

Temporal Evolution in the Networks of Porous Materials: From Homogenization to Instability

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S1. SIMULATION ALGORITHM

In our simulations, we tested two types of networks: (i) a topologically ordered (diamond-grid nodes) network, (ii) a topologically random network (2d and 3d). The random network is created using uniformly distributed points with on average $N_x \times N_y$ nodes in the horizontal and vertical directions. The randomly distributed points are then connected using a Delaunay triangulation. The diameter of each edge is sampled from either a uniform distribution with $[1, 14]$, log-normal distribution with $\mu = 3, \sigma = 0.48$, or truncated normal distribution with $\mathcal{N}(\mu = 7.0, \sigma = 3.6)$, where all of the distributions have a coefficient of variation close to 0.5. An external pressure is considered between the left-most nodes and the rightmost nodes ($p_{\text{left}} = 10, p_{\text{right}} = 0$). For each edge, assuming a Poiseuille flow, the fluid flow q and pressure difference δP_e are related through $q_e = C_e \delta P_e$, where $C_e = \pi r_e^4 / 8 \mu L_e$, L_e is the length of the tube, and μ is the viscosity of the fluid. We define $|q_e\rangle$ as the vector of flow through all the edges, and as a result $|q_e\rangle = C_e \Delta |P_n\rangle$ where $|P_n\rangle$ is the vector of pressure at all the nodes, and Δ the transpose of the network's oriented incidence matrix. The orientation (or direction) of an edge is arbitrary selected, and it only determines the positive direction for the fluid flow in that edge. Next, we need to use conservation of mass at the nodes to solve for the network pressure/flux at the nodes/edges. The conservation of mass at each node can be written as

$$|q_n\rangle = \Delta^T \mathbf{C} \Delta |P_n\rangle, \quad (\text{S1})$$

where $\mathbf{C} = \text{diag}(C_e^{(1)}, C_e^{(2)}, \dots, C_e^{(N_e)})$ is a diagonal matrix of edge conductances, and $|q_n\rangle$ is the vector of total incoming flow to each node. The total incoming flow to an internal node is zero inside the network due to the conservation of mass, and can only be non-zero at the boundary nodes. Without loss of generality, we renumber the boundary nodes to $1, 2, \dots, N_B$, where N_B shows the total number of nodes at the boundary. We re-partition Eq. (S1) to obtain

$$\left[\begin{array}{c|c} \Delta_b^T \mathbf{C} \Delta_b & \Delta_b^T \mathbf{C} \Delta_n \\ \hline \Delta_n^T \mathbf{C} \Delta_b & \Delta_n^T \mathbf{C} \Delta_n \end{array} \right] \left[\begin{array}{c} P_1^{BC} \\ P_2^{BC} \\ \vdots \\ P_{N_B} \\ \hline P_{N_B+1} \\ \vdots \\ P_{N_n} \end{array} \right] = \left[\begin{array}{c} q_1^{BC} \\ q_2^{BC} \\ \vdots \\ q_{N_B}^{BC} \\ \hline 0 \\ \vdots \\ 0 \end{array} \right] \rightarrow \left[\begin{array}{cc} \mathbf{A}_{bb} & \mathbf{A}_{bn} \\ \mathbf{A}_{nb} & \mathbf{A}_{nn} \end{array} \right] \left[\begin{array}{c} P_1^{BC} \\ P_2^{BC} \\ \vdots \\ P_{N_B}^{BC} \\ \hline P_{N_B+1} \\ \vdots \\ P_{N_n} \end{array} \right] = \left[\begin{array}{c} q_1^{BC} \\ q_2^{BC} \\ \vdots \\ q_{N_B}^{BC} \\ \hline 0 \\ \vdots \\ 0 \end{array} \right], \quad (\text{S2})$$

