Permeation Instabilities in Active, Polar Gels: Supplemental Material

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This Supplemental Material (SM) provides, in greater detail, the derivation of the dynamic equations and calculations that are relevant to the linear stability analysis. The outline of the SM is as follows. In Sec. I, Equations (2)-(7) of the Letter are derived, using the general framework of non-equilibrium thermodynamics. In Sec II, the equations are written explicitly in terms of the concentration, polarization, and strain variables, and are linearized around the steady state. Next, in Sec. III, we review the parameters that are used in our theory and relate them to convenient lengthscales and timescales. Then, we focus on the linear stability analysis. The stability of the system in the large-q limit is demonstrated in Sec. IV. As part of Sec. V, we detail the approximations used throughout our work. Finally, in Sec. VI, we analyze the linear stability in the passive case.

I. DERIVATION OF THE DYNAMIC EQUATIONS AND FORCE BALANCE EQUATIONS

In this Section we derive the dynamic equations [Eqs. (2)-(3) in the main text] and force-balance equation [Eqs. (4)-(7) in the main text] from the general framework of non-equilibrium thermodynamics. First, the free energy production rate is written in a convenient form, and constitutive relations are written, while respecting Onsager reciprocity. Next, we demonstrate how these equations can yield a two-fluid model.

A. Derivation of the dynamic equations

The time derivative of the free energy is given by (see similar cases in [1, 2])

$$\dot{F} = -\int \mathrm{d}\boldsymbol{r} \left[v_{\alpha\beta}\sigma^{\mathrm{d}}_{\alpha\beta} + h_\alpha \frac{D}{Dt} p_\alpha + H^B_{\alpha\beta}\frac{D}{Dt}B_{\alpha\beta} - J_\alpha \partial_\alpha \bar{\mu} + \Delta\mu r \right], \tag{S1}$$

where $v_{\alpha\beta} = \left(\partial_\alpha v_\beta + \partial_\beta v_\alpha\right)/2$ is the center-of-mass (COM) strain rate and $\sigma^{\mathrm{d}}_{\alpha\beta}$ is the symmetric, deviatoric stress tensor. The solvent orientational field is $h_\alpha = -\delta F/\delta p_\alpha$, while the co-rotational derivative of the polarization is given by $Dp_\alpha/Dt = \left(\partial_t + v_\beta\partial_\beta\right)p_\alpha + \omega_{\alpha\beta}p_\beta$, with $\omega_{\alpha\beta} = \left(\partial_\alpha v_\beta - \partial_\beta v_\alpha\right)/2$ being the COM vorticity tensor. The network molecular field is $H^B_{\alpha\beta} = -\delta F/\delta B_{\alpha\beta}$, while the co-rotational derivative of the strain tensor is $DB_{\alpha\beta}/Dt = \left(\partial_t + v_\gamma\partial_\gamma\right)B_{\alpha\beta} + \omega_{\alpha\gamma}B_{\gamma\beta} + \omega_{\beta\gamma}B_{\gamma\alpha}$. As $B_{\alpha\beta}$ is a symmetric tensor, so is $H^B_{\alpha\beta}$. The relative current between the two components is $J_\alpha = \phi\left(1-\phi\right)\left(v^n_\alpha - v^s_\alpha\right)$, while the relative chemical potential is $\bar{\mu} = \delta F/\delta\phi$. Finally, r is the rate associated with the active consumption of the energy density $\Delta\mu$.

The deviatoric stress tensor is related to the total stress tensor, $\sigma_{\alpha\beta}$, by

$$\sigma^{\mathrm{d}}_{\alpha\beta} = \sigma_{\alpha\beta} - \sigma^a_{\alpha\beta} + \rho\, v_\alpha v_\beta - \sigma^{\mathrm{Er,s}}_{\alpha\beta}, \tag{S2}$$

where $\sigma^a_{\alpha\beta}$ is the antisymmetric part of the total stress and $\rho v_\alpha v_\beta$ is the momentum transfer with ρ being the total mass density. We neglect this term hereafter. The last term in the equation above is the symmetric part of the Ericksen stress. The Ericksen stress tensor, $\sigma^{\mathrm{Er}}_{\alpha\beta}$, is given by [3]

$$\sigma^{\mathrm{Er}}_{\alpha\beta} = \left(f - n_{\mathrm{s}}\mu_{\mathrm{s}} - n_{\mathrm{n}}\mu_{\mathrm{n}}\right)\delta_{\alpha\beta} - \frac{\partial f}{\partial\left(\partial_\beta p_\gamma\right)}\partial_\alpha p_\gamma - \frac{\partial f}{\partial\left(\partial_\beta n_{\mathrm{s}}\right)}\partial_\alpha n_{\mathrm{s}} - \frac{\partial f}{\partial\left(\partial_\beta n_{\mathrm{n}}\right)}\partial_\alpha n_{\mathrm{n}}, \tag{S3}$$

where n_{n} and n_{s} are the network and solvent densities, respectively, and μ_{n} and μ_{s} are their chemical potentials.

It is customary to consider that the solvent and network components are each convected with their own velocity [4]. We, therefore, rewrite the free-energy production rate of Eq. (S1) in terms of a solvent convected derivative, L^{s}_t, and a network convected derivative, L^{n}_t, defined as

$$L^{\mathrm{s}}_t\, p_\alpha = \left(\partial_t + v^{\mathrm{s}}_\beta\partial_\beta\right)p_\alpha + \omega^{\mathrm{s}}_{\alpha\beta}p_\beta + \nu v^{\mathrm{s}}_{\alpha\beta}p_\beta, \tag{S4}$$

$$L^{\mathrm{n}}_t B_{\alpha\beta} = \left(\partial_t + v^{\mathrm{n}}_\gamma\partial_\gamma\right)B_{\alpha\beta} + \omega^{\mathrm{n}}_{\alpha\gamma}B_{\gamma\beta} + \omega^{\mathrm{n}}_{\beta\gamma}B_{\gamma\alpha} + \nu_1\left(v^{\mathrm{n}}_{\alpha\gamma}B_{\gamma\beta} + v^{\mathrm{n}}_{\beta\gamma}B_{\gamma\alpha}\right) + A_{\alpha\beta}, \tag{S5}$$

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where $\omega^{\rm s}_{\alpha\beta}$ and $\omega^{\rm n}_{\alpha\beta}$ are the solvent- and network vorticity tensors, respectively, while $v^{\rm s}_{\alpha\beta}$ and $v^{\rm n}_{\alpha\beta}$ are the solvent- and network strain-rate tensors, respectively. The term ν is the solvent shear-alignment parameter and ν_1 is (minus) the network's slip parameter. The tensor $A_{\alpha\beta}$ accounts for other geometric non-linearities, according to

$$A_{\alpha\beta} = \nu_2 v^{\rm n}_{\gamma\gamma} B_{\alpha\beta} + \nu_3 v^{\rm n}_{\gamma\gamma} B_{\delta\delta}\delta_{\alpha\beta} + \nu_4 B_{\gamma\gamma} v^{\rm n}_{\alpha\beta} + \nu_5 v^{\rm n}_{\gamma\delta} B_{\gamma\delta}\delta_{\alpha\beta}. \tag{S6}$$

Note that other terms are allowed by the polar symmetry, such as those containing contractions between $Q_{\alpha\beta}$ and either $B_{\alpha\beta}$ or $v^{\rm n}_{\alpha\beta}$. For simplicity, we set $\nu = \nu_1 = -1$ and $A_{\alpha\beta} = 0$, such that

$$L^{\rm s}_t\, p_\alpha = \left(\partial_t + v^{\rm s}_\beta \partial_\beta\right) p_\alpha - p_\beta \partial_\beta v^{\rm s}_\alpha, \tag{S7}$$

$$L^{\rm n}_t B_{\alpha\beta} = \left(\partial_t + v^{\rm n}_\gamma \partial_\gamma\right) B_{\alpha\beta} - B_{\gamma\beta}\partial_\gamma v^{\rm n}_\alpha - B_{\gamma\alpha}\partial_\gamma v^{\rm n}_\beta. \tag{S8}$$

The convected derivatives $L^{\rm s}_t$ and $L^{\rm n}_t$ thus reduce to vector and tensor Lie derivatives, respectively [5]. In particular, $L^{\rm n}_t B_{\alpha\beta}$ is the upper-convected derivative [6]. For $\nu_1 = 1$ and $A_{\alpha\beta} = 0$, the network convected derivative reduces to the lower-convected derivative.

