

Supplementary Materials for
Controlling thermoreversibility and hole conductivity in thermoresponsive ionic biogels using phase morphology for neurohaptics

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Sci. Adv. **12**, eace0777 (2026)
DOI: 10.1126/sciadv.aee0777

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Supplementary Note 1: Insights on the material design of Ionic Biogel

Thermodynamics of Ionic Biogel

Both the nucleated and bicontinuous systems comprise two distinct polymer networks: the PEDOT:PSS/ionic liquid network (Network A – ionic phase) stabilized by electrostatic interactions, and the gelatin (or agarose)/glycerol/salt network (Network B – biogel phase) stabilized by hydrogen bonding (**Figure S2a**). In the ionic system (PEDOT:PSS/EMIM-TFSI), the ionic liquid (EMIM-TFSI) acts as a strong plasticizer, profoundly influencing the electrostatic interactions between the PEDOT and PSS chains (40). The EMIM⁺ cations and TFSI⁻ anions intercalate between the PEDOT and PSS chains, effectively screening the Coulombic attractions and reducing the electrostatic binding energy. This screening leads to a softer polymeric backbone with extended hole delocalization. Concurrently, the biogel phase (gelatin-glycerol-salt) hydrogel matrix is stabilized through an extensive hydrogen-bonding network (3, 4). Gelatin forms triple-helical structures connected via hydrogen bonds between peptide backbone amide groups. The addition of glycerol introduces multiple hydroxyl groups capable of forming hydrogen bonds with both gelatin and water molecules, enhancing the stretchability and reversibility of the gel-sol transition of the hydrogel. The presence of salts, such as NaCl, can further modulate the hydrogen-bonding network by influencing the hydration shell and ionic strength, which not only affects the ionic conductivity but also the gelation kinetics and thermal responsiveness (3, 4).

After mixing all the components of the ionic biogel (described in the Methods section), both nucleated and bicontinuous ionic biogel system forms an amorphous state due to kinetic constraints of the biogel phase at room temperature (3, 4). Upon cooling to room temperature (after heating), the immiscibility between PEDOT and ionic liquid favours phase separation. However, changing the mixing sequence and thereby the modulus of the biogel phase, phase separation occurs in two distinct pathways: 1) liquid – liquid phase separation (LLPS) in nucleated systems with isolated PEDOT domains and 2) viscoelastic phase separation (VPS) in bicontinuous systems. All the thermodynamic potentials such as Gibbs' free energy and thermodynamic quantities such as Flory-Huggins's parameter are path-independent and only state functions. Therefore, the change of phase morphology due to the mixing-sequence can only result from some non – equilibrium dissipative phenomenon such as the difference in kinetics of the two ionic biogel system – nucleated (high mobility M_ϕ of the biogel phase) and bicontinuous (low mobility M_ϕ of the biogel phase during mixing) (32, 39). Intuitively, for the nucleated system, high mobility M_ϕ of the biogel phase brings the system into thermal equilibrium as the entropy is maximized to create distinct individual PEDOT phases, promoting complete phase separation, typically observed in LLPS, where compartmentalization arises from weak, dynamic interactions (24, 30). With the increase in the polymeric concentration in the nucleated system, the PEDOT phases tend to get more populated yet remain nucleated (**Figure S2b**). In comparison, for bicontinuous systems, low phase mobility M_ϕ of the amorphous biogel phase restricts the complete mixing and creates a percolated PEDOT phase with large phase boundaries, which increases the total interfacial enthalpy along with mixing enthalpy.

Viscoelastic Phase Separation (VPS) and Liquid – Liquid Phase Separation (LLPS) in Ionic Biogels Inspired by Membraneless Organelles

In LLPS, multivalent proteins and RNAs demix into droplets that behave as dynamic liquids: they create spherical boundary due to surface tension, coalesce rapidly, exchange components with the surroundings, and remodel composition in response to signalling. This fluidity underpins functions that require rapid turnover and selective concentration, such as RNA processing in stress granules and nucleolar ribosome biogenesis, where weak, reversible interactions create a dense phase without long-lived mechanical memory.

Biological function demands organizing chromosomes, buffering forces, or templating reactions, the crowded, crosslinked milieu drives condensates into regimes where viscoelasticity constrains their evolution. In VPS, one component or the surrounding milieu relaxes far more slowly than its partner, so demixing is coupled to the buildup and release of internal stress. The result is qualitatively different morphologies and kinetics: strands and space-spanning networks form, coarsening is delayed or partially arrested, and shape anisotropy persists. Tanaka's framework rationalizes these outcomes in terms of dynamic asymmetry between slow, network-forming macromolecules (for example, long RNAs or low-complexity protein polymers). Cells naturally realize this asymmetry through regulated multivalency, post-translational modification, and ATP-driven remodelling that tune the lifetime and connectivity of transient crosslinks. VPS thus provides a route to mechanically robust scaffolds and long-lived microenvironments, when retaining shape, resisting coalescence, or storing elastic energy is advantageous.

VPS

Following Tanaka (27), we treat the mixture as two interpenetrating continua with volume fraction $\phi(\mathbf{x}, t)$ of component 1 (the slow, stress-bearing component) and velocities $\mathbf{v}_1, \mathbf{v}_2$ for components 1 and 2. The volume-averaged velocity is $\mathbf{v} = \phi \mathbf{v}_1 + (1 - \phi) \mathbf{v}_2$ and incompressibility imposes $\nabla \cdot \mathbf{v} = 0$. Tanaka summarizes the viscoelastic model by the coupled equations:

$$\frac{\partial \phi}{\partial t} = -\nabla \cdot (\phi \mathbf{v}) - \nabla \cdot [\phi(1 - \phi)(\mathbf{v}_1 - \mathbf{v}_2)], \quad (\text{S1})$$

$$\mathbf{v}_1 - \mathbf{v}_2 = -\frac{1-\phi}{\zeta(\phi)} \left[\nabla \cdot \mathbf{\Pi} - \nabla \cdot \boldsymbol{\sigma}^{(1)} + \frac{\phi}{1-\phi} \nabla \cdot \boldsymbol{\sigma}^{(2)} \right], \quad (\text{S2})$$

$$\rho \frac{\partial \mathbf{v}}{\partial t} \simeq -\nabla \cdot \mathbf{\Pi} + \nabla p + \nabla \cdot \boldsymbol{\sigma}^{(1)} + \nabla \cdot \boldsymbol{\sigma}^{(2)}. \quad (\text{S3})$$

Here $\zeta(\phi)$ is the friction coupling the relative motion to stresses, $\mathbf{\Pi}$ is the osmotic stress derived from the mixing free energy (e.g., Flory–Huggins plus gradient penalty), and $\boldsymbol{\sigma}^{(k)}$ are the viscoelastic stresses carried by each component. The Maxwell-type viscoelasticity with composition-dependent plateau moduli $G_{S,B}(\phi)$ and relaxation times $\tau_{S,B}(\phi)$:

$$\frac{\partial \boldsymbol{\sigma}_j}{\partial t} + (\mathbf{v} \cdot \nabla) \boldsymbol{\sigma}_j = \mathbf{D} \cdot \boldsymbol{\sigma}_j + \boldsymbol{\sigma}_j \cdot \mathbf{D}^T - \frac{1}{\tau_j(\phi)} \boldsymbol{\sigma}_j + G_j(\phi)(\mathbf{D} + \mathbf{D}^T), j \in \{S, B\}, \quad (\text{S4})$$

where $\mathbf{D} = \nabla \mathbf{v}_r$ is the deformation-rate tensor for the rheologically relevant velocity $\mathbf{v}_r$, S, B subscripts denotes the shear and bulk metric. The thermodynamic driving force still enters through $-\nabla \cdot \mathbf{\Pi}$, but pattern selection is modified by stress–diffusion coupling in Eq. (S1) and by force balance in Eq. (S3).

LLPS as a special case of VPS (fluid limit of the viscoelastic model)

Tanaka shows how the viscoelastic model reduces to the familiar fluid description when stresses relax rapidly. Under dynamic symmetry (trivial stress division) the viscoelastic model reduces to a “symmetric viscoelastic model,” with kinetics:

$$\frac{\partial \phi}{\partial t} = -\nabla \cdot (\phi \mathbf{v}) + \nabla \cdot \left[\frac{\phi(1-\phi)^2}{\zeta} \nabla \cdot \boldsymbol{\Pi} \right], \quad (\text{S5})$$

$$\rho \frac{\partial \mathbf{v}}{\partial t} \simeq -\nabla \cdot \boldsymbol{\Pi} + \nabla p + \nabla \cdot \boldsymbol{\sigma}. \quad (\text{S6})$$

Finally, in the fluid limit where deformation is slow compared with internal rheological times, the viscoelastic stress reduces to a Newtonian viscous stress, $\nabla \cdot \boldsymbol{\sigma} = \eta \nabla^2 \mathbf{v}$, yielding the classical fluid model (model H):

$$\frac{\partial \phi}{\partial t} = -\nabla \cdot (\phi \mathbf{v}) + \nabla \cdot \left[\frac{\phi(1-\phi)^2}{\zeta} \nabla \cdot \boldsymbol{\Pi} \right], \rho \frac{\partial \mathbf{v}}{\partial t} \simeq -\nabla \cdot \boldsymbol{\Pi} + \nabla p + \eta \nabla^2 \mathbf{v} \quad (\text{S7})$$

Equivalently, Tanaka presents model H in the familiar form $\partial_t \phi = -\nabla \cdot (\phi \mathbf{v}) + L \nabla^2 \mu$ with

$$\rho \partial_t \mathbf{v} \simeq \mathbf{F}_\phi - \nabla p + \eta \nabla^2 \mathbf{v} \quad (\text{S8})$$

where

$$\mathbf{F}_\phi / k_B T = -\nabla \cdot \boldsymbol{\Pi} = -\phi \nabla \mu \quad (\text{S9})$$

This is the regime in which LLPS morphology is governed by capillarity and diffusion: off-critical mixtures relax toward droplet morphologies (nucleation–growth in the metastable region), and the standard droplet coarsening laws apply.