where $\mathbf{A}_{st} = \Delta_s^T \mathbf{C} \Delta_t$ and $s, t \in \{a, b\}$. The first N_B elements of the pressure vector (i.e., P_1, \dots, P_{N_B}) represent the pressure at the boundary nodes and the rest are the pressure for the internal nodes. In summary the above equations can be written as a combination of two set of linear equations

$$\mathbf{A}_{bb} |P_{BC}\rangle + \mathbf{A}_{bn} |P\rangle = |q_{BC}\rangle, \quad (\text{S3a})$$

$$\mathbf{A}_{nb} |P_{BC}\rangle + \mathbf{A}_{nn} |P\rangle = 0. \quad (\text{S3b})$$

where $|P_{BC}\rangle = [P_1^{BC}, \dots, P_{N_B}^{BC}]^T$ is the boundary nodes pressure vector, $|q_{BC}\rangle = [q_1^{BC}, \dots, q_{N_B}^{BC}]^T$ is the boundary nodes incoming fluid flux vector, and $|P\rangle = [P_{N_B+1}, \dots, P_{N_n}]^T$ is the unknown pressure vector for the rest of the nodes. If the pressure at the boundary nodes is given, then we can use Eq. (S3b) to solve for the internal pressure

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values $|P\rangle$, and then use Eq. (S3a) to find the required flux at the boundary nodes $|q_{BC}\rangle$. However, if fluid flux vector at the boundary nodes is given (i.e., $|q_{BC}\rangle$ is known), we need to simultaneously solve for the Eqs. (S3a) and (S3b) to find the boundary pressure vector $|P_{BC}\rangle$ and internal nodes pressure vector $|P\rangle$. In either case, solving Eq. (S3) results in the nodes' pressure vector and also the fluid flux vector at the boundary nodes. The fluid flux at each edge can then be calculated using $|q_e\rangle = C_e \Delta |P_n\rangle$. Next, given the flux at each edge q_e , we increase (decrease) the edge radius under erosion (clogging) using

$$\frac{dr_e}{dt} \propto \pm \frac{q_e}{r_e^n}, \quad \leftarrow \text{replace with general law.} \quad (S4)$$

and re-iterate the process to solve for the new fluid flux vector. We use a simple forward Euler for time integration. For each iteration, we choose the time step dt so that $\max(dr_e) = 0.1r_0$, where r_0 is the smallest radius among all edges. This condition guarantees that at each step a small amount of material is eroded and there's no sudden change in the network. We further test the convergence by decreasing $\max(\Delta r_{ij})$ to half and we observe that the average relative change in the flux vector is 1.2%, and the PDFs remain intact without any notable change. The network size for the 2d/3d networks is $N_x = 100, N_y = 50, N_z = 12$ unless mentioned otherwise. In either case, the network has an order of 10^3 edges. The code is publicly made available in a GitHub repository [1].

S2. ANALYTICAL RESULTS FOR FLUID FLUX EXP-TAIL PDF IN A DIAMOND NETWORK

As described in the main text, the PDF of flow in a topologically disordered network of tubes is the same as a structured diamond grid when the radius of the tubes are highly disordered. In this section, we repeat a derivation to show that the observed exponential distribution of fluid flux can be described using a mean-field approach on a structured grid [2-4]. Basically, the random distribution of the diameters along with the conservation of mass in the network are the two main ingredients resulting in an exponential tail distribution. In a diamond grid, the incoming flow to a node is redistributed among the outgoing edges (since fluid mass is conserved). Due to the randomness in the tube's diameter, the redistribution of the incoming flow to a node between the outgoing edges is random variable. The only condition required to hold is that the incoming flux should be equal to the outgoing flux. This model for the flow can be mapped one to one to the problem of force fluctuations in a bead pack [2-4] as shown in Fig. S1. In a bead pack, the force at each layer is redistributed to the next layer where the total force exerted on the next layer should equal to that of the previous layer. Given the above conditions, the flow at layer $L+1$ at node j can be