Inserting the convected derivatives and using integration by parts, Eq. (S1) transforms into

$$\begin{aligned}\dot{F} = -\int {\rm d}\boldsymbol{r}\, \Big[& v_{\alpha\beta}\delta\sigma_{\alpha\beta} + h_\alpha L^{\rm s}_t\, p_\alpha + H^B_{\alpha\beta} L^{\rm n}_t B_{\alpha\beta} + \Delta\mu r \\ & + J_\alpha \left(-\partial_\alpha \bar{\mu} + \frac{1}{1-\phi}\left(h_\beta \partial_\alpha p_\beta + \partial_\beta\left(h_\alpha p_\beta\right)\right) + \frac{1}{\phi}\left(\partial_\beta \sigma^{\rm el}_{\alpha\beta} - H^B_{\beta\gamma}\partial_\alpha B_{\beta\gamma}\right)\right)\Big].\end{aligned} \tag{S9}$$

In Eq. (S9), we have defined the stress

$$\delta\sigma_{\alpha\beta} = \sigma^{\rm d}_{\alpha\beta} - \frac{1}{2}\left(\sigma^{\rm el}_{\alpha\beta} + \sigma^{\rm el}_{\beta\alpha} - h_\alpha p_\beta - h_\beta p_\alpha\right), \tag{S10}$$

where $\sigma^{\rm el}_{\alpha\beta} = -2H^B_{\alpha\gamma}B_{\gamma\beta}$ is the elastic (Kirchhoff) stress [5] and $-h_\alpha p_\beta$ is the stress associated with polarization rotations. These are the reactive contributions that result from the convected derivatives.

The stress $\delta\sigma_{\alpha\beta}$ includes additional contributions to the deviatoric stress and, namely, the dissipative and active stress. As the network is viscoelastic, dissipation due to network viscosity is already included in the $H^B_{\alpha\beta}L^{\rm n}_t B_{\alpha\beta}$ term, and the viscous contribution to $\delta\sigma_{\alpha\beta}$ originates from the solvent alone. For this reason, it is convenient to rewrite the free-energy production rate in terms of the solvent strain rate, $v^{\rm s}_{\alpha\beta} = \left(\partial_\alpha v^{\rm s}_\beta + \partial_\beta v^{\rm s}_\alpha\right)/2$, as

$$\dot{F} = -\int {\rm d}\boldsymbol{r}\, \left[v^{\rm s}_{\alpha\beta}\delta\sigma_{\alpha\beta} + h_\alpha L^{\rm s}_t\, p_\alpha + H^B_{\alpha\beta} L^{\rm n}_t B_{\alpha\beta} + J_\alpha \delta f_\alpha + \Delta\mu r\right]. \tag{S11}$$

The conjugate of the relative current, δf_α, is a force density, given by

$$\delta f_\alpha = \frac{1}{\phi}\left(\partial_\beta \sigma^{\rm el}_{\alpha\beta} - \phi\partial_\alpha\bar{\mu} - H^B_{\beta\gamma}\partial_\alpha B_{\beta\gamma}\right) - \frac{1}{1-\phi}\left[\partial_\beta\left(\delta\sigma_{\alpha\beta} - h_\alpha p_\beta\right) - h_\beta \partial_\alpha p_\beta\right]. \tag{S12}$$

We interpret it below in Sec I B.

The free-energy production rate of Eq. (S11) is written as an integral over pairs of forces and conjugate fluxes. In each pair, we consider the first variable as the force, and the second as the flux. In particular, the choice of J_α as the force and of δf_α as the flux is in contrast to their physical units. This choice is more convenient, because J_α is more easily measurable and it appears in convected derivatives.

Our aim is to derive constitutive relations between forces and fluxes in a linear theory, close to equilibrium. Fluxes are related to forces with the same signature under time-reversal as their conjugate force, by *dissipative* couplings, and to forces with opposite signatures by *reactive* couplings. Reciprocal dissipative couplings are equal and have a positive contribution to the entropy production, while reciprocal reactive couplings have opposite signs and do not contribute to the entropy production [7].

We consider for the constitutive relations only the leading, zeroth-order terms in a gradient expansion. There are fifteen such coupling terms (number of independent terms in a symmetric 5×5 matrix). One of which, relating r and $\Delta\mu$, does not play any role in the dynamics of the gel. Below, we address the remaining terms and explain how we retain only eight of them. For simplicity, we consider scalar couplings. More complicated tensors, written in terms of $p_\alpha p_\beta$ and $B_{\alpha\beta}$ are generally applicable.

The polarization rate and strain rate are given by

$$L_t^{\mathrm{s}}\, p_\alpha^{(d)} = \frac{1}{\gamma_1} h_\alpha,$$
$$L_t^{\mathrm{s}}\, p_\alpha^{(r)} = \lambda J_\alpha, \tag{S13}$$
$$L_t^{\mathrm{n}} B_{\alpha\beta}^{(d)} = -\frac{1}{\tau}\frac{\partial B_{\alpha\beta}}{\partial \sigma_{\gamma\eta}^{\mathrm{el}}}\sigma_{\gamma\eta}^{\mathrm{el}},$$
$$L_t^{\mathrm{n}} B_{\alpha\beta}^{(r)} = \frac{1}{2}\xi\left(J_\alpha p_\beta + J_\beta p_\alpha\right), \tag{S14}$$

where (d) denotes the dissipative part of the flux and (r) its reactive part. The dissipative, diagonal term in the polarization rate is written in terms of the angular viscosity, γ_1. We have chosen the dissipative couplings to $\Delta\mu$ and $H_{\alpha\beta}^B$ to be zero. Both these terms are, to linear order, of the form $\sim B_{\alpha\beta}p_\beta$ (we disregard a term $\Delta\mu p_\alpha$ that simply renormalizes the parallel orientational field, $h_\parallel$, and has no physical meaning). Such a $B_{\alpha\beta}p_\beta$ term is still possible in the diagonal coupling to the orientational field h_α, due to a free-energy coupling of the form $B_{\alpha\beta}p_\alpha p_\beta$. The same argument explains why we neglect possible terms $\sim h_\alpha p_\beta$ or $\sim \Delta\mu p_\alpha p_\beta$ in the equation for the strain rate. In the biological context of multicellular migration, the latter corresponds to active matrix remodeling . The dissipative term in the strain-rate equation is a relaxation term, written in terms of the elastic stress, $\sigma_{\alpha\beta}^{\mathrm{el}}$, and relaxation time, τ. For the strain energy that we consider in the linear stability, $f_B = \mathrm{Tr}\left(\boldsymbol{B} - \ln \boldsymbol{B}\right)$, the relaxation term reduces to $-\left(B_{\alpha\beta} - \delta_{\alpha\beta}\right)/\tau$.

In both the polarization-rate and strain-rate equations, we neglect a possible reactive coupling to the solvent strain-rate, $v_{\alpha\beta}^{\mathrm{s}}$. This is because we have already made our choice of convective terms in the definitions of the derivatives L_t^{s} and L_t^{n} [Eq. (S7)]. We do consider reactive couplings to the relative current, J_α, which are allowed by the polar symmetry. These are the permeation-alignment and permeation-deformation terms (λ and ξ, respectively).

The fluxes $\delta\sigma_{\alpha\beta}$ and f_α^{rel} are given by

$$\delta\sigma_{\alpha\beta}^{(d)} = 2\eta_s v_{\alpha\beta}^s,$$
$$\delta\sigma_{\alpha\beta}^{(r)} = (1-\phi)\,\zeta\Delta\mu Q_{\alpha\beta}, \tag{S15}$$
$$\delta f_\alpha^{(d)} = \frac{1}{\gamma\phi\,(1-\phi)} J_\alpha,$$
$$\delta f_\alpha^{(r)} = -\left(\lambda h_\alpha + \xi H_{\alpha\beta}^B p_\beta + \nu\Delta\mu p_\alpha\right). \tag{S16}$$

For $\delta\sigma_{\alpha\beta}$, we consider the dissipative viscous stress, in terms of the solvent viscosity, η_{s}, and an active, reactive stress, $(1-\phi)\,\zeta\Delta\mu Q_{\alpha\beta}$, proportional to the solvent concentration. The solvent viscosity also depends on the solvent concentration. However, as the solvent strain rate vanishes in the steady state and appears only as a first-order correction term, this concentration dependence does not play a role in the linear theory. It is not taken into account hereafter. We neglect a possible, dissipative coupling between $\delta\sigma_{\alpha\beta}$ and J_α (or, equivalently, between δf_α and $v_{\alpha\beta}^{\mathrm{s}}$). A coupling between these fields is already included in the definition of δf_α [Eq. (S12)]. For the force δf_α, we consider a dissipative force due to network-solvent friction, written in terms of the mobility γ. The first two reactive terms are determined from the reciprocal, reactive couplings in the polarization rate and strain rate. The final reactive term, $\nu\Delta\mu p_\alpha$, gives rise to an active force.

