Letter

Oscillating chemical reactions enable communication between responsive hydrogels

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Responsive hydrogels can sense environmental stimuli and respond as actuators by expelling water and changing shape. In this Letter, we develop theory to demonstrate that groups of responsive hydrogels can also communicate with each other, by utilizing the effect of elastic deformation on chemical reaction dynamics. Specifically, we consider a system of two spatially separated chemically responsive hydrogels suspended in a solution in which a Belousov-Zhabotinsky (BZ)-type reaction occurs. Solving for the gel dynamics with the transport of solvent through the poroelastic network and the chemical kinetics, we show how the periodic swelling-deswelling oscillations of each gel can become coupled, and how this coupling can be exploited to send signals from one gel to the other via mechanical manipulation of the sender that affect the local (and thus global) frequency of oscillation.

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Introduction. Hydrogels are increasingly employed in the construction of biocompatible, flexible machinery [1] that exploits their soft, elastic, and porous properties. These gels comprise a cross-linked hydrophilic polymer surrounded by water molecules that are free to move, and volumetric changes of up to two orders of magnitude are observed when they are swollen in water from an initially dry state [2]. Of particular interest in the design of smart devices are responsive hydrogels, which lose their affinity for water in response to stimuli [3].

Through the choice of polymer, gels can be produced that respond to temperature [4], light [5], electromagnetic fields [6], and pH or chemical concentration [7]. Recent experiments have illustrated how programmable shape changes can be realized in chemically responsive hydrogels coupled to oscillating chemical reactions [8–10]. These systems have been used in the laboratory to model processes such as the beating of cardiac tissue [11] and peristaltic squeezing [12], and to transport droplets [13]. In addition to a wealth of experimental studies of these so-called BZ gels that employ the famous Belousov-Zhabotinsky (BZ) reaction [14], there has been much progress modeling such gels [15]. Models usually treat the hydrogel as a lattice of springs whose natural length varies in the presence of chemical species that are generated and destroyed following one of a number of popular models for the BZ reaction [16].

There exist abundant examples of gels that sense the products of chemical reactions and respond accordingly, but it is also possible for these gels to influence the dynamics of the reactions themselves, enabling feedback mechanisms. Nava-Medina *et al.* [10] constructed a BZ gel with a catalyst bonded to the polymer chains such that the deformation of the gel influences the rate of reaction. These two-way relationships between a gel and its environment can be exploited to develop new paradigms for smart devices that both sense and influence their surroundings, or coordinate in systems inspired by living matter.

Chemical signaling is used in biological systems both between organisms (e.g., quorum sensing within bacterial colonies) and within multicellular life (in hormonal signals between organs) [17]. Recent studies have used BZ gels as an analogue for such processes, leading to collective motion of "swarms" of gels that communicate using diffused products [18]. Inspired by these natural systems, we consider a pair of gels coupled to an oscillating reaction, with catalyst molecules bound to their polymer scaffolds. This system exhibits periodic oscillations in the gels, allowing signals to be sent through exploiting the diffusion of chemical solute in the solvent separating them. Signaling is achieved via mechanical manipulation of the "sender" gel, adjusting the local frequency of oscillation and hence the global dynamics in a manner akin to frequency modulation radio. Not only does this provide a model for intercellular communication, it also unlocks the development of smart hydrogel devices that coordinate complicated responses using insights from biology, employing tried-and-tested building blocks that are straightforward to implement in the real world.

Modeling foundations. We first seek a description of the response of a hydrogel to a chemical concentration field $c(\boldsymbol{x}, t)$. This is traditionally achieved using an expression for the energy density of the deformed gel, which is then used to find stresses on virtual springs joining points in the gel—the gel lattice-spring model of Ref. [15].

In the present study, we restrict our attention to gels undergoing uniaxial deformation and, instead of specifying a particular elastic and energetic law for the hydrogel, consider an osmotic pressure, linear in the polymer volume fraction ϕ,

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that changes in nature when a critical chemical concentration c^* is reached:

$$\Pi = \Pi_0 \frac{\phi - \phi_0(c)}{\phi_0(c)} \quad \text{with} \quad \phi_0(c) = \begin{cases} \phi_{00}, & c \leqslant c^*, \\ \phi_{0\infty}, & c > c^*, \end{cases} \tag{1}$$

where ϕ_0 is the equilibrium polymer fraction [19]. If $c > c^*$, the gel will deswell from an initial equilibrium $\phi \equiv \phi_{00}$, reversibly, to an equilibrium polymer fraction $\phi_{0\infty} > \phi_{00}$ in the absence of any mechanical constraints.