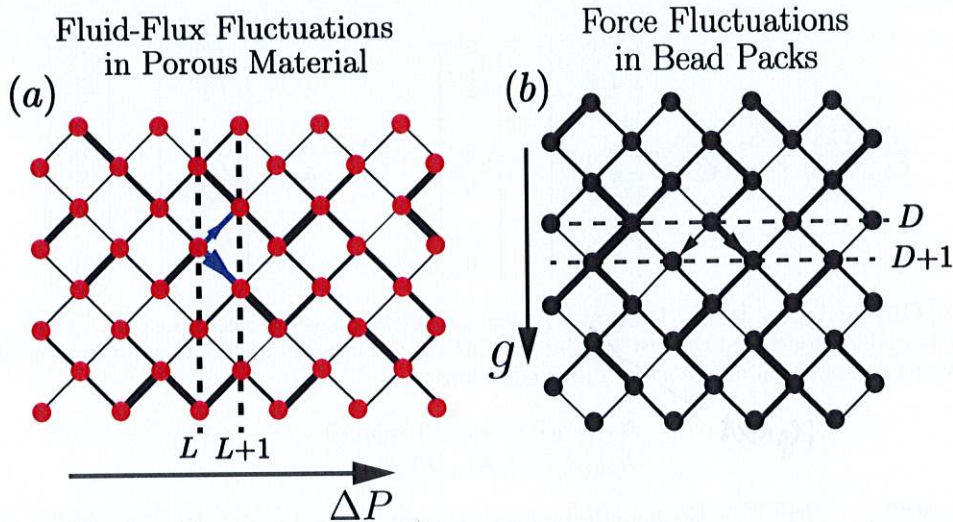


FIG. S1. (a) Schematic of diamond grid network of tubes. The incoming flow to each node is redistributed among the outgoing edges. The thickness of the lines shows the fluid flux transferred through that edge. (b) Schematic diagram showing beads (represented with nodes) and their contacts to the neighboring sites (represented with edges). The thickness of the edges show the force transferred through that contact.

→ ~~unnecessary~~ for completeness, we repeat the derivation of Refs 2-4 which shows that...

obtained as

$$q(L+1, j) = \sum_i w_{ij} q(L, i) = w_{i,i+1} q(L, i+1) + w_{i,i} q(L, i), \quad (S5)$$

where w_{ij} shows the weights by which the flow is redistributed, and $\sum_j w_{ij} = 1$ since the total fluid flux is conserved. Assuming a general distribution of for the weights, $\eta(w)$, we can use a mean-field approximation to find the distribution of q at the layer L , i.e., $p_L(q)$, as

$$p_L(q) = \prod_{j=1}^N \left\{ \int_0^1 dw_j \eta(w_j) \int_0^\infty dq_j p_{L-1}(q_j) \right\} \times \delta \left(\sum_j w_j q_j - q \right), \quad (S6)$$

where N is the number of outgoing edges (e.g., in our structured diamond grid $N = 2$) and $\delta(\cdot)$ is the Kronecker delta function. The constraint that q 's emanating downward should add up to one is accounted for through the definition of $\eta(w)$. Taking the Laplace transform of the above equation and defining $\tilde{p}(s) \equiv \int_0^\infty p(q) e^{-qs} dq$ one obtains

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$$\tilde{P}_L(s) = \left(\int_0^1 dw \eta(w) \tilde{P}_{L-1}(sw) \right)^N. \quad (S7)$$

The above equation gives a recursive relation for the Laplace transformed of fluid flux PDF, $\tilde{P}_L(s)$, where it gradually converges to a distribution $\tilde{P}(s)$ with which the PDF of fluid flux can be obtained. Solving the above equation for a structured diamond grid network, one can find that the converged PDF of the fluid flux becomes $p(q) = 4q \exp(-2q)$ [2-4], which is an exponential tailed distribution.

from finds

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I thought that
the exact form
is $q^a e^{-q}$?
please double-check.