These constitutive relations describe the dynamics of the strain and polarization fields, as well as the force-balance equation on the gel, using $\partial_\beta\sigma_{\alpha\beta} = 0$. Next, we demonstrate how they can be interpreted as a two fluid model, written in terms of separate force-balance equations for each of the components. Namely, the flux δf_α, conjugate to the relative current, is related to the relative force, f_α^{rel} between the two components.

B. Interpretation in terms of a two-fluid model

We consider the different contributions to the vanishing total force acting on the gel, $\partial_\beta\sigma_{\alpha\beta} = 0$. It includes the force resulting from the Ericksen stress [3],

$$\begin{aligned}\partial_\beta\sigma_{\alpha\beta}^{\mathrm{Er}} &= -n_{\mathrm{s}}\partial_\alpha\mu_{\mathrm{s}} - n_{\mathrm{n}}\partial_\alpha\mu_{\mathrm{n}} - h_\beta\partial_\alpha p_\beta - H_{\beta\gamma}^B\partial_\alpha B_{\beta\gamma}\\ &= -\partial_\alpha\delta P - \phi\partial_\alpha\bar{\mu} - h_\beta\partial_\alpha p_\beta - H_{\beta\gamma}^B\partial_\alpha B_{\beta\gamma},\end{aligned} \tag{S17}$$

where we have made use of the gel incompressibility $n_{\mathrm{s}} + n_{\mathrm{n}} = \rho/m$, with m being the molecular mass (assumed equal for both components), and have denoted $\delta P = \rho\mu_s/m$. For simple solvents and in the absence of elasticity, for which the Ericksen stress reduces to (minus) the osmotic pressure, $\delta P = P - \Pi$ is the difference between total pressure and osmotic pressure. The relative chemical potential is $\bar{\mu} = \rho\left(\mu_{\mathrm{n}} - \mu_{\mathrm{s}}\right)/m$.

The total force is thus given by

$$\begin{aligned}\partial_\beta \sigma_{\alpha\beta} &= \partial_\beta \sigma^{\rm el}_{\alpha\beta} - \phi \partial_\alpha \left(\bar{\mu} + \delta P\right) - H^B_{\beta\gamma} \partial_\alpha B_{\beta\gamma} \\ &\quad + \partial_\beta \left(\delta\sigma_{\alpha\beta} - h_\alpha p_\beta\right) - \left(1-\phi\right) \partial_\alpha \delta P - h_\beta \partial_\alpha p_\beta \\ &= 0. \end{aligned} \tag{S18}$$

The first line in the right-hand-side of the equation above is written in terms of network-dependent physical quantities, and the second line in terms of solvent-dependent ones. They can be interpreted as the force on the network and on the solvent, respectively, not including relative forces between the two components, which do not contribute to the total stress, $\sigma_{\alpha\beta}$.

We define

$$\begin{aligned} f^{\rm n}_\alpha &= \partial_\beta \sigma^{\rm el}_{\alpha\beta} - \phi \partial_\alpha \bar{\mu} - H^B_{\beta\gamma} \partial_\alpha B_{\gamma\beta}, \\ f^{\rm s}_\alpha &= \partial_\beta \left(\delta\sigma_{\alpha\beta} - h_\alpha p_\beta\right) - h_\beta \partial_\alpha p_\beta. \end{aligned} \tag{S19}$$

The force on the network is $f^{\rm n}_\alpha - \phi\, \partial_\alpha \delta P$ and the one on the solvent is $f^{\rm s}_\alpha - \left(1-\phi\right) \partial_\alpha \delta P$. Comparing to Eqs. (S12) and (S18), We find that,

$$\begin{aligned} f^{\rm n}_\alpha + f^{\rm s}_\alpha - \partial_\alpha \delta P &= 0, \\ \frac{1}{\phi} f^{\rm n}_\alpha - \frac{1}{1-\phi} f^{\rm s}_\alpha &= \delta f_\alpha. \end{aligned} \tag{S20}$$

A linear combination of these two equations yields

$$\begin{aligned} f^{\rm n}_\alpha - \phi \partial_\alpha \delta P &= \phi \left(1-\phi\right) \delta f_\alpha, \\ f^{\rm s}_\alpha - \left(1-\phi\right) \partial_\alpha \delta P &= -\phi \left(1-\phi\right) \delta f_\alpha. \end{aligned} \tag{S21}$$

These are the separate force-balance equations for the network and solvent, respectively, in a two-fluid model. It is now possible to identify the relative force between the two components, $\phi\left(1-\phi\right)\delta f_\alpha = f^{\rm rel}_\alpha$. Equation (S21) thus yields Eq. (5) in the main text. This explains the interpretation of the permeation-deformation and permeation-alignment couplings, which appeared originally as phenomenological, polar couplings in Eqs. (S13) and (S14), as relative forces between network and solvent.

In this derivation of the two-fluid model, we have made use of the fact that $\delta\sigma_{\alpha\beta}$ is a stress that originates only in the solvent, as was chosen in our constitutive relations, $\delta\sigma_{\alpha\beta} = 2\eta_{\rm s} v^{\rm s}_{\alpha\beta} + \left(1-\phi\right)\zeta \Delta\mu Q_{\alpha\beta}$. This choice neglects possible network contributions, proportional to $H^B_{\alpha\beta}$ and $\Delta\mu B_{\alpha\beta}$. A two-fluid model can be similarly derived when such network contributions are taken into account, as we demonstrate now.

We write $\delta\sigma_{\alpha\beta} = \delta\sigma^{\rm s}_{\alpha\beta} + \delta\sigma^{\rm n}_{\alpha\beta}$, where $\delta\sigma^{\rm s}_{\alpha\beta}$ is the solvent contribution and $\delta\sigma^{\rm n}_{\alpha\beta}$ is the network contribution of the form $\delta\sigma^{\rm n}_{\alpha\beta} = a H^B_{\alpha\beta} + b\Delta\mu B_{\alpha\beta}$. In this case, while Eq. (S11) still holds, δf_α is related differently to the relative force. Following the same arguments as above, we find that

$$\frac{f^{\rm rel}_\alpha}{\phi\left(1-\phi\right)} = \frac{1}{\phi}\left[\partial_\beta\left(\delta\sigma^{\rm n}_{\alpha\beta} + \sigma^{\rm el}_{\alpha\beta}\right) - \phi\partial_\alpha\bar{\mu} - H^B_{\beta\gamma}\partial_\alpha B_{\beta\gamma}\right] - \frac{1}{1-\phi}\left[\partial_\beta\left(\delta\sigma^{\rm s}_{\alpha\beta} - h_\alpha p_\beta\right) - h_\beta \partial_\alpha p_\beta\right]. \tag{S22}$$

This yields $\phi\left(1-\phi\right)\delta f_\alpha = f^{\rm rel}_\alpha - \partial_\beta \delta\sigma^{\rm n}_{\alpha\beta}$. Inserting this expression in Eq. (S11) yields

$$\dot{F} = -\int {\rm d}\boldsymbol{r} \left[v^{\rm s}_{\alpha\beta}\left(\delta\sigma^{\rm s}_{\alpha\beta} + \delta\sigma^{\rm n}_{\alpha\beta}\right) + h_\alpha L^{\rm s}_t p_\alpha + H^B_{\alpha\beta} L^{\rm n}_t B_{\alpha\beta} + \frac{J_\alpha}{\phi\left(1-\phi\right)}\left(f^{\rm rel}_\alpha - \partial_\beta\delta\sigma^{\rm n}_{\alpha\beta}\right) + \Delta\mu r \right]. \tag{S23}$$

This ensures that any choice of $\delta\sigma^{\rm n}_{\alpha\beta}$ will be incorporated in two constitutive relations, one conjugate to $v^{\rm s}_{\alpha\beta}$ and one conjugate to J_α. In this way, the reciprocal terms will depend consistently on the network strain rate, $v^{\rm n}_{\alpha\beta}$. The resulting two-fluid model is described by Eq. (S21), with the forces,

$$\begin{aligned} f^{\rm n}_\alpha &= \partial_\beta\left(\delta\sigma^{\rm n}_{\alpha\beta} + \sigma^{\rm el}_{\alpha\beta}\right) - \phi\partial_\alpha\bar{\mu} - H^B_{\beta\gamma}\partial_\alpha B_{\gamma\beta}, \\ f^{\rm s}_\alpha &= \partial_\beta\left(\delta\sigma^{\rm s}_{\alpha\beta} - h_\alpha p_\beta\right) - h_\beta\partial_\alpha p_\beta. \end{aligned} \tag{S24}$$

II. LINEARIZED VERSION OF THE EQUATIONS

In this section we derive the linearized version of the equations, which is used for the linear stability analysis. We first write the equations in full form, including explicit expressions for the fields that are derived from the free energy. Then, we solve the steady-state equations, and linearize around the steady-state solutions.