Changes in ϕ result from the flow of water through the scaffold, which can be quantified in a uniaxial geometry by the interstitial fluid flux

$$q = \frac{D(\Phi)}{\Phi}\frac{\partial \Phi}{\partial x} = \frac{k\Pi_0}{\mu_l}\left(\frac{1}{\Phi_0} + \frac{4\mu_s}{3\Pi_0\phi}\Phi^{\frac{1}{3}}\right)\frac{\partial \Phi}{\partial x}, \tag{2}$$

where $\Phi = \phi/\phi_{00}$, k is the gel permeability, μ_s is the shear modulus, μ_l is the dynamic viscosity of the water, and Π_0 is a constant osmotic modulus [20], as derived in *Appendix A: Derivation of the gel model.* If the initial thickness of swollen hydrogel is a_0 and the gel occupies the space $0 \leqslant x \leqslant a(t)$, swelling and drying are governed by the nonlinear diffusion equation

$$\frac{\partial \Phi}{\partial t} = \frac{\partial}{\partial x}\left[D(\Phi)\frac{\partial \Phi}{\partial x}\right] \quad \text{with} \quad \begin{cases} \Phi(a(t),\, t) = \Phi_0, \\ \partial_x \Phi(0,\, t) = 0, \end{cases} \tag{3}$$

with the boundary condition at $x = 0$ arising from no flow ($q = 0$) and that at $x = a(t)$ from a stress balance [20], again justified in the Appendix. Polymer conservation sets the thickness of the gel in terms of its initial value,

$$\int_0^{a(t)} \Phi \, \mathrm{d}x = a_0 \quad \text{so} \quad \frac{\mathrm{d}a}{\mathrm{d}t} = -\frac{D(\Phi_0)}{\Phi_0}\partial_x \Phi(a(t),\, t). \tag{4}$$

Reaction modeling. The best-known example of an oscillatory chemical reaction is the Belousov-Zhabotinsky reaction with a bromate oxidizer [14]. A number of different models for this reaction have been proposed in the literature [16], including the Brusselator [21], featuring four reaction steps:

$$A \to X, \quad 2X + Y \to 3X, \quad B + X \to Y + F, \quad \text{and} \quad X \to G.$$

In the present study, we will consider only this model (a general approach for autocatalytic reactions) in lieu of more complex models that are specific to the BZ reaction (such as the Oregonator) owing to its analytic simplicity and the fact it retains the key mechanisms driving chemical oscillations, but this model could easily be replaced with any other approach in the governing equations. In the case of an excess of A and B, the equations governing the reaction can be rescaled, and reaction dynamics are described by

$$\dot{X} = rR_X(X,\, Y) = r[A + X^2Y - (1 + B)X], \tag{5a}$$

$$\dot{Y} = rR_Y(X,\, Y) = r[-X^2Y + BX], \tag{5b}$$

where X and Y are the concentration of the two product species and r is a rate constant. The assumption that A and B cannot be depleted allows for the existence of solutions that perpetually oscillate, with a Hopf bifurcation where $B = 1 + A^2$ and the single fixed point becomes unstable. These oscillations take the form of limit cycles [22] whose approximate period is

$$T \approx (1 + B)^2/4rA, \tag{6}$$

when $B/A \gg 1$, as calculated in the Supplemental Material [23] through noting that the system takes the form of a relaxation oscillator. In spite of its apparent simplicity, solutions of the Brusselator system have been shown to provide good quantitative models for the processes that underpin BZ reactions [24], since many such oscillations decay very slowly in time.

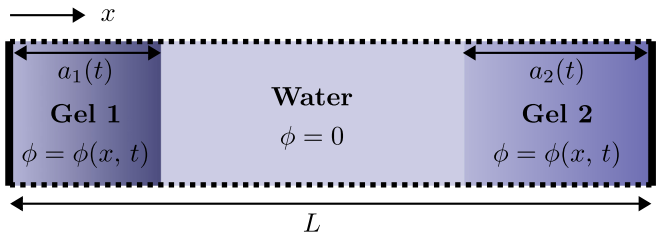


FIG. 1. A schematic illustrating the configuration of two gels in a closed container, separated by water. For clarity, the horizontal direction is shrunken, when actually we are modeling the case $a_1,\ a_2 \ll L$.