[I would consider removing section S2, it doesn't add much.]

S3. ROBUSTNESS TO TOPOLOGY AND INITIAL CONDITION

In order to check the robustness of our result with respect to the topology of the network, we run our simulations for a three-dimensional random network with random initialization of nodes in space (Fig. S2), and also for a two dimensional topologically ordered network with a diamond grid (Fig. S3), where both networks are initialized with a uniform random distribution for the diameter of the tubes. We find that regardless of the topology of the network, an erosion dynamics with $n > 3$ results in a homogenized network, while $n < 3$ results in the channelization instability. We further check the effect of initial randomness on the fate of the network. We use a two-dimensional diamond-grid network with a narrow uniform initial distribution of tube diameters around an average diameter d_0 with only a very small variation (3%), i.e., the tube diameters are sampled from $\mathcal{U}[d_0(1 - \epsilon), d_0(1 + \epsilon)]$. We again find that for the networks with $n < 3$ channels are formed, while for $n > 3$ the network stays homogenized.

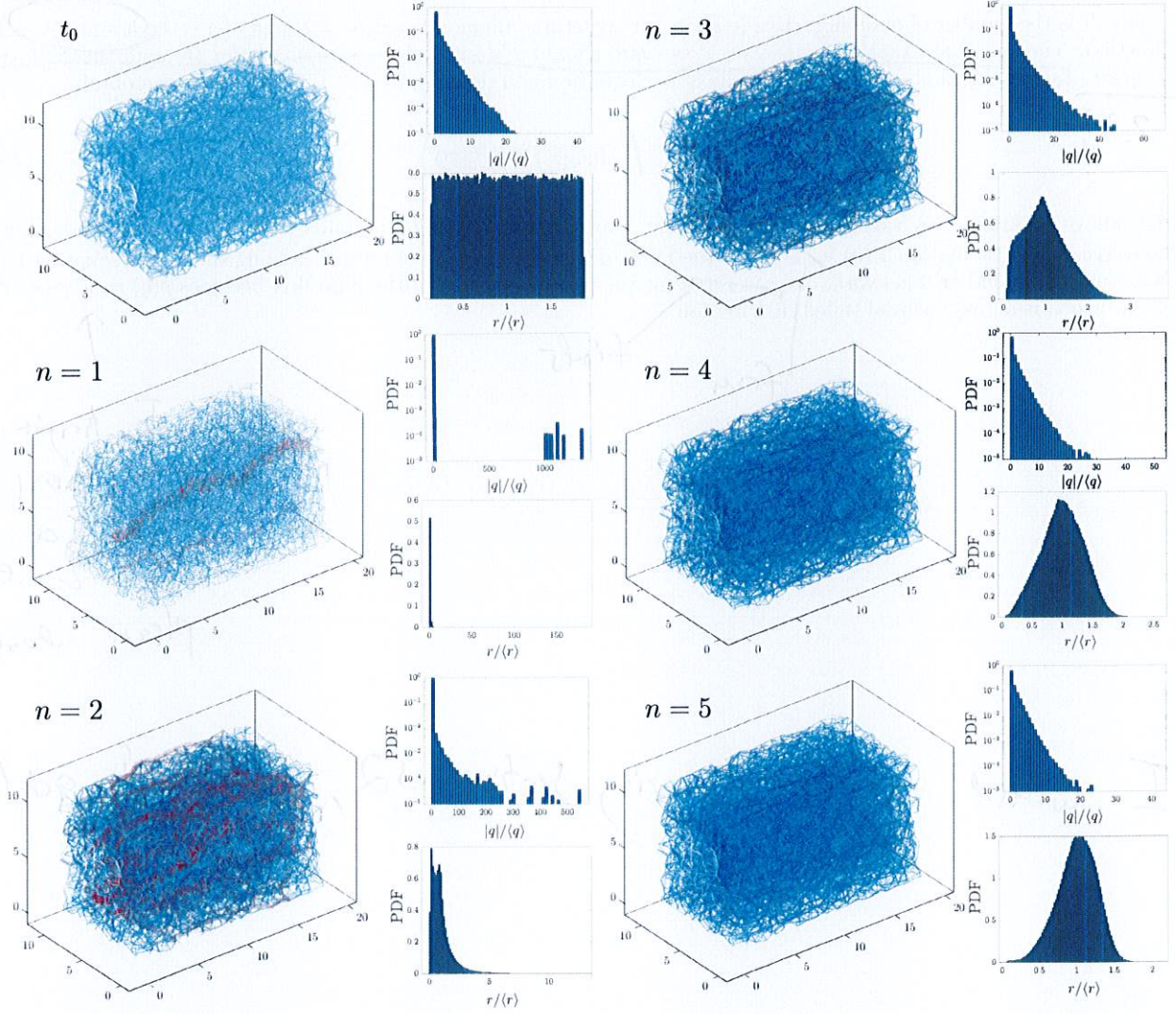


FIG. S2. Erosion in a topologically random 3D network of tubes with $N_x = 50, N_y = 12, N_z = 12$ and an initial uniform broad distribution of tube diameters randomly sampled from $\mathcal{U}[1, 14]$. Snapshots of the network, PDF of normalized fluid flux $q/\langle q \rangle$, and normalized edge radius distribution $r/\langle r \rangle$ at the initial time $t = 0$, and also after N erosion steps for different powers of erosion n are shown. We stop the erosion after N steps such that $\langle r \rangle = 2r_0$ where $r_0 = \langle r_{t=0} \rangle$. The erosion law is based on Eq. (1) in the main text where different powers of n correspond to different models of erosion.