A. Full form

We consider the free energy of Eq. (1) in the Letter,

$$F=\int dr\left[f_\phi+\frac{1}{2}G\phi\mathrm{Tr}\left(\boldsymbol{B}-\ln\boldsymbol{B}\right)+\left(1-\phi\right)^2\left(\frac{1}{2}K\left(\nabla\boldsymbol{p}\right)^2+K_d\nabla\cdot\boldsymbol{p}\right)-\frac{1}{2}h_\parallel\boldsymbol{p}^2\right]. \tag{S25}$$

Here, we have inserted the Flory Gaussian-chain free-energy density, $f_B=G\phi\mathrm{Tr}\left(\boldsymbol{B}-\ln\boldsymbol{B}\right)/2$ and have neglected the possible strain-polarization coupling, $f_{Bp}=0$. The resulting solvent orientational field is

$$h_\alpha=-\frac{\delta F}{\delta p_\alpha}=\partial_\beta\left[\left(1-\phi\right)^2K\partial_\beta p_\alpha\right]-2K_d\left(1-\phi\right)\partial_\alpha\phi+h_\parallel p_\alpha. \tag{S26}$$

Note that the polar-splay contribution to the orientational field is via the concentration gradient. Otherwise, this term can be integrated in the free energy to a boundary term (divergence theorem), which does not contribute to the orientational field. The network molecular field is given by

$$H^B_{\alpha\beta}=-\frac{\delta F}{\delta B_{\alpha\beta}}=-\frac{1}{2}G\phi\left(\delta_{\alpha\beta}-B^{-1}_{\alpha\beta}\right), \tag{S27}$$

and the elastic stress is

$$\sigma^{\mathrm{el}}_{\alpha\beta}=-2H^B_{\alpha\gamma}B_{\gamma\beta}=G\phi\left(B_{\alpha\beta}-\delta_{\alpha\beta}\right). \tag{S28}$$

The relative chemical potential reads

$$\bar{\mu}=\frac{\delta F}{\delta\phi}=\bar{\mu}_0\left(\phi\right)+\frac{1}{2}G\mathrm{Tr}\left(\boldsymbol{B}-\ln\boldsymbol{B}\right)-2\left(1-\phi\right)\left(\frac{1}{2}K\left(\nabla\boldsymbol{p}\right)^2+K_d\nabla\cdot\boldsymbol{p}\right), \tag{S29}$$

where $\bar{\mu}_0$ is derived from the mixing term, $\bar{\mu}_0=\delta f_\phi/\delta\phi-\partial_\alpha\delta f_\phi/\delta\partial_\alpha\phi$. Equations (S26)-(S29) relate the fields that appear in the forces and fluxes of the entropy production to the dynamic fields, ϕ, p_α and $B_{\alpha\beta}$, whose dynamics we analyze to linear order.

The concentration fields satisfies the continuity equation,

$$\partial_t\phi+\partial_\alpha\left(\phi v^n_\alpha\right)=0. \tag{S30}$$

The polarization and strain fields evolve according to Eqs. (2) and (3) in the main text of the Letter, and the velocities are related to the polarization, strain and concentration from the force-balance equations on the solvent and network [Eqs. (4)-(7) in the main text].

B. Steady state

We search for a steady-state with homogeneous fields $\phi=\phi_0$, $p_\alpha=p^0_\alpha=\delta_{\alpha1}$, $B_{\alpha\beta}=B^0_{\alpha\beta}$, a zero COM velocity, $v_\alpha=0$, and a homogeneous relative current, $J^0_\alpha=J_0p^0_\alpha$. As the system is homogeneous, all the gradient terms vanish, including the convective terms, and forces acting on the network and solvent. The relative current is determined from $f^{rel}_\alpha=0$.

The relative force depends on the parallel solvent orientational field, $h_\parallel$. This field is a Lagrange multiplier that ensures $p^2=1$. Its value is determined by projecting the polarization rate equation on the polarization. We find that $h_\parallel=-\gamma_1\lambda J_0$. In addition, the steady-state strain is given by $B^0_{\alpha\beta}=\delta_{\alpha\beta}+\xi\tau J_0p^0_\alpha p^0_\beta$. As $B_{\alpha\beta}$ is positive definite, this steady state is possible only for $\xi\tau J_0>-1$. The resulting molecular field is $H^{B\,(0)}_{\alpha\beta}=-G\phi_0\xi\tau J_0\left(1+\xi\tau J_0\right)^{-1}p^0_\alpha p^0_\beta/2$.

The equation $f^{rel}_\alpha p^0_\alpha=0$ then reduces to

$$\frac{1}{\gamma}J_0-\phi_0\left(1-\phi_0\right)\left(-\lambda^2\gamma_1J_0-\frac{1}{2}G\phi_0\xi^2\frac{\tau J_0}{1+\xi\tau J_0}+\nu\Delta\mu\right)=0. \tag{S31}$$

This is generally a quadratic equation in J_0. As our framework is formulated to linear order in J_0, we consider small $\xi\tau J_0$ and retain only linear terms. This yields the relative current,

$$J_0=\tilde{\gamma}\phi_0\left(1-\phi_0\right)\nu\Delta\mu, \tag{S32}$$

with the renormalized mobility

$$\tilde{\gamma}=\frac{\gamma}{1+\gamma\phi_0\left(1-\phi_0\right)\left(\lambda^2\gamma_1+\frac{1}{2}\eta_n\xi^2\right)}, \tag{S33}$$

where $\eta_n=G\phi_0\tau$ is the network viscosity. The mobility is effectively decreased, due to the new, polar, relative forces between the solvent and network. For simplicity, we consider in our work only linear terms in the new coupling terms, and neglect, $\gamma\phi_0\left(1-\phi_0\right)\left(\lambda^2\gamma_1+\frac{1}{2}\eta_n\xi^2\right)\ll1$. In this case, the mobility retains its original value, $\tilde{\gamma}\simeq\gamma$ and

$J_0 = \gamma\phi_0 (1-\phi_0)\,\nu\Delta\mu$. This approximation is re-examined in Sec. V of the SM.

C. Linearized equations

The stability is studied by introducing a small perturbation in the fields at point $\boldsymbol{r}$ and time t with a wave vector $\boldsymbol{q} = q\hat{y}$ and growth rate s. For simplicity, we focus on a 2-dimensional system,

$$\begin{aligned}
\phi &= \phi_0 + \phi^1 \exp(iqy + st)\,,\\
p_\alpha &= \delta_{\alpha 1} + p^1\delta_{\alpha 2}\exp(iqy+st)\,,\\
B_{xx} &= 1 + \xi\tau J_0 + B^1_{xx}\exp(iqy+st)\,,\\
B_{xy} &= B^1_{xy}\exp(iqy+st)\,,\\
B_{yy} &= 1 + B^1_{yy}\exp(iqy+st)\,.
\end{aligned} \tag{S34}$$

In particular, the correction to the polarization is in the y-direction to preserve $p^2 = 1$ to linear order. The amplitudes (denoted with superscript 1) are assumed small, and the equations are linearized in these amplitudes. To this linear order, we consider the following form of the mixing chemical potential,

$$\bar{\mu}^1_0 = \chi^{-1}\left(1 + l_\phi^2 q^2\right)\phi^1, \tag{S35}$$

where $\chi^{-1} = \delta^2 f_\phi/\delta\phi^2$ is the inverse osmotic compressibility, and l_ϕ is a Ginzburg-Landau type correlation length, which suppresses large-q concentration fluctuations.