Chemomechanical oscillations of a single gel. In experiments, Yoshida *et al.* [25] note that the swelling–deswelling cycle (the 'mechanical response') synchronizes with the chemical oscillation, with zero phase difference. Though the large number of parameters in this study make fitting our results to existing data a possible yet nonunique endeavor, Fig. 2 shows that this qualitative behavior seen in a single gel is captured here, with length changes occurring in phase with

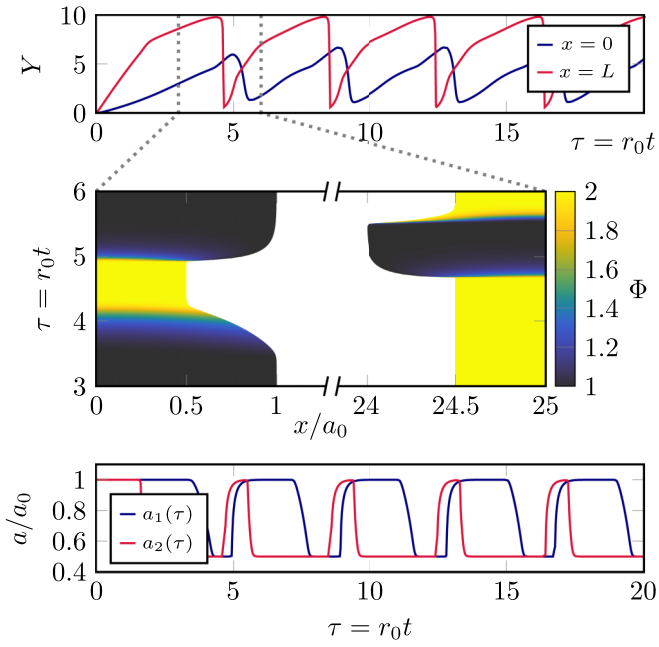


FIG. 2. Plots of the length of the two gels, profiles of the gel composition, and the concentration of the product Y at $x = 0$ and $x = L$ showing how the two gels shrink and reswell. In this plot, $A = 1$, $B = 5$, and $L = 25a_0$. The rate constant $r(\Phi) = r_0$ in the left-hand gel and $5r_0$ in the right-hand gel. The Damköhler number $Da_I = 1$ with $t_{\text{diff}} = 0.125t_{\text{pore}}$. The critical concentration $Y^* = 5$ and $\phi_{0\infty} = 2\phi_{00}$.

chemical concentration peaks. In the Supplemental Material [23], we discuss the nature of these oscillations, which cannot be proven to be limit cycles but exhibit near-periodicity with an approximately constant period.

Two-gel systems. Figure 1 illustrates the model of this article, where two gels occupy the regions $0 \leqslant x \leqslant a_1(t)$ and $L - a_2(t) \leqslant x \leqslant L$, respectively, with $L \gg a_1,\ a_2$. When fully swollen in the absence of any chemical species, $a_1 = a_2 = a_0$ and $\Phi \equiv 1$ in both gels. Reactions occur only in the hydrogel, where there is a catalyst bonded to the polymer scaffold, and the rate of reaction is proportional to the concentration of reagents, as in the experiments of Ref. [10]. The rate constant can depend on the polymer volume fraction, since the catalyst concentration is high in drier gels where the scaffold is tightly packed. To incorporate this effect, we replace r in Eq. (5) with $r(\Phi)$.

Gel dynamics are governed by the system (3), where we assume $D(\Phi) \equiv D$, a constant [26]. This equation can be coupled with an advection-diffusion-reaction model for the concentration of chemical species c,

$$\text{solute:} \quad \frac{\partial c}{\partial t} + q\frac{\partial c}{\partial x} = r(\Phi)R_c(X,\ Y) + \mathcal{D}\frac{\partial^2 c}{\partial x^2}, \tag{7a}$$

$$\text{gel:} \quad \frac{\partial \Phi}{\partial t} = D\frac{\partial^2 \Phi}{\partial x^2}, \tag{7b}$$

where c represents X or Y, $q = 0$ outside of the gel, and $\mathcal{D}$ is the molecular diffusion coefficient, equal to D_w in the water and D in the gel. We assume that the gels respond to the concentration of species Y, with rapid deswelling occurring above a critical threshold $Y = Y^*$.

Figure 2 shows an example solution to this coupled set of partial differential equations, illustrating how the shape of a gel and its composition both change in an oscillating pattern in response to chemical signals, which diffuse across the gap between gels. It is difficult to determine whether the oscillations seen here are limit cycles or evidence of quasiperiodicity, but it is clear from Fig. 2 and justified in the Supplemental Material [23] that at least near-periodic solutions persist for long times.