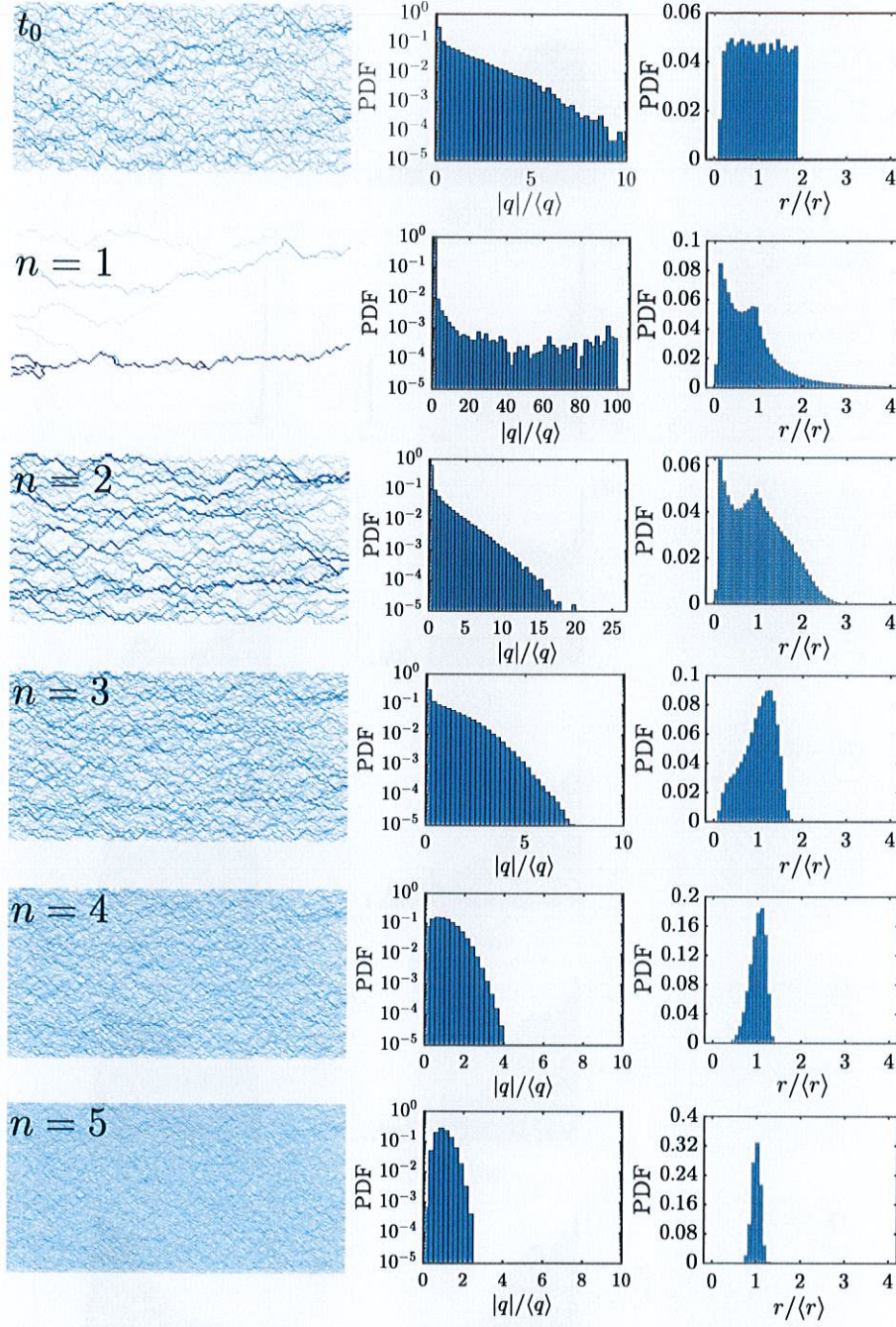


FIG. S3. Erosion in a diamond grid network with $N_x = 100, N_y = 50$ randomly distributed nodes and an initially broad distribution of tube diameter randomly sampled from $\mathcal{U}[1, 14]$. The initial condition is shown with the label $t = 0$ in the first row. Each row afterward corresponds to the simulation result after N steps such that $\langle r \rangle = 2r_0$ where $r_0 = \langle r_{t=0} \rangle$ or twice the initial average radius. The erosion law is based on Eq. (1) in the main text where different powers of n correspond to different models of erosion. The first column is a snapshot of the pore network, the second column is the PDF of normalized fluid flux $q/\langle q \rangle$, and the last column is the PDF of normalized radius $r/\langle r \rangle$.