The COM velocity and relative current are similarly expanded to linear order, $v_\alpha = v^1_\alpha \exp(iqy+st)$ and $J_\alpha = J_0 p^0_\alpha + J^1_\alpha \exp(iqy+st)$, together with the pressure difference, $\delta P = \delta P^1 \exp(iqy+st)$, where the reference pressure was conveniently taken to be zero. Incompressibility yields $v^1_2 = 0$. The pressure can then be determined from the y-component of the force-balance equation on the entire gel,

$$\delta P^1 = -2i\eta_s q\frac{J^1_y}{1-\phi_0} + \frac{1}{2}\zeta\Delta\mu\phi^1 + \phi_0 G B^1_{yy} - \phi_0\left[-2iK_d(1-\phi_0)\,qp^1 + \frac{1+l_\phi^2q^2}{\chi}\phi^1\right]. \tag{S36}$$

The first term on the RHS originates from the solvent viscosity, the second term is the compressional, active stress in the y-direction due to concentration variations, the third term is the compressional elastic stress in the y-direction, and the last two terms are components of the osmotic contribution to the pressure.

The components for the relative current can be found from the network force-balance equation. The x-component is simpler, because it does not involve pressure terms. We find that

$$\gamma^{-1}J^1_x = iqG\phi_0 B^1_{xy} + \nu\Delta\mu(1-2\phi_0)\,\phi^1 - \frac{1}{2}\xi G\phi_0^2(1-\phi_0)\,B^1_{xx}. \tag{S37}$$

The first term on the RHS is the force exerted by the shear strain. The second term is the correction to the x-component of the active, relative force, due to concentration fluctuations. The third term is the force due to the permeation-deformation mechanism. The permeation-alignment mechanism vanishes to linear order in λ.

The y-component of the relative current includes the contribution coming from the pressure. We find that

$$\begin{aligned}
\frac{1}{\gamma\phi_0(1-\phi_0)}\left[1 + 2\gamma\eta_s\frac{\phi_0}{1-\phi_0}q^2\right]J^1_y = \lambda\left[-(1-\phi_0)^2Kq^2p^1 - 2K_d(1-\phi_0)\,iq\phi^1\right] - \frac{1}{2}\xi G\phi_0 B^1_{xy} + \nu\Delta\mu p^1\\
+ iq\left(GB^1_{yy} - \left[-2iK_d(1-\phi_0)\,qp^1 + \frac{1+l_\phi^2q^2}{\chi}\phi^1\right] - \frac{1}{2(1-\phi_0)}\zeta\Delta\mu\phi^1\right),
\end{aligned} \tag{S38}$$

The coefficient on the LHS originates from the viscous term in the pressure. The first line of the RHS are the relative forces, including the contributions from permeation alignment ($\sim\lambda$), permeation deformation ($\sim\xi$) and active force ($\sim\nu\Delta\mu$). The second line of the RHS includes the divergence of the weighted difference between the network stress and solvent stress [compare with the expression for δf_α in Eq. (S12)].

The x-component of the solvent velocity is found from the x-component of the force-balance equation on the entire gel, which amounts to equating the shear stress to zero. This yields

$$v^{s(1)}_x = \frac{i}{\eta_s q}\left[G\phi_0 B^1_{xy} + (1-\phi_0)\,\zeta\Delta\mu p^1 + \gamma_1\lambda J_0 p^1\right], \tag{S39}$$

where the first term on RHS is the elastic shear stress, the second term is the active shear stress to linear order, and the last term is the stress associated with polarization rotation to linear order.

Substituting the above results for the velocities, as well as the expressions for the chemical potential and orientational

fields, we obtain linear equations in terms of only the dynamic fields. These equations are

$$
\begin{aligned}
0 &= \left(s + \frac{q^2}{1+l_\eta^2 q^2}\left(\frac{l^2}{\tau_\phi}\left(1+l^2q^2\right)+\frac{l_\eta^2}{4\tau_a}\right)+\frac{2\lambda l_p l_{\phi p}^2 q^2}{\tau_p\left(1-\phi_0\right)\left(1+l_\eta^2q^2\right)}\right)\phi^1 \\
&+\left(\frac{iJ_0 q}{1+l_\eta^2q^2}-\frac{2il_p l_{\phi p}^2 q^3}{\tau_p\left(1-\phi_0\right)\left(1+l_\eta^2q^2\right)}-\frac{i\lambda l_p^2 l_{\phi p}^2 q^3}{\tau_p\left(1+l_\eta^2q^2\right)}\right)p^1-\frac{\left(1-\phi_0\right)^2\phi_0 l_B^2}{\tau\left(1+l_\eta^2q^2\right)}\left(i\frac{\phi_0}{2}\xi q B_{xy}^1+q^2B_{yy}^1\right), \\
0 &= \left(i\lambda\frac{q}{1+l_\eta^2q^2}\left(\frac{l^2}{\tau_\phi}\left(1+l^2q^2\right)+\frac{l_\eta^2}{4\tau_a}\right)+\frac{2il_pq}{\tau_p\left(1-\phi_0\right)}\right)\phi^1 \\
&+\left(s+\frac{l_p^2q^2}{\tau_p}+\frac{\lambda J_0 l_\eta^2 q^2}{1+l_\eta^2q^2}+\frac{2\lambda l_p l_{\phi p}^2 q^2}{\tau_p\left(1-\phi_0\right)\left(1+l_\eta^2q^2\right)}\right)p^1-i\lambda\frac{\left(1-\phi_0\right)^2\phi_0 l_B^2}{\tau\left(1+l_\eta^2q^2\right)}qB_{yy}^1, \\
0 &= -\xi\frac{1-2\phi_0}{\phi_0\left(1-\phi_0\right)}J_0\phi^1+\left(s+\frac{1}{\tau}\right)B_{xx}^1-\frac{i\phi_0\left(1-\phi_0\right)\xi l_B^2 q}{\tau}B_{xy}^1, \\
0 &= \frac{i}{2}\xi\frac{q}{1+l_\eta^2q^2}\left(\frac{l^2}{\tau_\phi}\left(1+l^2q^2\right)+\frac{l_\eta^2}{4\tau_a}\right)\phi^1+\left(\frac{2\lambda l_{\phi p}^2 J_0}{l_\eta^2\left(1-\phi_0\right)^2}-\frac{\xi J_0}{2}\frac{2+l_\eta^2q^2}{1+l_\eta^2q^2}+\frac{\xi l_p l_{\phi p}^2 q^2}{\tau_p\left(1-\phi_0\right)\left(1+l_\eta^2q^2\right)}+\frac{1}{\tau_a}\right)p^1 \\
&+\left[s+\frac{1}{\tau}\left(1+2\phi_0^2\frac{l_B^2}{l_\eta^2}\right)+\frac{l_B^2q^2}{\tau}\right]B_{xy}^1+i\frac{l_B^2}{2\tau}\xi q\phi_0\left(1-\phi_0\right)\left(B_{xx}^1-\frac{1-\phi_0}{1+l_\eta^2q^2}B_{yy}^1\right), \\
0 &= -\left(\frac{4\lambda l_p l_{\phi p}^2 q^2}{\phi_0\left(1-\phi_0\right)\tau_p\left(1+l_\eta^2q^2\right)}+\frac{2}{\phi_0}\frac{q^2}{1+l_\eta^2q^2}\left(\frac{l^2}{\tau_\phi}\left(1+l^2q^2\right)+\frac{l_\eta^2}{4\tau_a}\right)\right)\phi^1 \\
&+\left(\frac{2i\lambda l_p^2 l_{\phi p}^2 q^3}{\tau_p\phi_0\left(1+l_\eta^2q^2\right)}-\frac{2iJ_0q}{\phi_0\left(1+l_\eta^2q^2\right)}+\frac{4il_p l_{\phi p}^2 q^3}{\tau_p\phi_0\left(1-\phi_0\right)\left(1+l_\eta^2q^2\right)}\right)p^1 \\
&+\frac{i\xi l_B^2 q}{\tau\left(1+l_\eta^2q^2\right)}\phi_0\left(1-\phi_0\right)^2B_{xy}^1+\left[s+\frac{1}{\tau}+\frac{2l_B^2q^2}{\tau}\frac{\left(1-\phi_0\right)^2}{1+l_\eta^2q^2}\right]B_{yy}^1.
\end{aligned}
\tag{S40}
$$

They describe the time evolution of the fields ϕ^1, p^1, B_{xx}^1, B_{xy}^1, and B_{yy}^1, respectively. For the sake of brevity, these equations are written in terms of several lengthscales and timescales that are defined below in Sec. III. The equations can be written in matrix form as, $\boldsymbol{M}\cdot\boldsymbol{x}=0$, where $\boldsymbol{x}=\left(\phi^1,p^1,B_{xx}^1,B_{xy}^1,B_{yy}^1\right)^T$ is a vector of the perturbed fields, and $\boldsymbol{M}$ is the dynamic matrix. Within the framework of linear stability, the dispersion relations $s(\boldsymbol{q})$ are obtained by solving $\det\boldsymbol{M}=0$, which is a fifth-order polynomial in s. The system is stable if $\mathrm{Re}\,s<0$ for all the eigenvalues.