VPS and LLPS leading to different phase morphology and rheology

Though both systems have phase separation akin to their positive mixing free energy, the difference in the phase morphology occurs due to non-dissipative forces like kinetics and interfacial forces. Though mixing free energy indicates the phase separation, other free energy needs to be considered. The total Gibbs free energy F for a nucleated system (LLPS - based) is given as:

$$F = \int \frac{\kappa}{2} |\nabla \phi|^2 + f_{elastic}(\phi) + f_{mix}(\phi) dV, \quad (\text{S10})$$

where

$$f_{mix}(\phi) = RT \left(\frac{\phi_A}{N_A} \ln \phi_A + \frac{\phi_B}{N_B} \ln \phi_B + \chi \phi_A \phi_B \right) \quad (\text{S11})$$

and

$$f_{elastic}(\phi) = \frac{G}{4} (\mathbf{W} - \mathbf{I}) : (\mathbf{W} - \mathbf{I}) + G_{biogel}. \quad (\text{S12})$$

In the above equation, G is the shear modulus of the polymeric network given by $G \sim G_0 \phi_p^3$ and $\mathbf{I}$ is the identity matrix, G_{biogel} is the elastic free energy due to the gelatin-based matrix, and $\mathbf{W}$ is the conformation matrix due to the polymeric matrix given by

$$\mathbf{W} \equiv \frac{1}{n_p} \left[\sum_{\ell=1}^{n_p} \frac{1}{(Z_\ell-1)} \sum_{m=1}^{Z_\ell-1} \frac{\mathbf{R}_m^\ell \mathbf{R}_m^\ell}{3a^2 N_e} \right], \quad (\text{S13})$$

where n_p is the number of polymeric chains and ℓ th polymer chain has Z_ℓ number of entangled segments with position vectors $\mathbf{R}_m^\ell$ directed from $\mathbf{r}_m^\ell$ to $\mathbf{r}_{m+1}^\ell$. At equilibrium, the average length of $\mathbf{R}_m^\ell$ is $a\sqrt{N_e}$, where N_e is the average number of segments existing between the two adjacent entangled segments along a chain. However, for a nucleated system, $n_p = 0$ as it lacks polymeric networks, making $\mathbf{W} - \mathbf{I} = \mathbf{0}$.

In comparison, for bicontinuous (VPS-based)

$$F = \int \frac{\kappa}{2} |\nabla \phi|^2 + f_{mix}(\phi) + f_{elastic}(\phi) dV, \quad (\text{S14})$$

where κ is the interfacial parameter. In this case, $\mathbf{W} - \mathbf{I} \neq \mathbf{0}$, and there is a trade-off between polymeric elastic free energy and G_{biogel} , with a high concentration of PEDOT:PSS resulting in $\frac{G}{4}(\mathbf{W} - \mathbf{I}):(\mathbf{W} - \mathbf{I}) \gg G_{biogel}$. The loss tangent for the bicontinuous system w.r.t. to gelatin-based matrix can be given as

$$\tan \delta_{\text{Bicontinuous}} = \frac{\Im \left\{ \frac{G}{4} (\mathbf{W} - \mathbf{I}):(\mathbf{W} - \mathbf{I}) \right\}}{\Re \left\{ \frac{G}{4} (\mathbf{W} - \mathbf{I}):(\mathbf{W} - \mathbf{I}) \right\}}. \quad (\text{S15})$$

The chemical potential μ is given as

$$\mu = \frac{\delta F}{\delta \phi} = \frac{\partial f}{\partial \phi} - \nabla \cdot \frac{\partial f}{\partial (\nabla \phi)}, \quad (\text{S16})$$

where F is related to the total Gibbs free energy and the total stress:

$$\boldsymbol{\sigma} = 2\mathbf{W} \cdot \frac{\delta F}{\delta \mathbf{W}} + G_{biogel} = 2G\mathbf{W} \cdot (\mathbf{W} - \mathbf{I}) + G_{biogel}. \quad (\text{S17})$$

The dynamics of these phases are given by:

$$\frac{\partial \phi}{\partial t} = \nabla \cdot \left(M_\phi \left(\nabla \mu - \frac{\nabla \cdot \boldsymbol{\sigma}}{\phi} \right) \right), \quad (\text{S18})$$

where M_ϕ is the mobility and considering that the mean velocity is zero. The spinodal line is given by the polymeric composition when $\nabla \mu = 0$ and the spinodal decomposition or unstable thermodynamic state occurs when $\nabla \mu < 0$. For LLPS, $\nabla \cdot \boldsymbol{\sigma} = \mathbf{0}$, and only mixing energy or osmotic stress is critical, whereas for VPS, stress forces like friction between the polymer and the biogel matrix play a critical role in phase morphology.

The molecular interactions between the biogel phase and the ionic phase for both nucleated and bicontinuous systems can be elucidated from a comprehensive series of characterizations.

Material Characterization of Ionic Biogel

X-ray photoelectron spectroscopy

X-ray photoelectron spectroscopy (XPS) analysis of the ionic biogel (nucleated 35 w/w%) reveals the presence of key carbon, nitrogen, and sulphur species, including CH_x (25.6%), $\text{C}-(\text{O}, \text{N})$ (27.4%), and SO_3 (6.2%) (**Figure S3**). The detection of sulfonate (SO_3) groups and thiols confirms the functionality of PEDOT:PSS, while fluorocarbon and siloxane signals suggest interactions with the ionic liquid EMIM-TFSI. Additionally, the presence of protonated amines ($\text{N}-\text{C}^+$, 2.9%) and carbonyl groups ($\text{C}=\text{O}$, 5.1%) underscores the role of the gelatin-glycerol matrix in stabilizing the ionic and electronic interfaces.

Raman spectroscopy

Raman spectroscopy at different temperatures was carried out for the ionic biogel with nucleated 35 w/w% (**Figure S4a**) and bicontinuous 100 w/w% (**Figure S4b**) systems. For the nucleated system, the Raman spectra reveal that the $\text{C}_\alpha=\text{C}_\beta$ symmetric stretching vibration peaks of PEDOT exhibit a red shift from 1423 cm^{-1} at 25°C and 30°C to 1417 cm^{-1} at 40°C and 50°C , with an accompanying increase in intensity up to 40°C , followed by a decrease at 50°C post-phase transition. This shift indicates that a higher proportion of benzoid moieties in PEDOT is converted to the quinoid structure due to oxidative charge transfer doping at elevated temperatures, resulting in a more planar polymer backbone (40). Concurrently, the Amide I band associated with the $\text{C}=\text{O}$ stretching vibrations in gelatin increases in intensity from 25°C to 40°C and then diminishes at 50°C , suggesting the formation and subsequent disruption of intermolecular hydrogen bonding interactions between gelatin and glycerol molecules. These findings indicate that elevated temperatures promote both the unwrapping of gelatin's helical structure and the planarization of PEDOT chains, leading to synergistic enhancements in the electrical and mechanical properties of the ionic biogel.

Fourier-transform infrared spectroscopy

The Fourier-transform infrared spectroscopy (FTIR) spectra reveal strong characteristic infrared absorption peaks of thiophene functional groups at $1,640\text{ cm}^{-1}$, with a markedly higher absorption intensity observed during heating for nucleated 35 w/w% (**Figure S5a**), 100 w/w% (**Figures S5b,e**) and bicontinuous 100 w/w% (**Figures S5c,f**), 200 w/w% (**Figures S5d,g**). This increase in absorption intensity at elevated temperatures suggests enhanced vibrational activity of the thiophene rings within the PEDOT chains, potentially due to increased conjugation length arising from thermal effects. Such changes may facilitate greater π -electron delocalization and contribute to the improved charge mobility or conductivity of the ionic biogel at higher temperatures. The Amide A band—corresponding to the $\text{N}-\text{H}$ and $\text{O}-\text{H}$ stretching vibrations exhibits a blue shift from $3,287\text{ cm}^{-1}$ at 25°C to $3,317\text{ cm}^{-1}$ at 45°C , along with notable peak broadening. This shift to higher wavenumbers indicates a decrease in hydrogen bonding interactions within the gelatin-glycerol-salt hydrogel matrix as temperature increases. The disruption of hydrogen bonds by thermal agitation strengthens the $\text{N}-\text{H}$ bonds, causing them to absorb at higher frequencies (blue shift). The observed peak broadening suggests increased molecular disorder and a wider distribution of hydrogen-bonding environments resulting from the enhanced thermal motion of gelatin chains (4).

Atomic force microscopy

The atomic force microscopy (AFM) images reveal a hierarchical porous architecture featuring interconnected pores that range from nanometres to micrometres in size for both nucleated (35 w/w%) (**Figure S6a**) and bicontinuous (100 w/w%) (**Figure S6b**) systems. This hierarchical porosity arises from the thermodynamically driven phase separation between the hydrophilic

gelatin-glycerol network and the more hydrophobic PEDOT:PSS/EMIM-TFSI domains during the gelation process.

Micro-computed tomography

The phase-separation can also be observed in micro-computed tomography for both nucleated (100 w/w%) (**Figure S7a**) and bicontinuous (100 w/w%) (**Figure S7b**) due to the difference in optical density of the ionic phase and gelatin-based biogel phase.

Room-temperature UV-Vis

The UV-Vis-NIR spectra exhibit a significant redshift, with the characteristic absorption peak of PEDOT appearing around ~950 nm (bicontinuous 100 w/w%) instead of the typical ~800 nm observed in pristine PEDOT films, whereas in nucleated 35 w/w%, there is no obvious peak (**Figure S8**). This pronounced shift to longer wavelengths indicates an increase in the effective conjugation length and enhanced delocalization of π -electrons along the PEDOT backbone within the ionic biogel matrix (40).

Wide-angle X-ray scattering

The wide-angle X-ray scattering (WAXS) patterns reveal notable temperature-dependent changes in the crystalline ordering of the PEDOT:PSS domains within the biogel matrix for nucleated (35 w/w%) (**Figures S9a,b**) and bicontinuous (100 w/w%) (**Figure S9c**) systems. Specifically, for nucleated 35 w/w%, the (200) d-spacing, corresponding to the scattering vector $q_z \sim 0.4 \text{ \AA}^{-1}$, exhibits an increase from 0.011 (a.u.) to 0.014 (a.u.) as the temperature is elevated from 25°C to 40°C, whereas no obvious change is observed for the bicontinuous system. The peak at $q_z \sim 0.87 \text{ \AA}^{-1}$ in WAXS patterns for the bicontinuous system, previously assigned to the stacking of PEDOT chains along the π - π direction, remains consistent in intensity and position at 25°C to 60°C, indicating stable π - π stacking interactions within the PEDOT domains across this temperature range. The unchanged ordering suggests that the planarization and increased conjugation of the PEDOT chains at elevated temperatures up to 60°C do not disrupt the overall stacking arrangement but may enhance charge transport by improving orbital overlap.