Coupled oscillator model. It is difficult to make predictions from this system of equations, and some effects, such as the advective term on the left-hand side of Eq. (7a), are less important than others, e.g., the reaction dynamics or the diffusion of species between hydrogels. There are three timescales in the problem, corresponding to chemical kinetics, diffusion in the water, and the poroelastic transport in the gels,

$$t_{\text{react}} = 1/r_0, \quad t_{\text{pore}} = \mu_l a_0^2/k\Pi_0, \quad \text{and} \quad t_{\text{diff}} = L^2/D_w, \tag{8}$$

where r_0 is a typical scale for $r(\Phi)$. In many cases, it is possible to reduce the generally applicable full syst0em (7) to a system of coupled oscillators. This behavior is well attested experimentally in studies of coupled BZ droplets, where rich coupled-oscillator dynamics have been observed [27].

Such a system of "diffusively coupled" Brusselators has been studied in the literature [28–30], where two oscillators are linked by the transport of chemical species between them. The analysis of these systems is mathematically complicated, with the existence, stability, and dynamics of oscillatory solutions dependent on the strength of coupling, as discussed in the Supplemental Material [23]. In order to deduce the coupling strength in our system, we must parameterize the diffusive transport of chemical species across the gap separating the two BZ gels.

First, we assume that $t_{\text{react}} \gg t_{\text{pore}}$. This latter timescale is set by diffusion through pores, and though this is typically of the order of minutes, the reaction timescale can be tuned to be slower by setting the amount of catalyst bonded to the gel. Scaling time with t_{react} in Eq. (7a) shows that the chemical concentration is approximately uniform within the gels, as illustrated in Eq. (B2). We now denote the concentrations in the two gels by X_1, X_2, Y_1, and Y_2.

The ratio $t_{\text{pore}}/t_{\text{react}}$ is equal to the first Damköhler number Da_I, and taking $Da_I \ll 1$ suggests that the gel reconfigures rapidly to changes in chemical concentration, instantaneously reaching $\Phi_0(Y)$, allowing us to neglect the influence of gel dynamics on the problem. In the water, scaling times with t_{react}, we assume that diffusion occurs on a much faster timescale than the reaction or diffusion in the gel, $t_{\text{diff}} \gg t_{\text{pore}} \gg t_{\text{react}}$, and so, as seen in Eq. (B3), the concentration fields are linear between the two hydrogels.

Thus, the flux of chemical species across the gel-water boundary can be approximated by the diffusive flux Q_c of chemical species c within the water,

$$Q_c = -D_w\frac{\partial c}{\partial x} = -\frac{D_w(c_2 - c_1)}{L - a_1 - a_2} \approx -\frac{D_w(c_2 - c_1)}{L}, \tag{9}$$

assuming that $a_0 \ll L$. Homogeneity within the gels implies that the chemical species delivered to the gel spreads out instantaneously, increasing the concentration at a rate $-Q_c/a_1$ in the left-hand gel and Q_c/a_2 in the right-hand gel. Finally, we introduce a nondimensional time $\tau = t/t_{\text{react}}$, and assume the simplest possible form for $r(\Phi) = r_0\Phi$, to give a system of coupled equations for the concentration c of X or Y in each gel,

$$\frac{\mathrm{d}c_i}{\mathrm{d}\tau} = \Phi_i R_c + \frac{Qa_0}{a_i}(c_j - c_i) \qquad Q = \frac{L}{a_0}\frac{t_{\text{react}}}{t_{\text{diff}}} \gg 1, \tag{10}$$

where $i = 1,\ 2$, $i \neq j$, and $\Phi_i = \phi_0(Y_i)/\phi_{00}$.

