Supplementary Information

1. Simulation validation

We apply beam elements to model the gel surface. The beam behaves like a real surface if it satisfies: (1) a uniform prestress is applied so that the tension in the beam matches the surface tension γ, (2) the tensile stiffness of the beam is so low that the deformation in the simulation negligibly affects the prestress and the cross-section Poisson's ratio is set to 0 so that deformation does not change the cross-section area. Then the surface tension γ is independent of deformation. (3) the bending stiffness of the beam is so low that the beam layer negligibly affects the deformation of the gel. To satisfy (1), we applied a uniform artificial thermal stress inside the beam using a uniform thermal expansion coefficient and a uniform temperature field. The thermal stress times the beam cross-section area matches the surface tension γ. To satisfy (2), we set the tensile stiffness of the beam to be much lower than the prestress so that the deformation negligibly affects the prestress. Also, the cross-section Poisson's ratio is set to 0 so that deformation does not change the cross-section area. In our simulation, we set the beam modulus to be $\sim 10^{-4}$ of the prestress. As shown in Fig. S1a, even for the case of the smallest surface tension ($\gamma/\mu L_0 = 0.02$), the maximum change in surface tension over the beam is negligible. To satisfy (3), we set the bending stiffness of the beam to be negligible compared to μL_0^2, where μ is the shear modulus of the gel, L_0 is the wavelength of the surface roughness, and our plane-strain simulation has the unit thickness. In our simulation, we set the bending stiffness to be $\sim 10^{-14}$ of μL_0^2. As shown in Fig. S1b, the maximum sectional moment in the beam, M, is negligible compared to μL_0^2. The parameters of the beam section are summarized in Table S1.

Table S1. Simulation parameters in beam section.

Beam parameter	Young's modulus	Section Poisson's ratio	In-plane width	Thickness
Input values	0.001	0	1	0.001

In osmocapillary phase separation, the free surface represented by the beam above is unstable in the implicit solver, ABAQUS Standard. To stabilize the simulation, we add an elastic layer above the beam (Fig. S1c). We tune the modulus of the elastic layer to ensure that the elastic energy to stabilize the beam is negligible compared to the surface energy of the beam layer (Fig. S1d). Note that since $\lambda_{dry} < 1.5$ in our studies, we did not use the simulation result for the whole range of $\lambda_1 < 5$. The $W_{ela}/\gamma L_0$ with $\lambda_1 < 1.5$ is less than 0.1%.
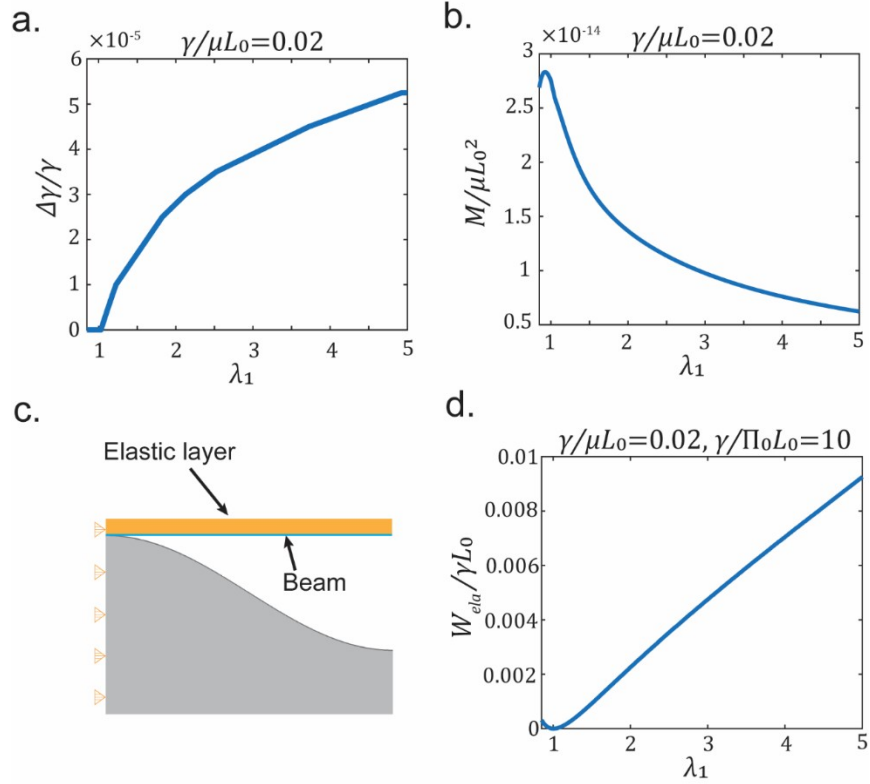
Figure S1. (a) The change in surface tension remains low during stretch. **(b)** The bending moment of the beam remains low during stretch. **(c)** An elastic layer is added to stabilize the simulation. **(d)** The elastic energy of the elastic layer remains low during stretch.