III. PARAMETERS OF THE THEORY AND NON-DIMENSIONALIZATION

Our theory includes 14 parameters: ϕ_0, l_ϕ, χ, G, K_d, K, τ, γ, γ_1, η_s, ξ, λ, $\nu\Delta\mu$, and $\zeta\Delta\mu$. These parameters have units that combine length, time, and energy. Therefore, from the Buckingham-Pi theorem, there are 11 independent, dimensionless parameters. It is convenient to introduce the active, relative current, $J_0=\gamma\phi_0\left(1-\phi_0\right)\nu\Delta\mu$, and 12 parameters in units of length and time.

These length scales are l_ϕ, $l_p=K/K_d$, $1/\xi$, $1/\lambda$, $l_B=\sqrt{D_B\tau}=\sqrt{G\gamma\tau/\left(1-\phi_0\right)}$, $l_{\phi p}=\sqrt{\gamma_1\gamma\phi_0\left(1-\phi_0\right)}$ and $l_\eta=\sqrt{2\gamma\eta_s\phi_0/\left(1-\phi_0\right)}$. l_η is proportional to the length over which dissipation due to network-solvent friction matches the dissipation due to the solvent viscosity. For poroelastic materials, l_η is proportional to the mesh size. $l_{\phi p}$ is proportional to the length over dissipation due to network-solvent friction matches the dissipation due to the polar rotational viscosity.

The time scales are τ, $\tau_p=l_p^2/D_p=K\gamma_1/\left[K_d\left(1-\phi_0\right)\right]^2$, $\tau_\phi=l_\phi^2/D_\phi=\chi l_\phi^2/\left[\gamma\phi_0\left(1-\phi_0\right)\right]$, and $\tau_a=\eta_s/\left[\left(1-\phi_0\right)\zeta\Delta\mu\right]$. The latter is an active time scale, that describes the rate in which the solvent need to be sheared, so that the viscous stress matches the active stress.

Among the above parameters, some can be negative. These are l_p, ξ, λ, J_0 (polar terms), and τ_a. It is possible to rescale the lengths by l_ϕ and times by τ_ϕ. This yields, together with ϕ_0, the 11 desired dimensionless parameters.

The large number of parameters makes the analysis of the system challenging. This is why we have made several approximations (see Sec. V) and have focused on novel permeation instabilities that arise to linear order in the polar terms J_0, ξ, and λ. Further analysis beyond the scope of our calculation is reserved for future studies. Furthermore, the 14 above-mentioned quantities are already a reduced number of system parameters. Other relevant quantities are, for example: additional elastic moduli (corresponding to other Poisson's ratios and possible nonlinearities),

bulk viscoelastic relaxation time and bulk viscosity (taken here as equal to the corresponding shear values), and configuration-dependent friction coefficients (e.g., $\gamma_{\alpha\beta} = \gamma\delta_{\alpha\beta} + \gamma_Q Q_{\alpha\beta}$). We did not retain such quantities for the purposes of our generic, physical theory. They may be relevant for a quantitative analysis of experiments.

IV. LARGE-WAVENUMBER STABILITY

While our hydrodynamic theory is valid for small q values, we test whether our dynamic variables are stable for large wavenumbers. We expand the coefficients of the determinant, $\det\boldsymbol{M}$, to highest order in q, and find that

$$s^5 + a_2 q^2 s^4 + a_4 q^4 s^3 + a_6 q^6 \left(s^2 + 2\frac{s}{\tau} + \frac{1}{\tau^2}\right) = 0, \tag{S41}$$

with the coefficients

$$\begin{aligned} a_2 &= \left(\frac{l_\phi^4}{l_\eta^2 \tau_\phi} + \frac{l_p^2}{\tau_p} + \frac{l_B^2}{\tau}\frac{1}{1+\lambda^2 l_{\phi p}^2}\right), \\ a_4 &= \frac{l_\phi^4}{l_\eta^2 \tau_\phi}\frac{l_p^2}{\tau_p} + \left(\frac{l_\phi^4}{l_\eta^2 \tau_\phi} + \frac{l_p^2}{\tau_p}\right)\frac{l_B^2}{\tau}\frac{1}{1+\lambda^2 l_{\phi p}^2}, \\ a_6 &= \frac{1}{\tau \tau_p \tau_\phi}\frac{1}{1+\lambda^2 l_{\phi p}^2}\frac{l_\phi^4 l_p^2 l_B^2}{l_\eta^2}. \end{aligned} \tag{S42}$$

There are two possible types of solutions. First, a relaxation solution $s = s_0$. In this case, only the parenthesis multiplying q^6 is required to vanish. These are the elastic relaxations, $s_{1,2} = -1/\tau$ of the two compressional strains, B_{xx} and B_{yy}. This result is related to the fact that we consider osmotic diffusion as the sole origin of strain diffusion. In this limit, B_{xx} has a vanishing diffusion coefficient for wave vectors in the y-direction. The diffusion coefficient of B_{yy}, on the other hand, decays to zero, because the effective mobility coefficient, γ, in the y-direction scales as $1/q^2$ in this limit, due to solvent compressibility (see Sec. VI).

Another type of solution is diffusion, $s = -Dq^2$. Inserting this solution in the equation, we find that the highest-order terms scale as q^{10} and D solves the equation $D^3 - a_2 D^2 + a_4 D - a_6 = 0$. The three solutions are given by $D = l_p^2/\tau_p$, corresponding to rotational diffusion, $D = l_B^2/\left[\left(1+\lambda^2 l_{\phi p}^2\right)\tau\right]$, corresponding to shear strain diffusion, and $D = l_\phi^4/\left(l_\eta^2 \tau_\phi\right)$. The latter diffusion constant is related to the concentration ϕ^1, but is different than the small-q diffusion coefficient, l_ϕ^2/τ_ϕ. The diffusion in this case is not osmotic, but describes mass-conserving compression/dilations in the y-direction. The local concentration is then given by $\phi_0 + \phi^1 = 1/v' \simeq \phi_0\left(1 - B_{yy}^1/2\right)$, where v' is the volume of a network element after a B_{yy}^1 deformation.

All the growth rates are negative, meaning that the system is stable for large q values. We emphasize that the theory is hydrodynamic. It was derived from a gradient expansion, and is adequate for small wave vectors. Terms that we have neglected may become important in the large-q limit. This Section serves only to demonstrate that our theory is consistent and does not result in large-q instabilities. We make no further predictions in this limit.

V. VALIDITY

The theory depends, as was explained in Sec. III, on 11 dimensionless parameters. For the sake of simplicity, we perform the analysis to linear order in the permeation-deformation and permeation-alignment parameters (ξ and λ, respectively). This implies a lengthscale l, such that ξl and λl are considered to be small. For example, the steady-state relative current, $J_0 = \gamma\phi_0\left(1-\phi_0\right)\nu\Delta\mu$ was derived assuming that $\gamma\phi_0\left(1-\phi_0\right)\left(\lambda^2\gamma_1 + \frac{1}{2}\eta_n\xi^2\right) \ll 1$. In terms of the lengthscales that we have introduced in Sec. III, this condition can be written as

$$\left(\lambda l_{\phi p}\right)^2 + \frac{1}{2}\left[\phi_0\left(1-\phi_0\right)\xi l_B\right]^2 \ll 1. \tag{S43}$$

Below we review similar conditions that were used as part of our calculation.