Biocompatibility

EMIM-TFSI and related imidazolium ionic liquids can exhibit toxicity in their neat form. Therefore, evaluating skin compatibility and biocompatibility of the formulated ionic biogel is essential for wearable use. In our material, EMIM-TFSI is incorporated within a physically crosslinked gelatin-glycerol matrix and a PEDOT:PSS-rich phase, which together create a mechanically stable composite expected to eliminate the activity and transport of freely mobile ionic-liquid species compared to the neat ionic liquid. Applying the nucleated 35 w/w% ionic biogel to the forearm skin of human subjects for one week evaluates the skin compatibility. Throughout the application period, no signs of skin irritation, inflammation, or adverse reactions are observed (**Figure S10a**). The biocompatibility of the ionic biogel is also confirmed with the MTT assay (**Figure S10b**). The cell viability of 89% and 110% in 24 h and 48 h is comparable to that of 100% and 127% from the control, supporting the proliferative effects of hydrogels on MCF-10A cells (**Figure S10c**). The demonstrated biocompatibility is critical to prevent adverse immune responses and ensure the longevity of the device.

Supplementary Note 2: Controlling the Gel–Sol Transition Temperature in Ionic Biogel System

Thermoreversible gels for through-hair neural interface must pass through the rheological crossover $\tan \delta = 1$, so they can be printed or extruded in the sol state and rapidly recover an adhesive gel once in contact with the scalp. This crossover temperature, T_{gel} , can be controlled by balancing the enthalpic strength and kinetic lifetime of hydrogen bonds that knit the matrix, against the geometric confinement imposed by the characteristic size ξ of the PEDOT:PSS domains that may isolate (nucleated system) or percolate (bicontinuous) to provide electronic conduction. At the gel point T_{gel} , the storage and loss moduli become equal, giving $\tan \delta = \frac{G''}{G'} = 1$ at a representative frequency ω (e.g., 1 Hz) (3, 4).

The relaxation time $\tau(T)$ follows the Arrhenius relation:

$$\tau(T) = \tau_0 e^{-\frac{E_a}{RT}}, \quad (\text{S19})$$

where E_a is the activation energy of the hydrogen bond breaking. Therefore, the gel crossover point T_{gel} yields:

$$T_{\text{gel}} \sim \frac{E_a}{R \ln \frac{\tau_0}{\tau(T_{\text{gel}})}}. \quad (\text{S20})$$

where τ_0 is the reference time constant and $\tau(T_{\text{gel}})$ is a fixed time constant for a particular frequency. Optimizing T_{gel} requires controlling the activation energy for hydrogen bond breaking at a constant probing frequency.

Gelatin matrices gel via cooperative hydrogen bonds ($\Delta H_h \approx 5\text{--}20 \text{ kJ}\cdot\text{mol}^{-1}$), so that $E_a \approx N_h \Delta H_h$ scales with hydrogen bond junction density N_h . Introduction of H-bond disruptors (e.g., glycerol and ionic liquids) lowers E_a and depresses T_{gel} , whereas increasing gelatin or water content (from PEDOT:PSS) raises E_a and hence T_{gel} . Moreover, using hydrophobic DES increases the hydrogen bond donor for increased hydrogen bond junction density, which increases E_a and hence T_{gel} to result in gel phase only. In comparison, hydrophilic DES and EMIM-OTF are soluble in PEDOT:PSS to hinder the hydrogen bonding from water, resulting in lowered E_a and decreased T_{gel} . Moreover, for bicontinuous systems, percolated PEDOT:PSS domains with an equivalent length ξ impose large geometric drag on network dynamics, with the characteristic geometric relaxation time scaling as:

$$\tau(T_{\text{gel}}) \propto \frac{\xi^2}{D(T_{\text{gel}})}. \quad (\text{S21})$$

where $D(T_{\text{gel}})$ is the diffusion constant of the matrix at temperature T_{gel} . Experimentally, increasing PEDOT:PSS/IL concentration first raises T_{gel} due to mild dehydration and interfacial crosslinking (increased E_a), but at higher polymeric loadings, network plasticization lowers E_a and decreases T_{gel} . Combining both effects gives:

$$T_{gel} \sim \frac{E_a}{R \ln \left(\frac{\tau_0}{\alpha \frac{\xi^2}{D(T_{gel})}} \right)}, \quad (S22)$$

where α is a proportional constant. In nucleated systems, E_a is more dominated and is influenced by both the concentration of PEDOT:PSS and the type of ionic liquids. In comparison, for bicontinuous systems, percolation length ξ becomes for dominated, which primarily depends on the concentration of PEDOT:PSS.

Rheological Characterization of Nucleated 35 w/w% Ionic Biogel

Temperature and time are critical factors in modulating and restoring the mechanical properties of thermoresponsive viscoelastic materials (3, 4, 29). The viscosity transitions from ~ 1.6 kPa·s at room temperature to ~ 18 Pa·s at high temperature (50°C) and then back to ~ 940 Pa·s at room temperature in 15 mins (**Figure S14a**). The loss modulus decreases from 490 Pa at room temperature to 210 Pa at 40°C , which marks the transition temperature of the ionic biogel as the loss modulus equals the storage modulus. In comparison, the gelatin-glycerol-salt composite biogel without PEDOT:PSS and ionic liquid exhibits a higher transition temperature of around 45°C . The higher storage modulus of 19 kPa (and loss modulus of 2.1 kPa) at room temperature reduces, but still to a relatively higher storage (and loss) modulus of 800 Pa at 45°C (**Figure S14b**). The change of loss tangent with temperature is greater and earlier for ionic biogel with 0.25 1/K at $\sim 39^\circ\text{C}$ than biogel with 0.1 1/K at $\sim 43^\circ\text{C}$. All the measurements are taken at 1 Hz and 0.05% shear strain. The material exhibits a shear-thinning property having a solid-liquid transition ($\tan \delta = 1$) at 190% shear strain with a dynamic viscosity of 400 Pa·s, sharply decreased from 1.3 kPa·s at 10% shear strain (**Figure S14c**). With the increase in frequency, the loss tangent $\tan(\delta)$ increases from 0.06 at 1 Hz to 2.4 at 100 Hz, and the viscosity decreases linearly from 7.7 kPa·s at 0.1 Hz to 11 Pa·s at 100 Hz (**Figure S14d**).

Repeated Rheological Characterization of Nucleated 35 w/w% Ionic Biogel

In this experiment, the temperature is ramped up from room temperature to 60°C at a rate of $5^\circ\text{C}/\text{min}$ and back to room temperature, along with holding for a 15-minute equilibrium period before repeating the cycle (**Figure S14e**). The loss tangent $\tan(\delta)$ increases from 0.1 at room temperature to 2.2 at 60°C , and then to 0.9 at room temperature. Keeping the material at room temperature for 15 minutes decreases the loss tangent from 0.9 to 0.2. In the second temperature cycle, the loss tangent increases to 1.7 at 60°C and recovers to 0.9 at room temperature again. Similarly, the dynamic viscosity decreases rapidly from 1.7 kPa·s at room temperature to 10 Pa·s at 60°C and then increases to 200 Pa·s at room temperature, followed by a further increase to 950 Pa·s after keeping it isothermal for 15 mins (**Figure S14f**). During the second cycle, the viscosity decreases to 30 Pa·s at 60°C and increases back to 210 Pa·s at room temperature. The storage modulus reduces from 6 kPa at room temperature to 110 Pa at 60°C for the second ramp-up, whereas the loss modulus reduces from 1.1 kPa at room temperature to 190 Pa at 60°C (**Figures S14g, h**). A similar pattern is observed during the temperature ramp-down, where both loss and storage moduli increase with decreasing temperature. For example, in the second ramp, storage (or loss) modulus increases from 60 (or 110) Pa at 60°C to 1 kPa at room temperature, probably due to metastability and a high efficiency of recovery for the gelatin chains to re-establish

hydrogen bonds. After the first cycle to remove excess water, the viscosity normalized by the viscosity at the end of the 2nd isothermal process remains almost unchanged (with only $\pm 3\%$ variation) at the end of the isothermal process after additional cycles ($N=1, 2$, and 3), showcasing a thermoreversible material (with minimal thermal fatigue) with metastable characteristics. However, there is a 10% increase in the normalized viscosity at the end of each ramp-down process (or at the start of the isothermal process), possibly due to the slower recovery time of the formation of hydrogen bonds (but with high efficiency) or altering the alignment of distinct phases. This result indicates that the recovery or recombination of hydrogen bonding (larger time constant) during the ramp-down exhibits a larger thermal hysteresis than the breaking of the hydrogen bonding (smaller time constant) of the gelatin chains during the ramp-up. A very similar trend is also observed in the ~ 3 -month-old sample kept at room temperature with only $\pm 4\%$ variation at the end of the isothermal process for $N=1, 2$, and 3 (**Figure S14i**).

Computational Model of Temperature-dependent Rheology using Spring-Damper

Viscoelastic materials exhibit both elastic (storage) and viscous (loss) characteristics, which are traditionally modelled using fundamental spring-damper systems, as in the Maxwell and Kelvin-Voigt models. The Maxwell Model, arranged in series, is well-suited for describing time-dependent stress relaxation, capturing the loss modulus under constant strain as the material dissipates energy over time. In contrast, the Kelvin-Voigt Model, arranged in parallel, effectively represents the creep recovery of the material, where the elastic response under constant stress can be quantified by the storage modulus, especially in cyclic loading scenarios. While these classic models provide foundational insights into viscoelastic behaviours, they require adaptation to accurately represent thermoresponsive materials, where changes in the modulus are temperature-dependent, and shear-thinning behaviour induces nonlinear dependencies on both temperature and shear rate.