Linking gel mechanics to reaction dynamics. The dynamics of the system of Eq. (10) can be influenced by varying the reaction rate in one gel but not the other. We achieve this by imposing a mechanical strain on the second gel, such that we can control the concentration of catalyst in the reaction occurring there. Applying a uniform vertical compressive strain E to the gel changes the horizontal stress balance at the interface with water, and hence changes the polymer fraction in both swollen and deswollen states [31]:

$$\frac{\Phi - \Phi_0(c)}{\Phi_0(c)} = \frac{\mu_s}{\Pi_0}(1 + E - \Phi^{1/3}). \tag{11}$$

Therefore, assuming there are no mechanical constraints on the left-hand gel as it shrinks and swells,

$$\Phi_1 = \begin{cases} 1, & Y_1 \leqslant Y^*, \\ \Phi_{0\infty}, & Y_1 > Y^*, \end{cases} \qquad \Phi_2 = \begin{cases} \Phi_2^-, & Y_2 \leqslant Y^*, \\ \Phi_2^+, & Y_2 > Y^*, \end{cases} \tag{12}$$

with $\Phi_2^{\pm}$ obtained by solving Eq. (11) below and above $Y_2 = Y^*$. The gel widths are determined by conservation of

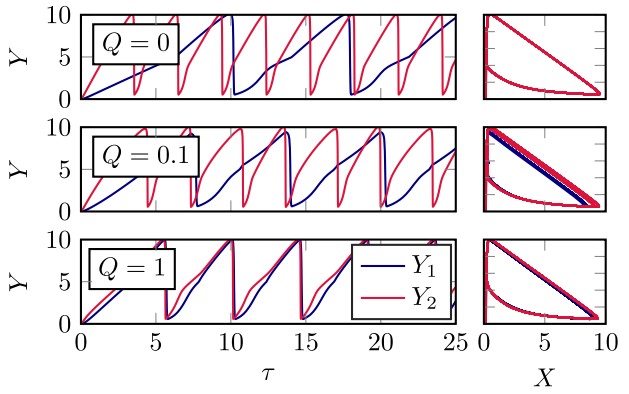


FIG. 3. Solutions to the simplified coupled oscillator model (10) for different coupling strengths Q when $A = 1$ and $B = 5$. Here, $\phi_{0\infty} = 2\phi_{00}$ and $Y^* = 5$, and the second gel is compressed with $E = 1$ and $\mu_s/\Pi_0 = 100$. On the right, phase-plane plots of Y against X showing behavior close to periodicity when $X_i = Y_i = 0$.

polymer,

$$a_1 = \Phi_1^{-1} a_0 \quad \text{and} \quad a_2 = (1 - E)^{-1} \Phi_2^{-1} a_0. \tag{13}$$

Solving Eq. (10) shows the existence of periodic behavior, just as in the single-oscillator case. Figure 3 illustrates how changing the coupling strength leads to synchronization of the two nonlinear oscillators. When $Q = 0$, both reactions occur independently, with the two systems exhibiting different periods. As the strength of coupling increases, the periods synchronize as integer multiples of each other before equalizing at an intermediate value between the natural periods of each oscillator. The process governing this coupling for intermediate Q is explained in the Supplemental Material [23].

In the large-Q regime, it is apparent that $X_1 = X_2$ and $Y_1 = Y_2$, and so Eq. (10) reduces to those for a single oscillator of the form of Eq. (5) with rate $r = (\Phi_1 + \Phi_2)/2$. We can therefore use an analogous approach to that for the single oscillator (6) to find the oscillation period,

$$T = \frac{(1+B)^2}{2A^2(\Phi_\infty + \Phi_2^+)} + \frac{2Y^*}{A} \frac{(\Phi_\infty - 1) + (\Phi_2^+ - \Phi_2^-)}{(1+\Phi_2^-)(\Phi_\infty + \Phi_2^+)}, \tag{14}$$

provided that $(1+B)^2/4A > Y^*$, as also derived in the Supplemental Material.

Strong coupling occurs when the diffusivity in the water is high, reaction rate constant sufficiently low, and when the separation between gels is not too large. In general, $D_w \sim 10^{-9}\,\mathrm{m}^2\,\mathrm{s}^{-1}$ [32], and r_0 is approximated well by the reciprocal of the characteristic timescale for the reaction. Therefore, $Q \gg 1$ when

$$\text{reaction timescale} \gg (10^9\,\mathrm{m}^{-2} a_0 L)\,\mathrm{s}. \tag{15}$$

For millimeter-scale systems, this requires reactions to occur on a timescale of minutes, or hours in the case of centimeter-scale systems, both reasonable choices seen in experiments [9].

This allows for a signal to be transmitted between two spatially separated gels by modulating the frequency of the first gel's oscillations through varying the compression E in time. In the limit of strong coupling, Eq. (14) gives the oscillation period that both gels settle to, and this is seen to be dependent on the polymer fraction $\Phi_2^\pm$, allowing measurements of the behavior of the left-hand gel to be used to deduce the state of the right-hand gel.

