $\Lambda_0 e^{S/S_c}$. The top left and bottom right insets show that both the particle path tortuosity τ and characteristic penetration length Λ_0 increase with disorder Σ . All simulations use $\mu = 10^{-3}$ Pa-s, $\bar{d}_t = 1$ mm, $d_g = 1.2\bar{d}_t$, and $\xi = 10^{-8}$ m in media composed of 120×120 throats.

particle penetration is promoted by pore space disorder. Both of these differences with the 1D case result from the increased connectivity of the pore space in 2D: The latter provides an additional degree of freedom for particle squeezing and advection through throats with the fastest flow speeds, which follows tortuous paths through the medium that avoid the smallest throats [20]. As shown in the upper left inset of Fig. 3, our simulations confirm that the tortuosity τ of these paths increases with the degree of disorder Σ . Moreover, the critical value of S required for particle penetration is approximately two orders of magnitude lower than in the 1D case—again due to the increased pore space connectivity [39], since fluid can now route through the N throats transverse to a clogged one, thereby reducing its flow rate by a factor of N (which

is indeed $\sim 10^2$ in the simulations). Further exploring how the transverse transport of particles is enhanced by medium disorder will be a useful direction for future work.

Cooperative dynamics of multiple particles. Next, to probe how multiple hydrogel particles are transported, we repeat our 2D simulations with two particles at different initial transverse separation distances from each other. Remarkably, when both particles are initially next to each other (separated by one throat), the mean penetration length is $\approx 30\%$ larger than in the single-particle case, as shown by comparing the squares to circles in Fig. 4(a). This effect diminishes as the initial transverse separation between particles increases, as we show in the Supplemental Material [39]; as a limiting case, when the particles are separated by as many as 40 pore throats, their penetration into the medium is similar to the single-particle case, as shown by the diamonds in Fig. 4(a). Alternatively, this unexpected effect is promoted as more hydrogel particles are added to the medium, as we also show in the Supplemental Material [39]. This cooperative effect is also sensitive to the relative orientation of the particles: Penetration is only promoted when the particles are close to each other in the transverse, not longitudinal, direction [39]. Taken altogether, these results indicate that the cooperative unclogging of multiple hydrogel particles arises from orientation-dependent alterations in the flow through the medium induced by individual particles. Close inspection of the fluid velocities around a single stuck hydrogel particle confirms and elucidates this expectation. As shown in Fig. 4(b), each particle induces a dipolarlike perturbation to the fluid velocity field characteristic of low Reynolds number flow around an obstacle [41–43]. Thus, when one particle clogs or squeezes through a pore, it redirects fluid to neighboring pores, enabling nearby particles to also squeeze through tight pores.

Discussion. Our work demonstrates that the penetration of hydrogel particles into a porous medium is governed by the competition between the flow-induced viscous stress on a particle and the mechanical stress required to squeeze it through constrictions—providing a quantitative foundation for their applications in energy, industry, medicine, and sustainability.

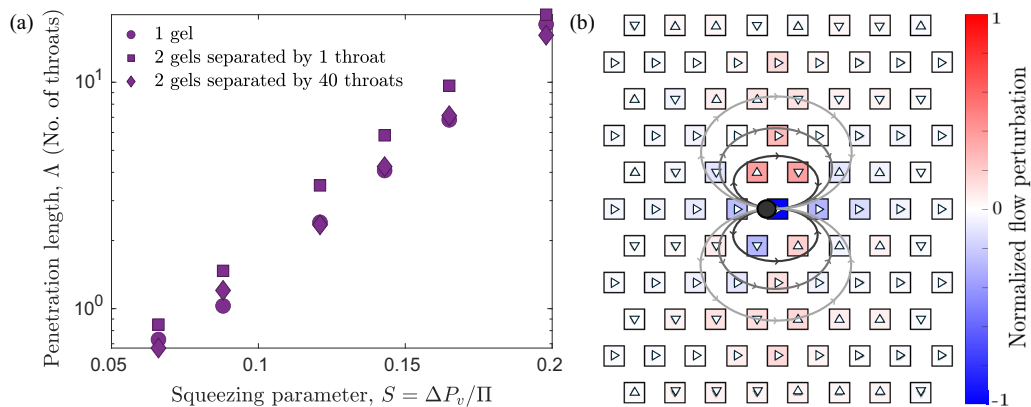


FIG. 4. Cooperative unclogging of hydrogel particles is mediated by dipolarlike flow perturbations. (a) Mean penetration length Λ (each point derived from 10^3 realizations) as a function of S for a single particle (circles), two particles separated transversely by a single throat (squares), or two particles separated transversely by 40 throats (diamonds), in the same 2D medium with $\Sigma = 0.1$ and 120×120 throats. The second case has $\approx 30\%$ more penetration. (b) Dipolarlike perturbation to the flow induced by a particle clogging a pore. The color bar shows the relative change in the flow speed caused by a hydrogel particle; sketched streamlines show the dipolarlike nature of the flow.

Our simulations highlight the pivotal role of dimensionality and disorder: In 1D, increasing disorder hinders particle penetration, whereas in higher-dimensional media, greater disorder actually promotes deeper penetration due to the availability of alternative flow paths. Moreover, the simulations reveal the counterintuitive finding that increasing the particle concentration enhances penetration rather than promoting clogging, as in other systems [44,45]. This cooperative unclogging occurs through dipolarlike alterations in the local flow field, where particles redirect fluid to neighboring pores, facilitating the transport of nearby particles through tight constrictions. Experimentally testing the predictions made here—for example, by visualizing the transport of hydrogel particles through micro/millifluidic obstacle arrays created using lithography or 3D printing [46]—will be an important direction for future work. Moreover, it will be interesting to explore how our findings extend to impermeable but still deformable particles, such as cells, vesicles [47], emulsion droplets [48,49], and colloidal aggregates [50].

Acknowledgments. It is a pleasure to acknowledge Baojun Bai for useful discussions, as well as funding to S.K. through the Chateaubriand Fellowship of the Office for Science & Technology of the Embassy of France in the United States, to L.T. through the INTPART program from the Research Council of Norway (Project No. 309139), to M.R. through the Princeton Center for Complex Materials MRSEC (NSF Grant No. DMR-2011750), and to S.S.D. through NSF Grant No. DMR-2011750, the Project X Innovation Fund, the Camille Dreyfus Teacher-Scholar Program of the Camille and Henry Dreyfus Foundation (SSD). This material is also based upon work by SSD supported by the U.S. Department of Energy's Office of Energy Efficiency and Renewable Energy (EERE) under the Geothermal Technologies Office (GTO) Innovative Methods to Control Hydraulic Properties of Enhanced Geothermal Systems Award No. DE-EE0009790.

Data availability. The data that support the findings of this article are openly available at [51].

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