The Kelvin-Voigt model combines a spring and a damper in parallel, describing a material that behaves elastically when subjected to a constant strain:

$$\sigma(t) = E\varepsilon(t) + \eta \frac{\partial \varepsilon(t)}{\partial t}, \quad (\text{S23})$$

where $\sigma(t)$ is the stress, $\varepsilon(t)$ is the strain, E is the elastic modulus of the individual spring, and η is the viscosity of the individual damper. For materials that exhibit complex, multi-stage recovery of storage and loss moduli, the Kelvin-Voigt model can be generalized by introducing several spring-damper units in parallel, each with distinct moduli (E_i) and viscosities (η_i):

$$\sigma(t) = \sum_{i=1}^N E_i \varepsilon(t) + \eta_i \frac{\partial \varepsilon(t)}{\partial t}. \quad (\text{S24})$$

Each spring-damper unit contributes to a unique time constant $\tau_i = \eta_i / E_i$, allowing the model to simulate various rates of recovery. The storage modulus (G') and loss modulus (G'') can then be expressed as:

$$G'(t, T) = \sum_{i=1}^N [E_i e^{-t/\tau_i}], \quad (\text{S25})$$

$$G''(t, T) = \sum_{i=1}^N \left[\frac{\eta_i}{\tau_i} e^{-\frac{t}{\tau_i}} \right]. \quad (\text{S26})$$

For the ionic biogel, viscosity $\eta_i(T)$ typically decreases with temperature T . However, the rheological parameters such as viscosity and modulus are not symmetrical, probably due to the difference in time scales of breaking and formation of hydrogen bonds; therefore, the time constants for temperature ramp up (τ_{breaking}) and down (τ_{recovery}) are different. At a constant shear rate (1 Hz), shear strain (0.05%), and temperature rate ($\pm 5^\circ\text{C}/\text{min}$) (positive or negative for increase or decrease in temperature with zero for the isothermal condition), the dynamics of rheological properties under cyclic heating can be modelled with two distinct time constants:

- Time constant for breaking hydrogen bonds (τ_{breaking}), which describes a rapid reduction in viscosity and modulus upon heating;
- Time constant for recovering hydrogen bonds (τ_{recovery}), which governs the increased viscosity and modulus upon cooling, describing the reformation or "healing" of the structure.

To capture these behaviours, the generalized Kelvin-Voigt model with parallel spring-damper elements can be extended, with temperature-dependent moduli ($E_i(T)$) and viscosities ($\eta_i(T)$) that integrate decay/recovery time constants into the storage and loss modulus equations for each element. The moduli $G'(t, T)$ and $G''(t, T)$ can be extended as follows:

$$G'(t, T) = \sum_{i=1}^N \left[E_i(T) e^{-t/\tau_{\text{breaking}}} \mathcal{H}(t_{\text{breaking}} - t) + E_i(T) (1 - e^{-t/\tau_{\text{recovery}}}) \mathcal{H}(t - t_{\text{recovery}}) \right] \quad (\text{S27})$$

$$G''(t, T) = \sum_{i=1}^N \left[\frac{\eta_i(T)}{\tau_{\text{breaking}}} e^{-t/\tau_{\text{breaking}}} \mathcal{H}(t_{\text{breaking}} - t) + \frac{\eta_i(T)}{\tau_{\text{recovery}}} (1 - e^{-t/\tau_{\text{recovery}}}) \mathcal{H}(t - t_{\text{recovery}}) \right] \quad (\text{S28})$$

where $\mathcal{H}$ is the Heaviside step function, dividing the temperature ramp up ($t < t_{\text{breaking}}$) and down ($t > t_{\text{recovery}}$) phases.

Supplementary Note 3: Synergistic Thermodiffusion and Thermogalvanic Effect

The observed thermoelectric behaviour in the ionic biogel composite arises from the synergy of thermodiffusion and thermogalvanic effects, each of which has been studied separately in previous works (54). Thermogalvanic cells rely on temperature-dependent redox reactions, whereas thermodiffusion-based ionic thermoelectric devices leverage ion transport under thermal gradients. While both effects hold promise for low-grade heat harvesting, attempts to combine these effects within a single ionic thermoelectric (i-TE) system remain relatively limited. In nucleated 35 w/w%, the Seebeck coefficient reaches approximately 21 mV/K under a temperature difference of 0–3 K (dominated by the synergy of thermogalvanic and thermodiffusion) and decreases to about 4.6 mV/K for gradients between 3–8 K (probably dominated by either of the mechanisms). The observed behaviour can be attributed to a non-linear coupling of ion migration with redox-driven electrochemical potentials. The thermodiffusion mechanism, guided by the Soret effect, drives ions along the temperature gradient. In the reported composite, the gelatin–ionic liquid matrix provides a porous, quasi-solid environment that enhances ionic mobility. Simultaneously, the thermogalvanic mechanism leverages the temperature-dependent redox potential of the PEDOT:PSS component. In ionic biogel, the PEDOT:PSS domains undergo entropy-driven changes that, when aligned with ionic thermodiffusion, produce an additional voltage contribution. At lower temperature gradients (0–3 K), both the ionic and redox contributions combine effectively to generate higher thermoelectric voltages. This synergy decreases at larger temperature differences, possibly due to the saturation of ion migration pathways and a diminished redox response, leading to lower effective Seebeck coefficients. The resulting thermopower profile suggests that the composite's performance can be fine-tuned by the interplay between thermodiffusion and thermogalvanic effects, as well as by optimizing composition and nanostructure to enhance ionic selectivity and redox activity.

Supplementary Note 3: Mechanical Properties of Nucleated Ionic Biogel

The nucleated ionic biogel (35 w/w%) shows a smaller Young's Modulus of 33 ± 0.5 kPa compared to 71 kPa for the non-ionic biogel (**Figure S53a**). The ionic biogel could be stretched to 380% with a tensile strength of 84 ± 0.1 kPa. The self-healing property of the material allows the recovery of Young's modulus to 33 kPa as in pristine condition after cutting and healing at room temperature for 12 hours, but with lowered tensile strength (26 ± 0.2 kPa) and stretchability (120%). However, if the temperature is raised above the transition temperature, the material exhibits a 100% self-healing capability for both tensile strength and stretchability (as in the repeated temperature cycles). The material also shows an instantaneous self-healing property accelerated by its inherent adhesive property (**Figure S53b**). The ionic biogel has a very low shear modulus of 6.75 kPa (**Figure S53c**). However, the material exhibits a strain-stiffening effect for larger strain (>20%) in the compression test with a plateau modulus of ~ 50 kPa (**Figure S53d**).

Thermogravimetric analysis (TGA) is carried out by ramping the temperature at a rate of $5^\circ\text{C}/\text{min}$ to 150°C , followed by holding for 5 mins. The ionic biogel shows a much better water-retention property ($\sim 97\%$ at 70°C) compared to the normal biogel ($\sim 88\%$) (**Figure S53e**). The ionic biogel also retains about 91% of its weight in the first week and saturates around 89% till 3 weeks, whereas the biogel counterpart loses about 20% of its weight in the first week (**Figures S53f,g**). The hierarchically porous structure of the ionic biogel from phase separation also exhibits a high water vapour transmission rate (WVTR) of ~ 1417 g/m²/day, which is reasonably close to that of 1725 g/m²/day from the open bottle and more than the maximum transdermal water loss of 1050 g/m²/day (typically from axilla) (**Figure S53h**) as measured according to the ASTM E96-95 standard. As a result, the ionic biogel with high air and moisture permeabilities makes it particularly suitable for comfortable, long-term use on the skin surface in varying complex scenarios.

One of the most important benefits of phase-separation is the strain-insensitivity of the ionic biogel, which only increases its resistance by 100% after reaching its strain limit of 380%, which represents the record high strain insensitivity among all types of hydrogels to our knowledge (21, 44) (**Figure S53i**). The material shows noticeable changes (>1%) in resistance only after >30% strain. The material also shows negligible hysteresis, with resistance returning to its original value with only a 1% change.

Supplementary Note 4: Charge Injection Capacity and Wearable Stimulator

Charge injection capacity (CIC) is one of the most crucial parameters for evaluating the performance of electrical stimulation in neuroprosthetics (21). The CIC of the nucleated 35 w/w% ionic biogel is 10 mC/cm² at 33 Hz and 5 mC/cm² at 67 Hz (**Figure S54a**), which are comparable to existing materials used for similar applications (20). The CIC of ionic biogel is assessed by applying programmed current stimulation waveforms with an amplitude of 10 mA at 0.5 Hz using the galvanostatic mode of Autolab (**Figure S54b**). The high CIC indicates that the ionic biogel can inject higher amounts of charge without exceeding safe voltage limits or causing electrode polarization, thereby maintaining signal fidelity and reducing the risk of tissue damage. Besides the remarkable electrochemical stability, the ionic biogel also maintains a stable CIC value over prolonged charging and discharging cycles during cyclic CIC measurements, ensuring reliable performance during extended use (**Figure S54c**).

Wearable Stimulation Array

The large, stretchable patch designed on an adhesive, hydrophobic VHB tape exploits the ionic biogel as the skin-interfaced conductive medium for electrical stimulation (**Figure S54d**). By systematically varying the electrode pairs within the array, optimal configurations are identified for targeting the ulnar nerve to elicit haptic sensations localized near the pinkie finger region. While TENS of varied current amplitude in the range from 2 to 3.9 mA (depending on the pain threshold) at 150 Hz (duration of 3 s, pulse duration of 0.2 ms, and 450 number of pulses) is applied to the right arm, the corresponding force sensation is quantitatively measured on the left arm using a force sensor, allowing us to assess bilateral cortical processing of somatosensory inputs.