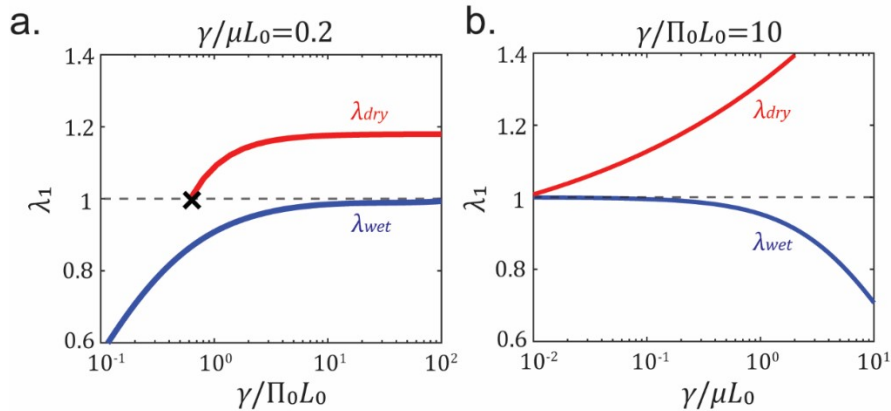## 2. Critical stretches varies with $\gamma/\mu L_0$ and $\gamma/\Pi_0 L_0$

Figure S2. (a) Under a fixed $\gamma/\mu L_0$, a larger $\gamma/\Pi_0 L_0$ shifts both λ_{dry} and λ_{wet} to larger stretches, and the gaps between λ_{dry} and λ_{wet} are roughly unchanged. **(b)** Under a fixed $\gamma/\Pi_0 L_0$, a larger $\gamma/\mu L_0$ widens the gap between λ_{dry} and λ_{wet}, both λ_{dry} and λ_{wet} converges to 1 if $\gamma/\mu L_0$ is extremely small.

3. The swelling-dependent Poisson's ratio

According to the Flory-Rehner model, the free energy density $W(F) = W_{ela} + W_{mix}$, where:

$$W_{ela} = \frac{NkT}{2}\left(\lambda_1^2 + \lambda_2^2 + \lambda_3^2 - 3 - 2\log\left(\lambda_1\lambda_2\lambda_3\right)\right), \tag{S1}$$

$$W_{mix} = \frac{kT}{\Omega}\left((\lambda_1\lambda_2\lambda_3 - 1)\log\left(\frac{\lambda_1\lambda_2\lambda_3 - 1}{\lambda_1\lambda_2\lambda_3}\right) - \chi\frac{1}{\lambda_1\lambda_2\lambda_3}\right). \tag{S2}$$