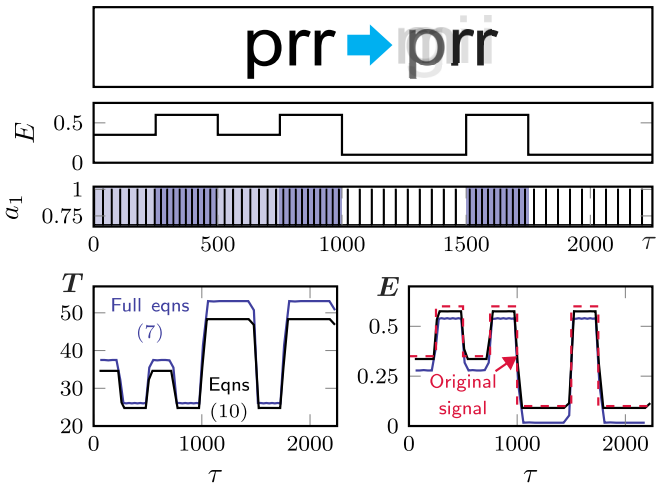


FIG. 4. To send the text PRR, the letters are converted to values {16, 18, 18}, which are then given ternary representations {121, 200, 200}. Each ternary digit sets $E(\tau)$, defined piecewise in sections of length $\Delta\tau = 250$, and we can solve the simplified oscillator model (10) and the full governing equation (7) to find the effect on the period of gel oscillation. To produce this plot, $A = 1$, $B = 15$, $Y^* = 5$, and $\Phi_\infty = 1.5$. When solving the full equations, we take $Da_I = 1$ and $L = 25a_0$ with $t_{\mathrm{diff}} = 0.125 t_{\mathrm{pore}}$. This leads to $Q = 40$, used in the solution of Eq. (10).

For the sake of analytic simplicity, we restrict our attention to stiff gels with $\mu_s/\Pi_0 \gg 1$, where Eq. (11) implies that $\Phi_2^\pm$ are both $(1+E)^3$. Thus, we can use Eq. (14) to deduce the compressive strain E on the second gel from a measurement of the period T. If the imposed strain changes sufficiently slowly that the oscillations in both gels settle to a steady period before E changes again, this can be used to send information from one gel to another through a frequency-modulated signal, as illustrated in Fig. 4.

Here, the strain $E(\tau)$ on the second gel is varied and the time period of oscillations in the first gel's size is measured. This signal is reproduced near-perfectly from a solution to the gel system modeled by coupled oscillators, as would be expected given that our expression for oscillation period is based on this model, but perhaps more importantly this same expression, applied to a numerical solution of the full system of governing Eq. (7), is seen to give an accurate prediction of the forcing signal.

Small discrepancies arise due to the fact that the strongly coupled oscillator model neglects small differences in concentration between the two gels, explaining why the black curve in Fig. 4 does not exactly match the signal. Larger differences occur between the signal predicted from the solution to the full equations, resulting from gel reconfiguration, spatial variation in concentration within the hydrogels, and a nonlinear concentration profile, all of which we have neglected here. In spite of this, it is clear that the oscillator model allows for the strain $E(\tau)$ to be deduced accurately through a measurement of the response period and can be applied to deduce this information without having to solve for the full dynamics.

Conclusions. In this article, we have introduced and modeled a frequency modulation paradigm for communication between responsive hydrogels, creating a basis for the development of increasingly complex gel machines with separate components. By exploiting the influence of mechanical manipulation on the dynamics of an oscillating chemical reaction occurring in a gel, we showed how local changes in one hydrogel can affect the global dynamics of a system of oscillating gels that are coupled through diffusion of chemical species.

Much as organ systems can send out chemical signals in the form of hormones in multicellular life, or colonies of microorganisms can inform others of danger or resources [17], this chemical signaling allows us to build up complex hydrogel machines that comprise multiple communicating parts. Mechanical input can be converted to chemical signals that then result in morphological changes, allowing for physical strains to be transmitted across a distance, in much the same way as a traditional telephone converts sound waves into an electrical signal and back again.

This has twofold importance: First, it illustrates how artificial systems constructed from hydrogels can act as analogues for important biological processes such as quorum sensing, and collective dynamics can result from an external input. For example, if one gel is placed under strain, others nearby can receive this information and move away or respond accordingly. Second, we can use these communication processes to chain together hydrogels to achieve complex goals—one immediate example could be coordinating a number of responsive hydrogel pumps placed at different positions to maintain a constant concentration of chemical species.