Supplementary Note 5: Perceived Sensation

On Day 1, the perceived force sensation is increased from 0.07 N to 0.5 N as the stimulation current applied to the ulnar nerve is increased from 3.1 mA to 3.9 mA, till the subject reports perceived pain (pain threshold) (**Figure S62**). Notably, on Day 2, the baseline force perception at the same stimulation current of 3.1 mA increases to 0.3 N, suggesting a possible sensitization or adaptation effect within the nervous system. A decrease in the pain threshold (current amplitude at which the pain onsets) is observed on Days 4 and 7, with similar force sensations of ~ 0.1 N being elicited at a significantly lower stimulation current of 2 mA. Specifically, on Day 4, a stimulation current of 3 mA can induce a sensation force that is about 842% of the value induced by the 3.1 mA stimulation on Day 1. Although the sensation force on Day 4 exhibits a similar trend and range to that observed on Day 7, there is a slight increase of $\sim 20\%$ at 3 mA (the higher end of the current range). These findings indicate potential neuroplastic changes occurring over repeated stimulation sessions, possibly involving peripheral sensitization mechanisms that enhance neural responsiveness to electrical stimuli (5, 6).

Supplementary Note 6: Multiphysics Modelling of Temperature-Driven Phase Field Flow and Electrochemical Performance of a Cylinder-Shaped Self-Balancing Electrode System

The ion transport within the ionic biogel and consequently the on-site amplification of the electrode signal is directly influenced by the ionic biogel's flow dynamics, which are governed by its viscosity, a parameter intrinsically linked to the local temperature distribution. These complex interdependencies are elucidated by the phase-field dynamics model of the ionic biogel and air at the interface between the electrode tip and the scalp, along with non-isothermal flow and transport of dilute species in COMSOL Multiphysics. The temperature at the electrode tip actively maintained at 320 K and the scalp temperature remained at approximately 300 K generate a thermal gradient. Coupled with the different thermal conductivities of the involved materials, a spatially and dynamically varying temperature field is created within the ionic biogel, which in turn modulates its viscosity according to a temperature-dependent relationship (**Figure S63a**). The use of the tip-shaped OECT starts with the contact of the ionic biogel with the scalp surface. The viscosity of the ionic biogel decreases with increasing temperature near the heated tip, which enhances the flow velocity v of the ionic biogel to flow more readily, displacing the air at the interface (**Figure S63b**). Furthermore, the ion transport within the ionic biogel is directly affected by these flow characteristics. The convection of ions due to the flow of ionic biogel, combined with diffusion and possible electrostatic interactions, leads to spatiotemporal variations in ion concentration $C(r, t)$, which is described by the convection-diffusion equation:

$$\frac{\partial C(r, t)}{\partial t} + v \cdot \nabla C(r, t) = D \nabla^2 C(r, t) + \frac{ze}{k_B T} D C \nabla \phi, \quad (\text{S29})$$

where D is the diffusion coefficient, z is the ionic valence, e is the elementary charge, k_B is the Boltzmann constant, v is the velocity, and ϕ is the electric potential (**Figure S63c**).

Equations Involved

Heat Transfer in Fluids and Solids (*Domain: Ionic Biogel, Air, and Tips*)

The governing equation for heat transfer in a fluid and solid domain is given by:

$$\rho C_p \frac{\partial T}{\partial t} + \rho C_p \mathbf{u} \cdot \nabla T = \nabla \cdot (k \nabla T) + Q, \quad (\text{S30})$$

where ρ is density, C_p is the specific heat capacity at constant pressure, T is temperature, $\mathbf{u}$ is the velocity vector, k is the thermal conductivity, and Q is the heat source term.

Non-Isothermal Laminar Flow (*Domain: Ionic Biogel*)

For incompressible laminar flow coupled with heat transfer, the Navier-Stokes equations are expressed as:

$$\rho \left(\frac{\partial \mathbf{u}}{\partial t} + (\mathbf{u} \cdot \nabla) \mathbf{u} \right) = -\nabla p + \nabla \cdot \mu (\nabla \mathbf{u} + \nabla \mathbf{u}^T) + \mathbf{F}, \quad (\text{S31})$$

$$\nabla \cdot \mathbf{u} = 0, \quad (\text{S32})$$

where p is pressure, μ is the temperature-dependent dynamic viscosity, and $\mathbf{F}$ is the external force (e.g., gravity or capillary force).

Phase Field Model (*Domain: Ionic Biogel and Air*)

The Cahn-Hilliard formulation used in the Phase Field module describes the evolution of the phase field variable ϕ , which distinguishes between fluid ($\phi = 1$) and air ($\phi = -1$). The governing equations are:

$$\frac{\partial \phi}{\partial t} + \mathbf{u} \cdot \nabla \phi = \nabla \cdot (M_\phi \nabla \mu_\phi), \quad (\text{S33})$$

$$\mu_\phi = \frac{\delta F}{\delta \phi} = \lambda \nabla^2 \phi + \frac{\partial f(\phi)}{\partial \phi}, \quad (\text{S34})$$

where M_ϕ is the mobility, $F = \frac{\lambda}{2} |\nabla \phi|^2 + f(\phi)$ is the free energy, $f(\phi) = \frac{\gamma}{4} (\phi^2 - 1)^2$ is the double-well potential, and γ is the surface tension parameter.

Electrostatics (*Domain: Ionic Biogel and Tissue*)

The Electrostatics module solves for the electric potential V using Poisson's equation:

$$\nabla \cdot (\epsilon \nabla V) = \Psi, \quad (\text{S35})$$

where ϵ is the permittivity of the medium and Ψ is the charge density.

Transport of Diluted Species (*Domain: Ionic Biogel and Tissue*)

The transport of ionic species in the ionic biogel can be modelled by the Nernst-Planck equation:

$$\frac{\partial C}{\partial t} + \nabla \cdot \mathbf{N} = R, \quad (\text{S36})$$

$$\mathbf{N} = -D \nabla C - \frac{zFD}{RT} C \nabla V + C \mathbf{u}, \quad (\text{S37})$$

where C is the concentration of species, D is the diffusion coefficient, z is the valence of the species, F is the Faraday constant, and R is the universal gas constant.

Supplementary Note 7: Signal Processing Methods for EEG Analysis during Electrical Stimulation

To process the EEG signals effectively and mitigate potential artifacts, we employed a rigorous multi-step approach that included filtering, downsampling, and time-frequency analysis. Each step was designed to preserve the integrity of the neural signals while eliminating confounding noise from stimulation and environmental sources. The following sections provide a detailed account of the methods and underlying principles.

1. Data Acquisition and Initial Processing

The EEG signals were acquired at a sampling frequency of $f_s = 4,000$ Hz (or 1000 Hz), a sufficiently high rate to capture the rapid dynamics of neural activities and avoid aliasing. High sampling rates are critical in the initial stages to ensure that all frequency components are accurately recorded.

To mitigate external electrical interference and improve signal quality, the grounding electrode was positioned at the neck, along the electrical pathway between the ulnar stimulation site and the EEG recording electrodes. This strategic placement minimized the effect of stray currents and muscle activity from regions unrelated to the stimulation, ensuring a stable reference for the recorded signals. Such a grounding configuration is crucial for suppressing electrical noise introduced by stimulation artifacts, as well as reducing variability from physiological sources near the head.

2. Noise Reduction Using Empirical Mode Decomposition (EMD)

To address stimulation-induced noise, we employed Empirical Mode Decomposition (EMD) as an initial filtering technique (63). EMD is a data-driven method that decomposes signals into a series of intrinsic mode functions (IMFs), each representing simple oscillatory modes within the data. This approach does not rely on predefined basis functions, making it well-suited for non-stationary and nonlinear signals such as EEG. Due to the limited number of channels (only three in this study), traditional techniques such as Independent Component Analysis (ICA), commonly used for multi-channel artifact removal, were not feasible. In contrast, EMD allowed us to effectively isolate and remove noise components without relying on the statistical assumptions required for ICA.

The decomposition of the EEG signal $x(t)$ into N IMFs is expressed mathematically as:

$$x(t) = \sum_{k=1}^N \text{IMF}_k(t) + r(t), \quad (\text{S38})$$

where $\text{IMF}_k(t)$ represents the k -th intrinsic mode function, characterized by a symmetric waveform with a zero mean and the same number of extrema and zero crossings, and $r(t)$ is the residual signal, representing the slowly varying trend or baseline.

Each IMF is extracted iteratively using the sifting process, which identifies local maxima and minima, interpolates them to form an upper and lower envelope, and subtracts their mean from the signal. The process repeats until the resulting IMF satisfies the criteria of zero mean and symmetric oscillations. For artifact removal, IMFs dominated by stimulation noise and high-amplitude artifacts (e.g., frequencies around the stimulation frequency) were identified and excluded. The residual signal and the remaining IMFs, containing neural activities, were retained for further analysis. This method effectively isolated stimulation noise from the underlying neural signals, preserving the spectral and temporal fidelity of the EEG data.

3. Bandpass and Notch Filtering

To further refine the signal, we applied a bandpass filter with a passband tailored to the frequency range of neural activities of interest. The bandpass filter was implemented using a zero-phase finite impulse response (FIR) design to avoid phase distortion, ensuring that the temporal structure of the EEG signals was preserved. Additionally, notch filters were employed to suppress specific interference frequencies: 60 Hz to remove power-line noise, and 150 Hz (or stimulation frequency) to eliminate noise from electrical stimulation. The notch filters were designed as second-order infinite impulse response (IIR) filters with a quality factor $Q=30$, ensuring effective attenuation of the target frequencies without excessively broad attenuation bands.

4. Downsampling

The sampling frequency of 4000 Hz (or 1000 Hz) was used to enable low input skin contact impedance, achieving a better signal-to-noise ratio. After filtering, the data were downsampled from 4000 Hz (or 1000 Hz) to 500 Hz. Downsampling reduces computational complexity while retaining the frequency components of interest, as neural signals of relevance typically fall below 250 Hz. Prior to downsampling, an anti-aliasing low-pass filter was applied to prevent spectral distortion. This filter was designed with a cutoff frequency of 200 Hz, well below the Nyquist limit of the reduced sampling rate.

5. Time-Frequency Analysis Using Gabor Spectrogram

To investigate the time-frequency dynamics of the EEG signals, we employed the Gabor spectrogram, a method that provides high-resolution spectrotemporal representations by convolving the signal with Gaussian-windowed sinusoidal functions. The spectrogram $S(f, t)$ was computed as:

$$S(f, t) = \int_{-\infty}^{\infty} z(\tau)g(t - \tau)e^{-i2\pi f\tau} d\tau, \quad (\text{S39})$$

where $g(t)$ is the Gaussian window defined as:

$$g(t) = e^{-t^2/(2\sigma^2)}. \quad (\text{S40})$$

The window width σ was optimized to balance time and frequency resolution, considering the trade-off imposed by the uncertainty principle. The resulting spectrogram provided insights into the evolution of neural activities across multiple frequency bands, revealing the temporal modulation of oscillatory patterns.