Here $\lambda_1, \lambda_2, \lambda_3$, are the principle stretches, N is the number of polymer chains per unit volume in the dry state, kT is the temperature in the unit of energy, Ω is the volume per solvent molecule, χ is a parameter describing the interaction between the solvent and the polymer. Next, we apply a volumetric strain $\varepsilon = \delta V/3V$ and expand the following expressions to the linear order of ε:

$$\frac{dW_{ela}}{d\varepsilon} = NkT\left(\left(\lambda_1^2 + \lambda_2^2 + \lambda_3^2\right) - 3\right) + NkT\varepsilon\left(\lambda_1^2 + \lambda_2^2 + \lambda_3^2 + 3\right), \tag{S3}$$

$$\frac{dW_{mix}}{d\varepsilon} = \frac{kT}{\Omega}\left(3 + 3J\,log\left(\frac{J-1}{J}\right) + \frac{3\chi}{J}\right) + \frac{kT}{\Omega}\varepsilon\left(-3 + \frac{9J}{J-1} + 6J\,log\left(\frac{J-1}{J}\right) - \frac{12\,\chi}{J}\right). \tag{S4}$$

Here $J = \lambda_1\lambda_2\lambda_3$.

The hydrostatic pressure on the gel is $p = \frac{1}{3}\frac{\partial W}{\partial\varepsilon} = p_0 + K\varepsilon$, where K is the bulk modulus. Then,

$$K = \frac{NkT\left(\lambda_1^2 + \lambda_2^2 + \lambda_3^2 + 3\right)}{3} + \frac{kT}{\Omega}\left(-3 + \frac{9J}{J-1} + 6J\,log\left(\frac{J-1}{J}\right) - \frac{12\,\chi}{J}\right). \tag{S5}$$

The shear modulus $G = NkT/J^{1/3}$[1], then the Poisson's ratio can be calculated by:

$$\nu = \frac{3K - 2G}{2(3K + G)}. \tag{S6}$$

In general, ν depends on both the swelling ratio J and the ratio between $\lambda_1, \lambda_2, \lambda_3$ in addition to the network parameters $N\Omega$ and χ. In Fig. S3, we plot the swelling dependent ν for different material properties. For typical hydrogels $\chi < 0.5, N\Omega < 1$, the Poisson's ratio is close to 0.5.
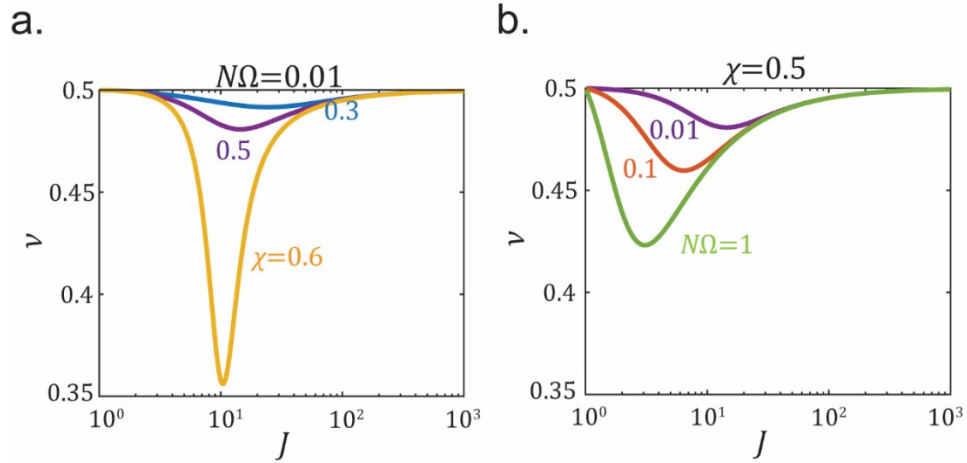
Figure S3. The swelling-dependent Poisson's ratio under **(a)** different polymer-solvent interaction parameter χ and **(b)** different crosslinking density $N\Omega$.

Reference

1. Li, J., et al., *Experimental determination of equations of state for ideal elastomeric gels.* Soft Matter, 2012. **8**(31): p. 8121-8128.