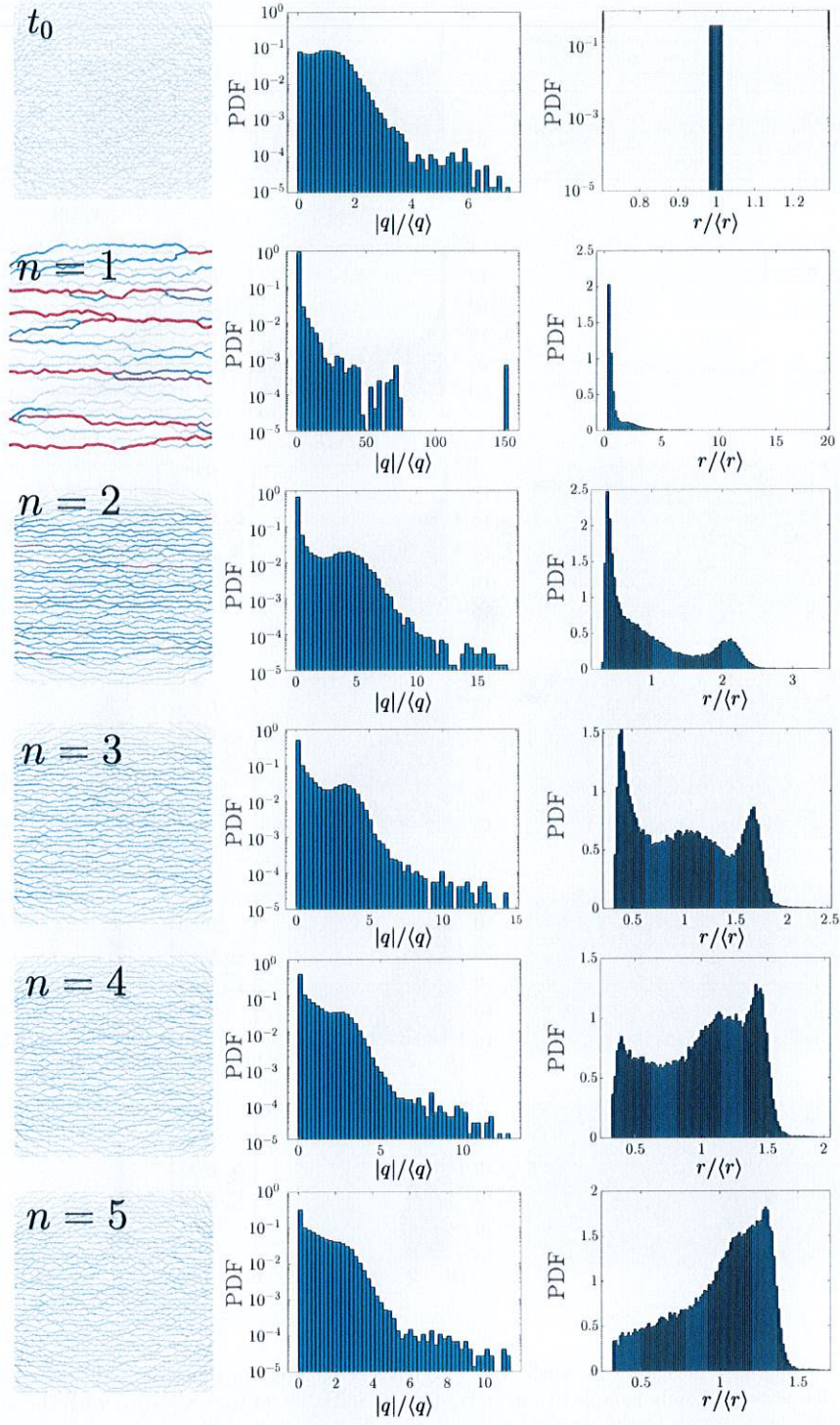


FIG. S4. Erosion in a structured diamond grid network with $N_x = 50, N_y = 50$ and an initially narrow distribution of tube diameters randomly sampled from $\mathcal{U}[d_0(1 - \epsilon), d_0(1 + \epsilon)]$ where d_0 is the average diameter and $\epsilon = 0.03$. The initial condition is shown with the label $t = 0$ in the first row. Each row afterward corresponds to the simulation result after N steps such that $\langle r_{t=N} \rangle = 2r_0$ where $r_0 = \langle r_{t=0} \rangle$ or twice the initial average radius. The erosion law is based on Eq. (1) in the main text where different powers of n correspond to different models of erosion. The first column is a snapshot of the pore network, the second column is the PDF of normalized fluid flux $q/\langle q \rangle$, and the last column is the PDF of normalized radius $r/\langle r \rangle$.

Never referred to!