Supplementary Note 8: Effect on mechanoreceptors during electrical stimulation

Recent advances in neuroprosthetics aim to restore motor control and sensory feedback for amputees through transcutaneous electrical stimulation (TENS) of peripheral nerves. However, the reliance on subjective assessments limits precision and repeatability, thereby hindering robust closed-loop neuroprosthetics. Neurohaptics addresses this gap by objectively quantifying neural responses, notably through biomarkers like EEG-based event-related desynchronization (ERD) in sensorimotor cortical regions such as C1 - C4, and Cz, traditionally associated with mechanoreceptor-driven tactile processing. Determining whether ERD occurs in response to TENS, directly activating sensory nerves, would validate its potential as an objective indicator of cortical engagement in sensory feedback loops.

As these mechanoreceptors convert mechanical vibrations into temporally patterned afferents, the somatosensory cortex receives a frequency-tuned neural response that engages corresponding cortical circuits with precise temporal specificity. Such alignment between peripheral mechanoreceptors' response and cortical desynchronization states underscores a hierarchical coding mechanism, where optimally matched frequencies enhance the fidelity of sensory processing and increase modulation of the cortical network activity. The reduction of alpha power, i.e., ERD, during haptic sensation suggests that active cortical engagement in processing the sensory input may modulate neural responses to minimize interference from artificial sensation. The observed ERD during electrical stimulation is consistent with known mechanisms of sensory processing and sensorimotor integration, where cortical neurons desynchronize their firing rates during active engagement, leading to a reduction in EEG power in specific frequency bands(57). Moreover, the progressive decrease in pain threshold and increased force perception over repeated sessions imply neuroplastic adaptations within the somatosensory pathways(6). These adaptations could involve synaptic potentiation, alterations in receptor sensitivity, or changes in neural network connectivity(5), reflecting the nervous system's ability to adapt to repeated stimuli. Such plasticity is fundamental for learning and memory and has significant implications for rehabilitation and neuroprosthetics applications.

Current Methods of Analysing Response of Mechanoreceptors during Mechanical Stimulation

The response of mechanoreceptors to different frequencies of tactile stimulation is currently measured through the following methods.

Microneurography: Microneurography is a precise technique used to directly measure the activity of individual mechanoreceptor afferents. This method involves inserting a fine needle electrode into a peripheral nerve to record the electrical activity of single nerve fibres. While highly accurate, this technique is invasive and can only record from a limited number of nerve fibres at a time.

Psychophysical Methods: Researchers often use psychophysical experiments to indirectly measure mechanoreceptor responses. These studies involve presenting various tactile stimuli and asking participants to make judgments about the sensations they experience. For example, participants might be asked to discriminate between different frequencies of vibration or to rate the perceived intensity of stimuli.

Intracranial EEG: Intracranial EEG, where electrodes are placed directly on or in the brain, provides high spatial and temporal resolution for measuring neural responses to tactile stimuli (2).

This method has revealed that neural responses to tactile stimulation can be recorded mainly in insular and opercular regions, as well as in the amygdala and fronto-temporal cortex.

Frequency-Dependent Mechanosensation in Mechanoreceptors

The human somatosensory system is finely tuned to detect mechanical stimuli across a wide range of frequencies, enabling precise discrimination of textures, vibrations, and pressure. This capability arises from the specialized roles of mechanoreceptors in the skin, each of which exhibits unique frequency response characteristics. These receptors, classified broadly into rapidly adapting (RA) and slowly adapting (SA) types, demonstrate resonant frequency behaviours that dictate their contributions to sensory perception.

Resonant Frequencies of Mechanoreceptors: Mechanoreceptors such as Meissner's corpuscles, Pacinian corpuscles, Merkel cells, and Ruffini endings are distributed throughout the dermis and epidermis. Each receptor type is optimized for specific frequency ranges. Meissner's corpuscles, located just beneath the epidermis, respond to low-frequency vibrations (2–50 Hz) and are highly sensitive to fluttering or light touch. This frequency range is particularly significant in detecting fine textures and dynamic skin deformations during object manipulation. Pacinian corpuscles, in contrast, are situated deeper in the dermis and are exquisitely sensitive to high-frequency vibrations (50–1,000 Hz). These corpuscles act as high-pass filters, allowing them to detect rapid oscillatory stimuli, such as those generated during the use of tools or interaction with surfaces that produce fine vibrations. Their peak sensitivity at around 250 Hz corresponds to the resonant frequency of the receptor's lamellar structure, which amplifies these signals.

Correlation Between Resonant Frequency and Sensation: The frequency-dependent sensitivity of mechanoreceptors enables the encoding of distinct tactile sensations. The low-frequency sensitivity of Meissner's corpuscles contributes to the perception of softness and slippage, providing critical feedback during grip adjustments. Pacinian corpuscles, by contrast, are essential for detecting surface texture and vibratory cues, such as the fine-grained properties of sandpaper or the vibration of a smartphone. Slowly adapting receptors, including Merkel cells and Ruffini endings, exhibit different resonant behaviours. Merkel cells, which are tuned to static pressure and low-frequency vibrations, are critical for high-acuity spatial discrimination, such as reading Braille or identifying object shapes. Ruffini endings respond to sustained stretch and skin deformation, contributing to proprioception and the perception of skin tension during movements.

Interaction of Mechanoreceptors Across Frequencies: The overlap in frequency sensitivity among these receptors ensures a rich, multimodal sensory experience. For instance, during interactions with textured surfaces, low-frequency signals may activate Meissner's corpuscles, whereas high-frequency components simultaneously stimulate Pacinian corpuscles. This interplay allows the brain to integrate diverse tactile inputs, creating a comprehensive representation of the stimulus.

Supplementary Figure 1

Classes of Electrically Conducting or Semiconducting Hydrogels: *Bicontinuous System*

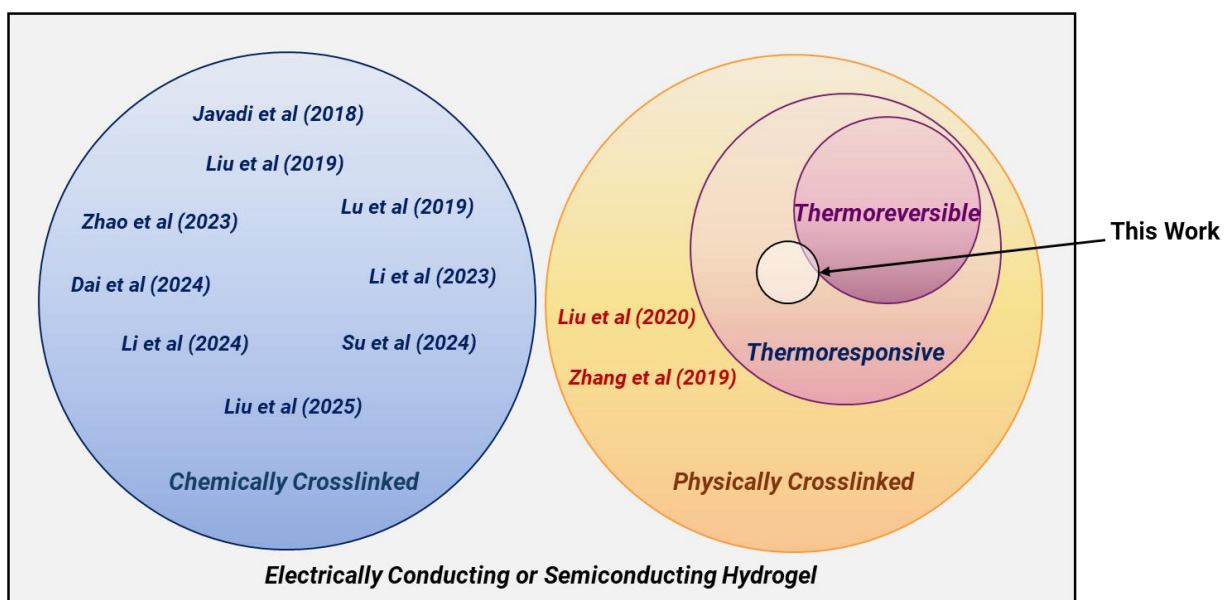


Figure S1: Classes of electrically conducting or semiconducting hydrogels. (8, 9, 11, 12, 14, 22, 46–49, 51)