A. Permeation deformation ($\xi \neq 0$)

The eigenvector associated with the concentration and polarization variables, which is a linear combination of them for $q = 0$, has a dispersion relation $s = iuq - Dq^2$. The velocity, u, to quadratic order in ξ, is given by

$$u = \sqrt{\frac{2}{1-\phi_0}\frac{l_p}{\tau_p}\left(J_0 + J_\xi\left[1 - J_0\xi\left(\tau+\tau_a\right)\right]\right)}, \tag{S44}$$

where

$$J_\xi = \frac{1}{2}\frac{\xi}{\tau_a}\left[\phi_0\left(1-\phi_0\right)\right]^2\frac{l_B^2}{1+2\phi^2 l_B^2/l_\eta^2}. \tag{S45}$$

Therefore, we have considered the limit where $|J_0\xi\left(\tau+\tau_a\right)| \ll 1$. The diffusion coefficient, D, to quadratic order in ξ, reads

$$D = \frac{1}{2}\left(D_p + D_\phi + D_\zeta\right) + \frac{1}{2}D_\xi\left[1 + \xi\left(\frac{1-\phi_0}{4}\frac{\tau_a}{\tau}\frac{\tau_p}{l_p}\left(\frac{l_\phi^2}{\tau_\phi} + \frac{l_\eta^2}{4\tau_a}\right)\left(1 + 2\phi_0^2\frac{l_B^2}{l_\eta^2}\right) - J_0\left(\tau+\tau_a\right)\right)\right]. \tag{S46}$$

Therefore, in addition to our previous condition, we require here that

$$\left|\xi\frac{\tau_a}{\tau}\frac{\tau_p}{l_p}\left(\frac{l_\phi^2}{\tau_\phi} + \frac{l_\eta^2}{4\tau_a}\right)\right| \ll 1. \tag{S47}$$

The eigenvector associated with the shear-strain variable has a dispersion relation $s = s_0 - Dq^2$. The relaxation time, up to quadratic order in ξ is given by

$$s_0 = -\frac{1}{\tau}\left[1 + \frac{\eta_n}{\eta_s} + \left(\frac{1}{2}\phi_0\left(1-\phi_0\right)l_B\xi\right)^2\right]. \tag{S48}$$

We have thus considered $\phi_0\left(1-\phi_0\right)l_B\xi \ll 1$ (as was required for the steady-state relative current). Note, however, that this new term demonstrates a new possible mechanism for elastic relaxation through a feedback between the shear strain and relative current.

The diffusion coefficient, up to quadratic order in ξ, is

$$D = D_B - D_\xi\left(1 - J_0\xi\left(\tau+\tau_a\right) + \xi\frac{\tau_p}{l_p}\frac{\tau_a}{\tau^2}\left(1 + 2\phi_0^2\frac{l_B^2}{l_\eta^2}\right)\left[\frac{1}{2}\left(\frac{1-\phi_0}{\phi_0}\right)^2 l_\eta^2\left(1 + 2\phi_0\frac{l_B^2}{l_\eta^2}\right) + \left(1-\phi_0\right)\frac{\tau}{4}\left(\frac{l_\phi^2}{\tau_\phi} + \frac{l_\eta^2}{4\tau_a}\right)\right]\right). \tag{S49}$$

Therefore, in addition to our previous conditions, we consider here that

$$\frac{1}{2}\left(\frac{1-\phi_0}{\phi_0}\right)^2\left|\frac{\xi l_\eta^2}{l_p}\frac{\tau_a\tau_p}{\tau^2}\right| \ll 1. \tag{S50}$$

The permeation instabilities that we report in the Letter are allowed by these conditions. They can be satisfied with sufficiently large active stresses, corresponding to small τ_a and large J_ξ and D_ξ in absolute values.

B. Permeation alignment ($\lambda \neq 0$)

We examine the corrections to our results in the presence of the permeation-alignment coupling, due to higher-order terms in the parameter, λ. We find that the eigenvector associated with the polarization has a constant growth rate ($\sim q^0$), which is cubic in λ, $s_0 = -\lambda^3 l_{\phi p}^2 J_0$. In particular, this leads to an instability for $\lambda J_0 < 0$. We consider this term to be negligible, $s_0\tau \ll 1$. In this case, as is presented in the main text, the eigenvector associated with polarization and concentration has a growth rate $s = iuq - Dq^2$. The velocity, u, is unchanged in this limit. The diffusion coefficient, however, has a correction

$$D = \frac{1}{2}\left[\left(1 + \frac{1}{2}\lambda^2 l_{\phi p}^2\right)D_p + D_\phi + D_\zeta + D_\lambda\right]. \tag{S51}$$

This correction increases the effective angular diffusion, and can only be stabilizing. It is negligible for $\lambda l_{\phi p} \ll 1$ (as was required for the steady-state relative current).

This condition also allows for finite $\lambda J_0\tau$, as is required for the possible strain instability in the main text, with negligible $s_0\tau$. The polarization-concentration instability is also allowed by this condition. The velocity u can become imaginary depending on the sign of $l_p J_0$ and λJ_0, and the diffusion coefficient can become negative, for example, for

sufficiently large elastic modulus, $G\chi \gg 1$, such that D_λ is large compared to D_ϕ in absolute value. Finally, the diffusion coefficient of B_{yy} has a correction of order $s_0\tau$, which is negligible within our limits.

VI. PASSIVE INSTABILITY

In the main text, we have presented active permeation instabilities that occur in the presence of new polar coupling terms. It is worth noting that a passive, polar gel can also become unstable, even in the absence of the permeation-alignment and permeation-deformation mechanisms [8, 9]. This instability originates from the coupling between the polarization and concentration. Its eigenmode is a linear combination of the concentration and polarization for $q = 0$. Below, we derive the criterion for this instability from the dynamic equations. This derivation allows to analyze the effects of the new permeation couplings that are allowed even in the absence of activity, $\Delta\mu = 0$. We demonstrate that the instability criterion is unaffected by the new permeation couplings.

A. Passive instability for $\lambda = \xi = 0$

As before, we linearize around the homogeneous steady state in the passive case (with $J_0 = 0$), for a wave-vector that is perpendicular to the original polarization, and write the equations in Fourier space. First, we ignore the solvent viscosity and network elasticity. The equations for the polarization and concentration are

$$\begin{aligned} sp^1 &= \frac{1}{\gamma_1}h_2 = -\frac{1}{\gamma_1}\left[(1-\phi_0)^2 Kq^2p^1 + 2K_d(1-\phi_0)iq\phi^1\right], \\ s\phi^1 &= -iqJ_y^1 = -\gamma\phi_0(1-\phi_0)q^2\left[\chi^{-1}\left(1+l_\phi^2q^2\right)\phi^1 - 2i(1-\phi_0)K_dqp^1\right]. \end{aligned} \tag{S52}$$

In terms of the lengthscales and time scales that we have introduced above, these equations are given by

$$\begin{aligned} sp^1 &= -\frac{1}{\tau_p}\left(l_p^2q^2p^1 + \frac{2i}{1-\phi_0}l_pq\phi^1\right), \\ s\phi^1 &= -q^2\left[\frac{l_\phi^2}{\tau_\phi}\left(1+l_\phi^2q^2\right)\phi^1 - \frac{2i}{1-\phi_0}l_{\phi p}^2\frac{l_p}{\tau_p}qp^1\right]. \end{aligned} \tag{S53}$$

This set of linear equations has a non-trivial solution when the determinant of coefficients vanishes,

$$s^2 + \left[\frac{l_p^2}{\tau_p} + \frac{l_\phi^2}{\tau_\phi}\left(1+l_\phi^2q^2\right)\right]q^2s + \frac{l_p^2}{\tau_p}\left[\frac{l_\phi^2}{\tau_\phi}\left(1+l_\phi^2q^2\right) - \left(\frac{2}{1-\phi_0}\right)^2\frac{l_{\phi p}^2}{\tau_p}\right]q^4 = 0. \tag{S54}$$

The linear term in s is always positive. The solution has a positive real part only when the coefficient independent of s is negative. An instability occurs for

$$\left(\frac{2}{1-\phi_0}\right)^2\frac{l_{\phi p}^2}{\tau_p} > \frac{l_\phi^2}{\tau_\phi}\left(1+l_\phi^2q^2\right). \tag{S55}$$

This is equivalent to $K < 4K_d^2\chi/\left(1+l_\phi^2q^2\right)$. This mechanism involves only diagonal transport coefficients and originates only from the concentration-polarization coupling in the free energy.

We now consider the effects of the solvent viscosity and network elasticity. Adding the solvent viscosity merely renormalizes the friction according to $1/\gamma \to \left(1+l_\eta^2q^2\right)/\gamma$ [see Eq. (S38)]. Network elasticity introduces a network force due to the elongation stress in the y-direction, $iqG\phi_0B_{yy}^1$. The strain variable, because of the convected derivative, evolves as

$$\left(s+\frac{1}{\tau}\right)B_{yy}^1 = \frac{2}{\phi_0}iqJ_y^1. \tag{S56}$$

The new equation for the current is, therefore,

$$\frac{1+\left[l_\eta^2 + 2(1-\phi_0)^2\frac{l_B^2}{1+s\tau}\right]q^2}{\gamma\phi_0(1-\phi_0)}J_y^1 = -iq\left[\frac{l_\phi^2}{\tau_\phi}\left(1+l_\phi^2q^2\right)\phi^1 - \frac{2i}{1-\phi_0}l_{\phi p}^2\frac{l_p}{\tau_p}qp^1\right] \tag{S57}$$

The elasticity can be considered as an s-dependent correction to the friction coefficient. Note that, assuming an instability ($s > 0$), the new friction coefficient remains positive, and our previous analysis holds with a renormalized γ. As the instability criterion is independent of γ, it is still given by $K < 4K_d^2\chi/\left(1+l_\phi^2q^2\right)$.