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Data availability. No data were created or analyzed in this study.

Appendix A: Derivation of the gel model. In this study, we use a linear-elastic-nonlinear-swelling (LENS) model to describe the hydrogel dynamics, basing our description of chemically responsive gels on a recent description of temperature responsive gels [33]. However, all models of poroelasticity are built on a foundation of conservation of the two components of the system—the solid phase (in this case the polymer chains making up the gel scaffold) and the liquid phase (the water in the pore spaces) [2]. In this case, we assume that both of these phases are individually incompressible and that ϕ represents the local polymer volume fraction, and $\phi_w = 1 - \phi$ is the local water volume fraction, so that

$$\frac{\partial \phi}{\partial t} + \frac{\partial}{\partial x}(\phi u_p) = 0 \quad \text{and} \quad \frac{\partial \phi_w}{\partial t} + \frac{\partial}{\partial x}(\phi_w u_w) = 0, \tag{A1}$$

where u_p is the polymer velocity and u_w is the fluid velocity. Adding these two equations shows that

$$\phi u_p + \phi_w u_w = \text{constant}. \tag{A2}$$

If, as in the geometry considered in Fig. 1, there is a rigid boundary, neither water nor polymer can pass and so the average velocity of the two phases is zero: If polymer moves in one direction, water must move to fill the space it has left, and vice versa. Equation (A2) shows that the same assumption must be applied throughout the gel, and so $\phi u_p = -\phi_w u_w$. Defining $q = \phi_w(u_w - u_p)$ as the relative flux of water molecules against a reconfiguring scaffold, Eq. (A1) also shows that

$$\frac{\partial \phi}{\partial t} = \frac{\partial}{\partial x}(\phi q). \tag{A3}$$

The deswelling and reswelling behavior of a chemically responsive hydrogel when the external chemical field is changed occurs as the polymer chains reconfigure in the presence of the solute species and their affinity for water molecules is accordingly modified. In the case of BZ gels, it is often the concentration of $[\mathrm{Ru}(bpy)_3]^+$ ions that modifies the structure of the polymer chains and leads to changes in the equilibrium degree of hydration [15]. The simplest way to encode this is through an equilibrium polymer fraction ϕ_0 that depends on the local concentration c, i.e.,

$$\phi_0(c) = \begin{cases} \phi_{00}, & c \leqslant c^*, \\ \phi_{0\infty}, & c > c^*. \end{cases} \tag{A4}$$

A gel's propensity to swell or deswell is governed by the osmotic pressure Π, and in the absence of any external mechanical constraints, we expect $\phi \to \phi_0$ after water has been expelled or imbibed, and hence $\Pi = 0$ at the equilibrium polymer fraction. In the present study, we therefore choose a linear osmotic pressure $\Pi = \Pi_0(\phi - \phi_0)/\phi_0$ that can be expected to fit the behavior close to the equilibrium polymer fraction well [19] and give a good qualitative description of swelling and deswelling behavior.

The transient evolution of polymer fraction when c is changed is governed by the interstitial fluid flow q. It is standard practice in poroelasticity to link q to gradients in pore or "pervadic" [34] pressure p through Darcy's law, such that

$$q = -\frac{k}{\mu_l}\frac{\partial p}{\partial x}, \tag{A5}$$

where k is the permeability and μ_l the dynamic viscosity of water. These gradients in pore pressure must balance gradients in bulk (effective) stress in the gel material itself such that the entire system is in force-free equilibrium, i.e.,

$$\frac{\partial}{\partial x}(\sigma_{\text{eff}} - p) = 0. \tag{A6}$$

There are a number of different models for the effective stress in a hydrogel: In the absence of any elastic stresses, we would expect σ_{eff} to only have a contribution from osmotic pressure Π, but adding in the effects of polymer chain elasticity leads to models ranging from the fully linear approach of Ref. [26] to hyperelastic models [2,35]. In the present study, we have used the LENS theory that permits large deviations from equilibrium polymer fraction but linearizes around small deviatoric strains, as introduced by Refs. [20,36] for its analytic simplicity and good agreement with fully nonlinear approaches using only a small number of macroscopically meaningful parameters [33]. In this case,

$$\frac{\partial \sigma_{\text{eff}}}{\partial x} = \frac{1}{\phi}\frac{\partial \Pi}{\partial x} + \frac{4\mu_s}{3\phi}\left(\frac{\phi}{\phi_{00}}\right)^{1/3}\frac{\partial \phi}{\partial x}, \tag{A7}$$

where μ_s is the shear modulus, assumed independent of ϕ (as is indeed the case in many nonlinear models [33,35]). This