Supplementary Figure 2

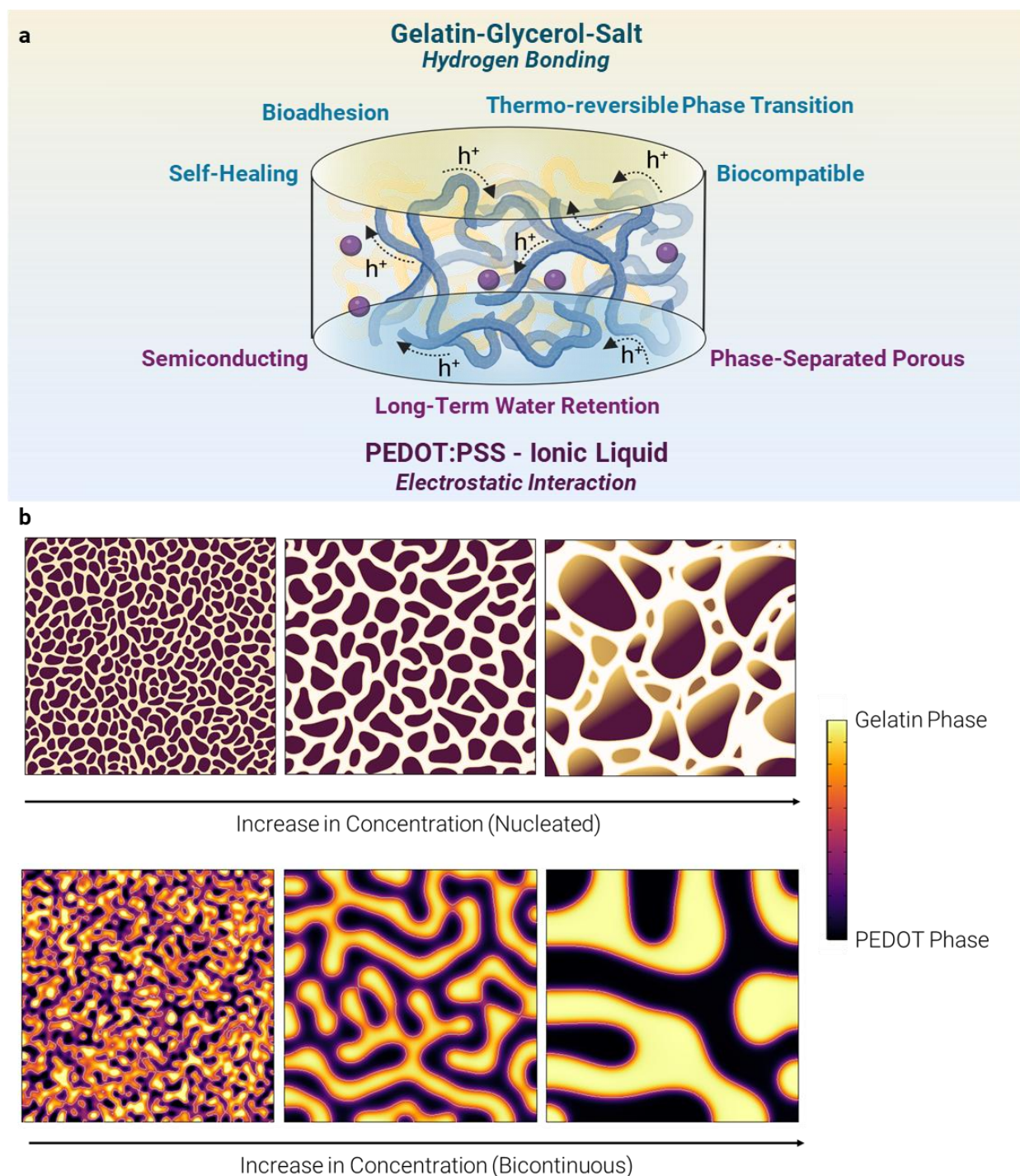


Figure S2: Thermodynamics and Kinetics of Mixing in Ionic Biogel. (a) Schematic of the synergistic effect of the biogel phase (gelatin-glycerol-salt) and ionic phase (PEDOT:PSS-Ionic Liquid). (b) Schematics for the effect of increasing the PEDOT:PSS concentration in the nucleated system (increase in PEDOT:PSS phase size) and the bicontinuous system (onset of percolation of the PEDOT:PSS network).

Supplementary Figure 3

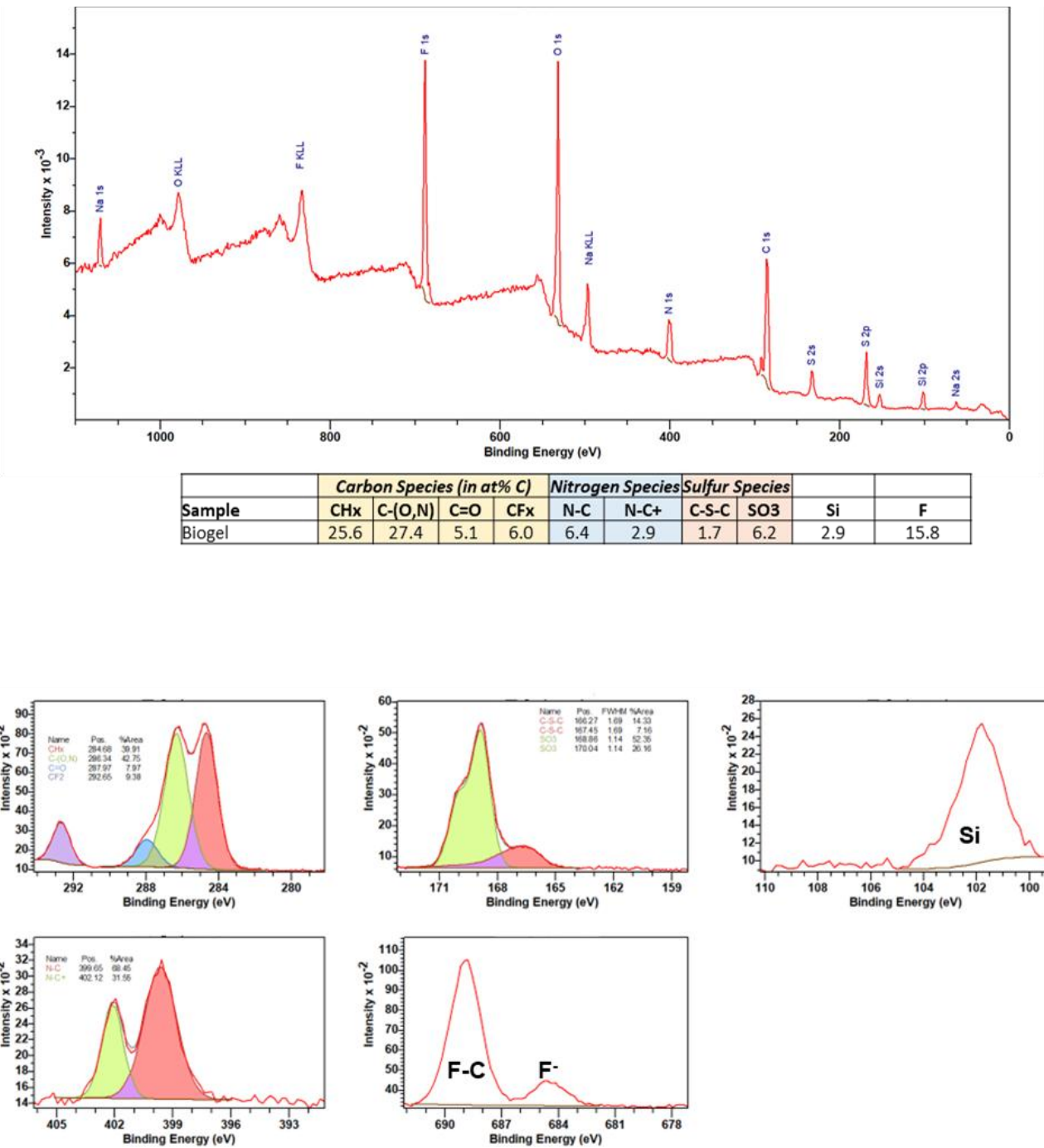


Figure S3: XPS analysis of ionic biogel nucleated 35 w/w%.

Supplementary Figure 4

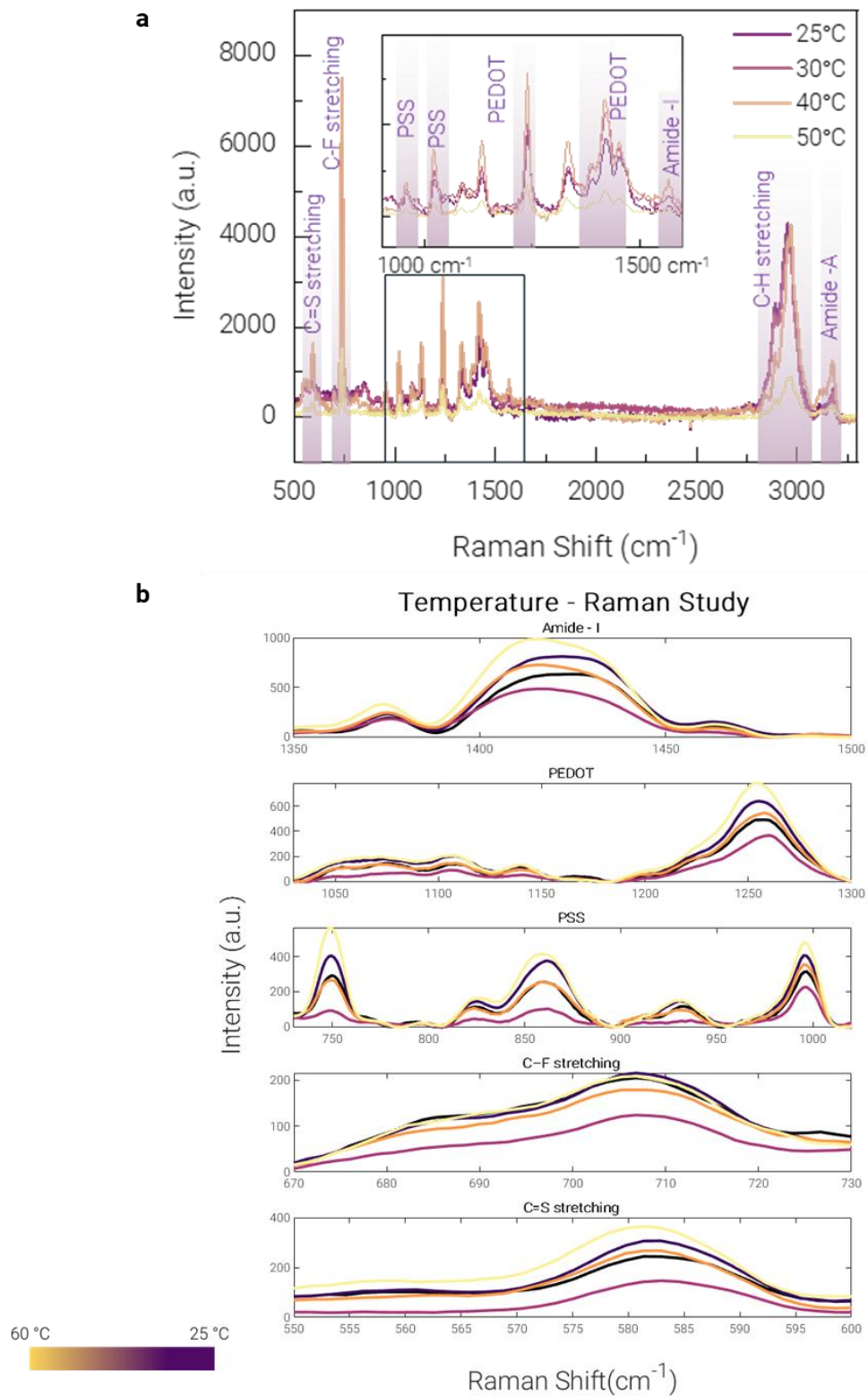


Figure S4: Raman-Temperature Analysis of (a) Nucleated 35 w/w% and (b) Bicontinuous 100 w/w%.