B. Passive instability with permeation alignment ($\lambda \neq 0$)

The permeation alignment induces a relative force between the network and solvent, which modifies the relative current,

$$J_y^1 = -iq\left[\frac{l_\phi^2}{\tau_\phi}\left(1+l_\phi^2 q^2\right)\phi^1 - \frac{2i}{1-\phi_0}\, l_{\phi p}^2 \frac{l_p}{\tau_p} q p^1\right] - \lambda \frac{l_{\phi p}^2}{\tau_p}\left(l_p^2 q^2 p^1 + \frac{2i}{1-\phi_0} l_p q \phi^1\right), \tag{S58}$$

and enters the polarization-rate equation as $sp^1 = h_y^1/\gamma_1 + \lambda J_y^1$. The equations for the polarization and concentration now read

$$\begin{aligned} sp^1 &= -\frac{l_p^2 q^2}{\tau_p}\left[1+\lambda\left(\lambda + \frac{2}{1-\phi_0}\frac{1}{l_p}\right) l_{\phi p}^2\right] p^1 - \left[\frac{2}{1-\phi_0}\left(1+\lambda^2 l_{\phi p}^2\right) + \frac{\lambda l_\phi^2}{l_p}\frac{\tau_p}{\tau_\phi}\left(1+l_\phi^2 q^2\right)\right] i \frac{l_p}{\tau_p} q \phi^1, \\ s\phi^1 &= -\frac{l_\phi^2}{\tau_\phi} q^2 \left[1 + l_\phi^2 q^2 + \frac{2}{1-\phi_0} l_p \lambda \frac{l_{\phi p}^2}{l_\phi^2}\frac{\tau_\phi}{\tau_p}\right]\phi^1 + \left(\frac{2}{1-\phi_0} + \lambda l_p\right) l_{\phi p}^2 i \frac{l_p}{\tau_p} q^3 p^1. \end{aligned} \tag{S59}$$

A non-trivial solution exists for

$$s^2 + \left[\frac{l_p^2}{\tau_p} + \frac{l_\phi^2}{\tau_\phi}\left(1+l_\phi^2 q^2\right) + \lambda\left(\lambda + \frac{4}{1-\phi_0}\frac{1}{l_p}\right) l_{\phi p}^2 \frac{l_p^2}{\tau_p}\right] q^2 s + \frac{l_p^2}{\tau_p}\left[\frac{l_\phi^2}{\tau_\phi}\left(1+l_\phi^2 q^2\right) - \left(\frac{2}{1-\phi_0}\right)^2 \frac{l_{\phi p}^2}{\tau_p}\right] q^4 = 0. \tag{S60}$$

The permeation-alignment parameter, λ, enters the equation only in the coefficient of the term linear in s. It can induce a new instability only if the term linear in s is negative. The minimal value of the coefficient is obtained for $\lambda = -2/\left[(1-\phi_0)\, l_p\right]$. In this case, the equation reduces to

$$s^2 + \left[\frac{l_p^2}{\tau_p} + \frac{l_\phi^2}{\tau_\phi}\left(1+l_\phi^2 q^2\right) - \left(\frac{2}{1-\phi_0}\right)^2 \frac{l_{\phi p}^2}{\tau_p}\right] q^2 s + \frac{l_p^2}{\tau_p}\left[\frac{l_\phi^2}{\tau_\phi}\left(1+l_\phi^2 q^2\right) - \left(\frac{2}{1-\phi_0}\right)^2 \frac{l_{\phi p}^2}{\tau_p}\right] q^4 = 0. \tag{S61}$$

The coefficient of the term linear in s can become negative only if the constant term in s is itself negative. Therefore, the criterion for instability is not changed. In this calculation we did not treat the viscosity and elasticity explicitly. They can be absorbed in γ (and the resulting lengthscales), as was explained above.

C. Passive instability with permeation deformation ($\xi \neq 0$)

Permeation deformation induces a relative force between the network and solvent, which modifies the relative current,

$$\left(1 + \left[l_\eta^2 + 2\left(1-\phi_0\right)^2 \frac{l_B^2}{1+s\tau}\right] q^2\right) J_y^1 = -iq\left[\frac{l_\phi^2}{\tau_\phi}\left(1+l_\phi^2 q^2\right)\phi^1 - \frac{2i}{1-\phi_0}\, l_{\phi p}^2 \frac{l_p}{\tau_p} q p^1\right] - \frac{1}{2}\xi\left[\phi_0\left(1-\phi_0\right)\right]^2 \frac{l_B^2}{\tau} B_{xy}^1. \tag{S62}$$

Here we treat the viscosity and elasticity explicitly. This is required because the strain evolves differently, due to the permeation-deformation mechanism. The remaining strains evolve as

$$\begin{aligned} \left(1+s\tau\right) B_{xy}^1 &= \frac{1}{2}\xi\tau J_y^1 + iq\tau v_x^{n(1)}, \\ \left(1+s\tau\right) B_{xx}^1 &= \xi\tau J_x^1. \end{aligned} \tag{S63}$$

The relative current in the x-direction is induced by the shear stress and permeation-deformation contribution, according to

$$J_x^1 = \frac{l_B^2}{\tau}\phi_0\left(1-\phi_0\right)\left[iqB_{xy}^1 - \frac{1}{2}\phi_0\left(1-\phi_0\right)\xi B_{xx}^1\right]. \tag{S64}$$

The solvent velocity in the x-direction is found from the force-balance on the gel in the x-direction. Equating the total shear stress to zero yields $iq\eta_s v_x^{s(1)} = -G\phi_0 B_{xy}^1$. The network velocity is found from $v_x^{n(1)} = v_x^{s(1)} + J_x^1/\left[\phi_0\left(1-\phi_0\right)\right]$, as

$$v_x^{n(1)} = \frac{l_B^2}{\tau}\left[iqB_{xy}^1 - \frac{1}{2}\phi_0\left(1-\phi_0\right)\xi B_{xx}^1\right] + 2i\phi^2 \frac{l_B^2}{l_\eta^2}\frac{1}{q\tau} B_{xy}^1. \tag{S65}$$

We can now find the shear strain in terms of the relative current

$$B_{xy}^1 = \frac{1}{2}\xi\tau\left(1 + s\tau + 2\phi^2\frac{l_B^2}{l_\eta^2} + \frac{1+s\tau}{1+\frac{\tau}{\tau_\xi}+s\tau} l_B^2 q^2\right)^{-1} J_y^1, \tag{S66}$$

where $\tau/\tau_\xi = \left[\phi_0\left(1-\phi_0\right)\xi l_B\right]^2/2$. Inserting this back in the equation for the relative current yields

$$J_y^1 = -iq\frac{\frac{l_\phi^2}{\tau_\phi}\left(1+l_\phi^2 q^2\right)\phi^1 - \frac{2i}{1-\phi_0}\,l_{\phi p}^2\,\frac{l_p}{\tau_p}qp^1}{1+\left[l_\eta^2+2\left(1-\phi_0\right)^2\frac{l_B^2}{1+s\tau}\right]q^2+\frac{1}{2}\frac{\tau}{\tau_\xi}\left(1+s\tau+2\phi^2\frac{l_B^2}{l_\eta^2}+\frac{1+s\tau}{1+\frac{\tau}{\tau_\xi}+s\tau}l_B^2q^2\right)}^{-1}. \tag{S67}$$

Therefore, the permeation-deformation mechanism results in a renormalized $s-$ and $q-$dependent friction coefficient. As the new coefficient is positive for $s>0$, We can renormalize our lengthscales and timescales, as we have done above, and obtain the same criterion for instability as in the $\lambda=\xi=0$ case.

We have assumed in our calculations that ϕ^1 and p^1 do not vanish. The solution $\phi^1=0$ and $p^1=0$, results in $J_y^1=0$, and therefore $B_{xy}^1=0$. This infers that B_{xx}^1 and B_{yy}^1 vanish as well. This shows that our calculation above holds for any eigenvector of the linear equations.

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