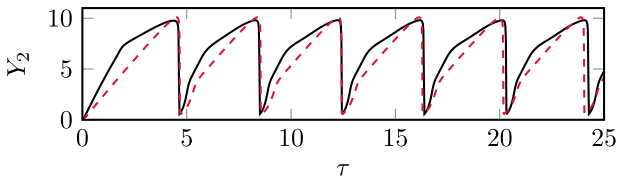


FIG. 5. Plots of Y evaluated at $x = L$ (solid black line) and Y_2 (dashed red line) in the full model of Eq. (7) and the coupled ordinary differential equation system of Eq. (10), respectively. The same parameters are used as in Fig. 2, and we therefore take $Q = 40$ to produce the approximate plot.

expression, combined with Eq. (A5), leads to the interstitial fluid flux expression of Eq. (2).

At the interface between gel and water, we require a boundary condition arising from stress balances. The pervadic pressure is continuous across phase boundaries [34] and is the only component of the stress tensor in the fluid domain away from the gel. Therefore, imposing continuity of normal stress requires $\sigma_{\text{eff}} = 0$ at $x = a(t)$. In the absence of any deviatoric strains, the effective stress just has a contribution from the osmotic pressure, forcing $\Pi = 0$ at the interface and setting $\phi = \phi_0$ here, while adding deviatoric strains (for example, by compressing the hydrogel) changes the balance and gives a drier interfacial polymer fraction, as seen in Eq. (11).

Appendix B: Nondimensionalization and reduction of the full governing equations. Equation (7) can be nondimensionalized by introducing a time variable $\tau = r_0 t$ scaled with the characteristic reaction timescale $t_{\text{react}} = 1/r_0$, and scaling all lengths with a_0, we let $\tilde{x} = x/a_0$. Thus, recalling that $q = (D/\Phi)\partial\Phi/\partial x$, Eq. (7a) becomes

$$\frac{\partial c}{\partial \tau} + \frac{D}{r_0 a_0^2 \Phi}\frac{\partial \Phi}{\partial \tilde{x}}\frac{\partial c}{\partial \tilde{x}} = \frac{r(\Phi)}{r_0}R_c(X, Y) + \frac{\mathcal{D}}{r_0 a_0^2}\frac{\partial^2 C}{\partial \tilde{x}^2}. \tag{B1}$$

Since $t_{\text{pore}} = a_0^2/D$ and $t_{\text{react}} = 1/r_0$, in the gel

$$\frac{\partial c}{\partial \tau} + \frac{t_{\text{react}}}{t_{\text{pore}}\Phi}\frac{\partial \Phi}{\partial \tilde{x}}\frac{\partial c}{\partial \tilde{x}} = \frac{r(\Phi)}{r_0}R_c(X, Y) + \frac{t_{\text{react}}}{t_{\text{pore}}}\frac{\partial^2 c}{\partial \tilde{x}^2}. \tag{B2}$$

When $t_{\text{react}} \gg t_{\text{pore}}$, the only admissible balances in this equation involve balancing advection and diffusion, and it is clear that we expect reaction dynamics to play a role, so we must have $\partial c/\partial \tilde{x} = 0$ at leading order. In the water,

$$\frac{\partial c}{\partial \tau} = \frac{L^2}{a_0^2}\frac{t_{\text{react}}}{t_{\text{diff}}}\frac{\partial^2 c}{\partial \tilde{x}^2}, \tag{B3}$$

and hence $\partial^2 c/\partial \tilde{x}^2 = 0$ since $t_{\text{react}} \gg t_{\text{diff}}$ and $L \gg a_0$. Gel dynamics follow with

$$\frac{\partial \Phi}{\partial \tau} = \frac{t_{\text{react}}}{t_{\text{pore}}}\frac{\partial^2 \Phi}{\partial \tilde{x}^2}. \tag{B4}$$

The assumption $t_{\text{react}} \gg t_{\text{pore}}$ shows that $\Phi \equiv \Phi_0(Y)$ in both gels. The coupled system of oscillators reached in Eq. (10) can be compared with the full numerical solutions to Eq. (7), as illustrated in Fig. 5, showing strong agreement with the full numerical result, capturing the period of oscillations accurately but the transient behavior differs significantly owing to reconfiguration of the gel, not captured in the reduced model.

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