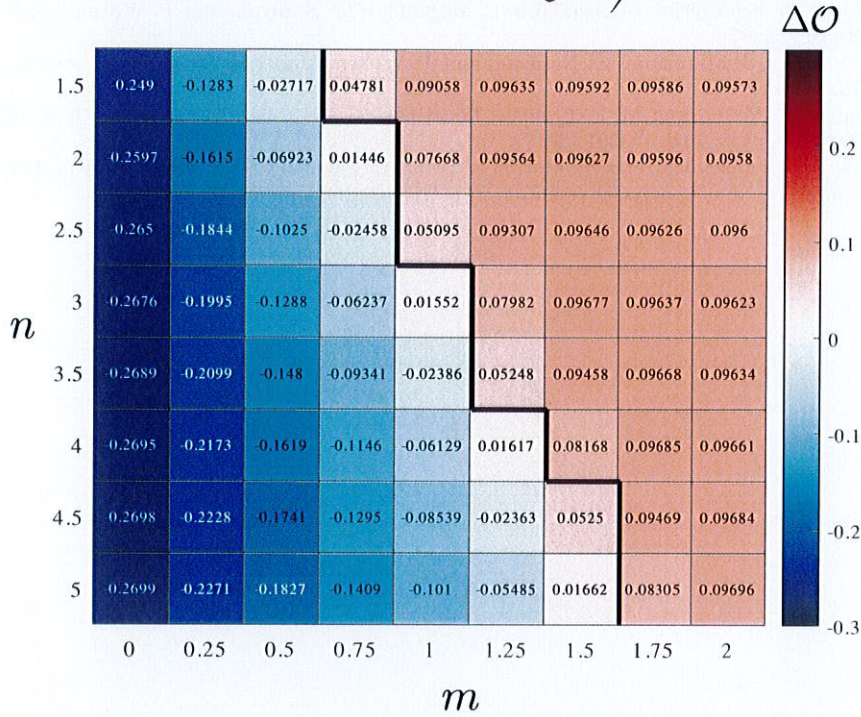
S4. AVERAGE CHANGE IN THE ORDER PARAMETER

In order to quantify the network behavior shown in Fig. 4, we calculate the change in the order parameter for different m, n averaged over 100 simulations with different random initial conditions, and the heat-map results are shown in Fig. S5. The positive or negative change in the order parameter shows the network's change toward homogenization or channelization. The boundary between the two phases (homogenization and channelization) calculated using simple model introduced in the main text is shown with a solid black line here, and it can be seen that it agrees well with the order parameter change.

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main text.



of the main text

FIG. S5. The heat map for the average change in the order parameter corresponding to the network shown in Fig. 4. The order parameter shown in the heat map here is an average of 100 different simulations with different random initialization of tube diameters. The black line shows the boundary between two phases calculated using the model introduced in the main text.

S5. CLOGGING DYNAMICS

Besides erosion, another change in the network is the deposition/sedimentation of material on the boundary walls of the porous material. We refer to this dynamical change a "clogging" process as opposed to erosion. Contrary to erosion, the clogging behavior may cause some edges to block which effectively alters the network of connectivity and network behavior. This change in the connection between nodes through edges getting blocked can drastically alter porous structure behavior, e.g., causes a huge difference between effective and true porosity [5]. Despite the drastic change of network with blockages, we can still focus on the *initial* change in the order parameter. The derivative of order parameter can be written as

$$\frac{dO}{dt} = \sum_{ij} \sum_{kl} \frac{\partial O}{\partial q_{ij}} \frac{\partial q_{ij}}{\partial C_{kl}} \frac{\partial C_{kl}}{\partial t} \quad (S8)$$

where the last term changes sign from erosion to clogging, i.e., $\partial C_{kl} / \partial t = \pm \alpha \pi q_{kl} / r_{kl}^{n-3} \mu_{kl}$ for erosion and clogging respectively. As a result, the magnitude of change in the order parameter equals that of erosion. Note that in Eq.

replace with general formula.

(S8), the second term depends on the network topology, and pore throat clogging results in the change of network topology at later times. At short times, however, similar to the erosion, a phase transition exists at $n = 3$. When $n < 3$ the network moves toward homogenization during the clogging process and when $n > 3$ the flow moves toward the development of channeling instability. At later times, this initial trend, however, might not hold true due to the aforementioned complex changes in the connectivity network during the clogging process.

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