Supplementary Figure 5

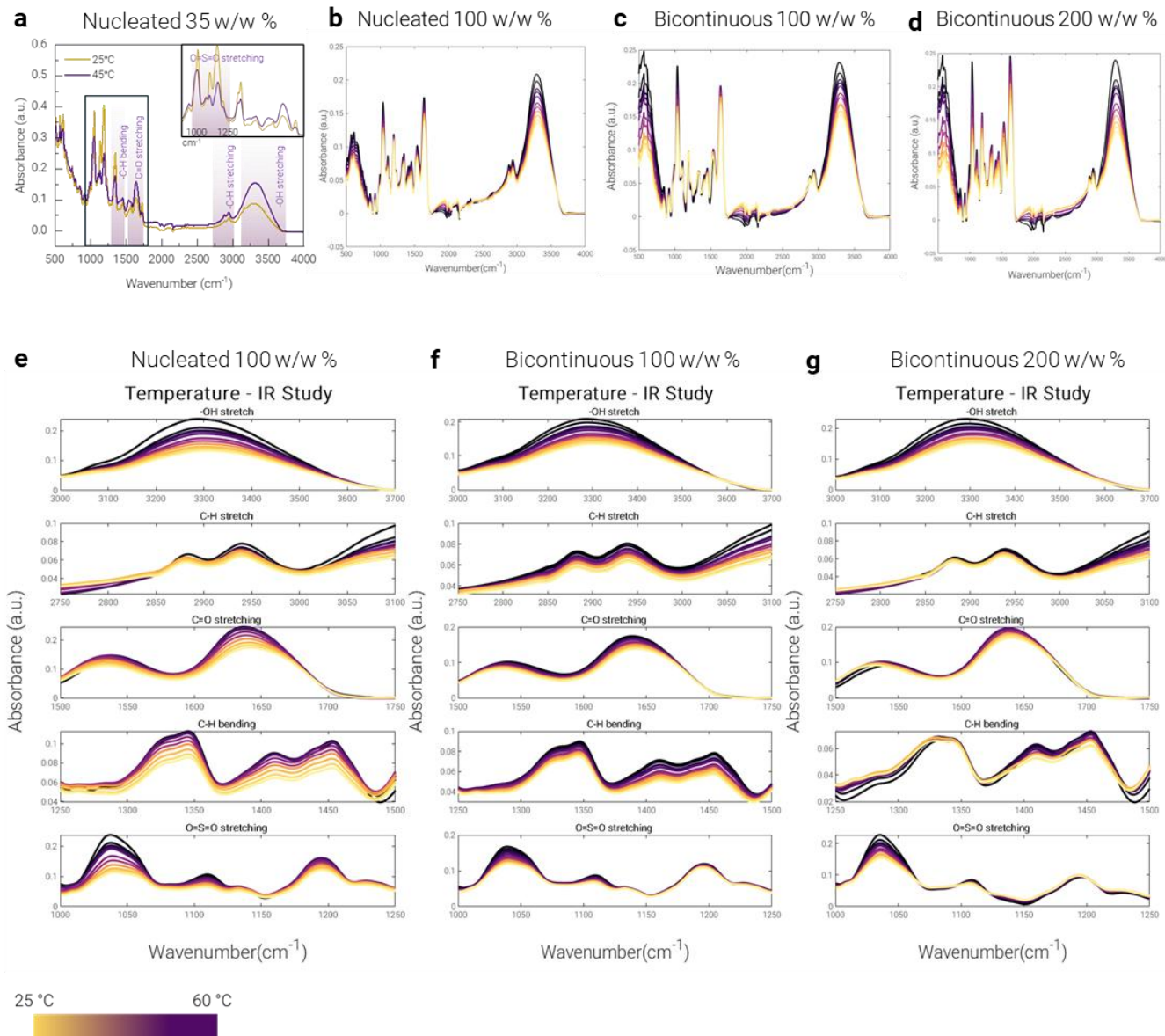
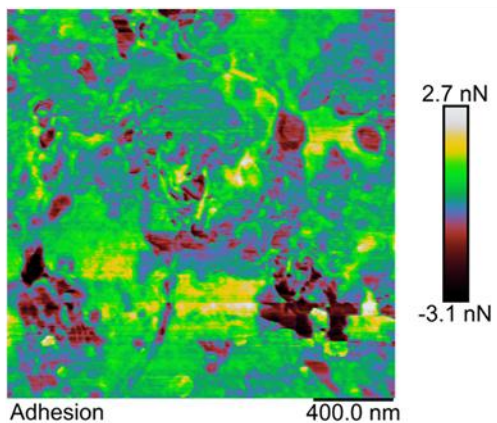
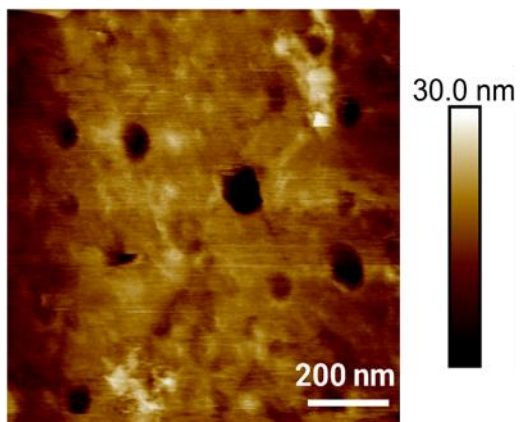


Figure S5: FTIR-Temperature Analysis of (a) Nucleated 35 w/w% and (b) 100 w/w%, and bicontinuous (c) 100 w/w% and (d) 200 w/w%. Functional group analysis as a function of temperature for (e) nucleated 100 w/w%, and bicontinuous (f) 100 w/w% and (g) 200 w/w%.

Supplementary Figure 6

a Nucleated 35 w/w %



b Bicontinuous 100 w/w %

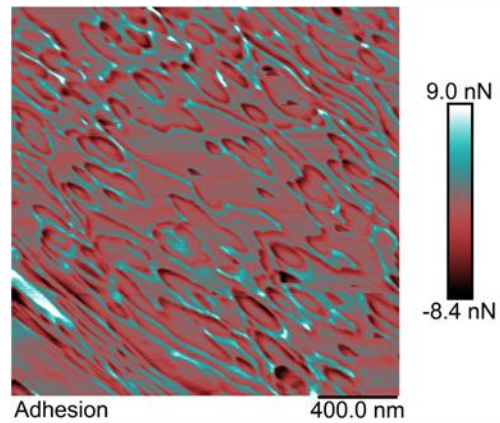
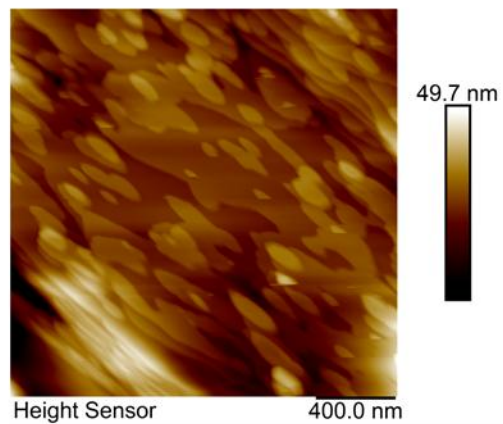


Figure S6: AFM Analysis of (a) Nucleated 35 w/w% and (b) Bicontinuous 100 w/w% to confirm the hierarchical porous nature of the material.

Supplementary Figure 7

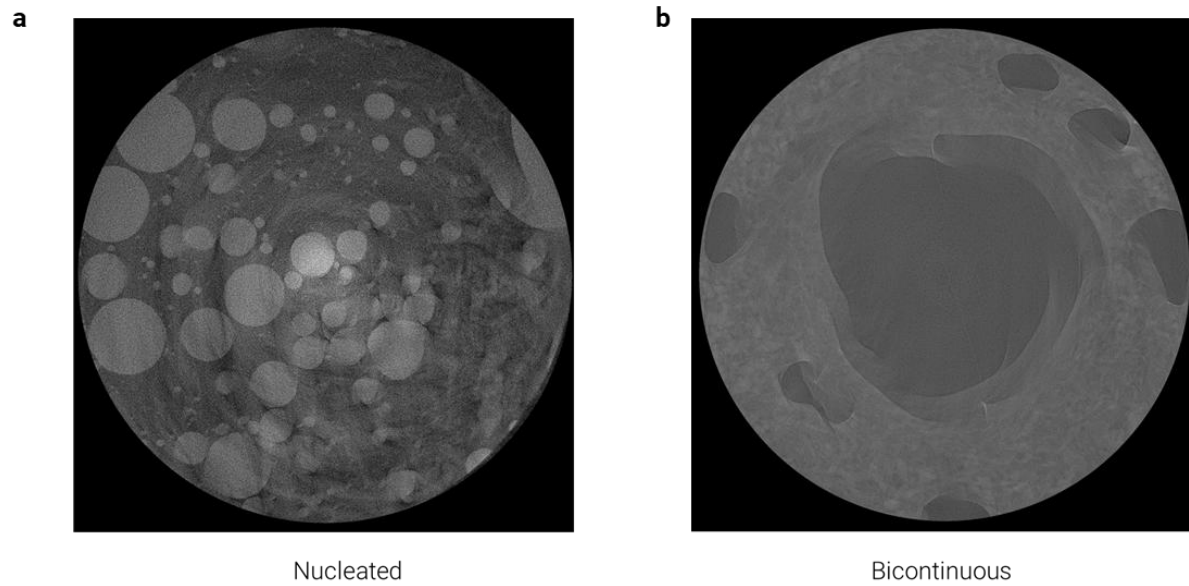


Figure S7: Micro-Computed Tomography of (a) Nucleated 100 w/w% and (b) Bicontinuous 100 w/w%. The field of view is 2 mm x 2mm.

Supplementary Figure 8

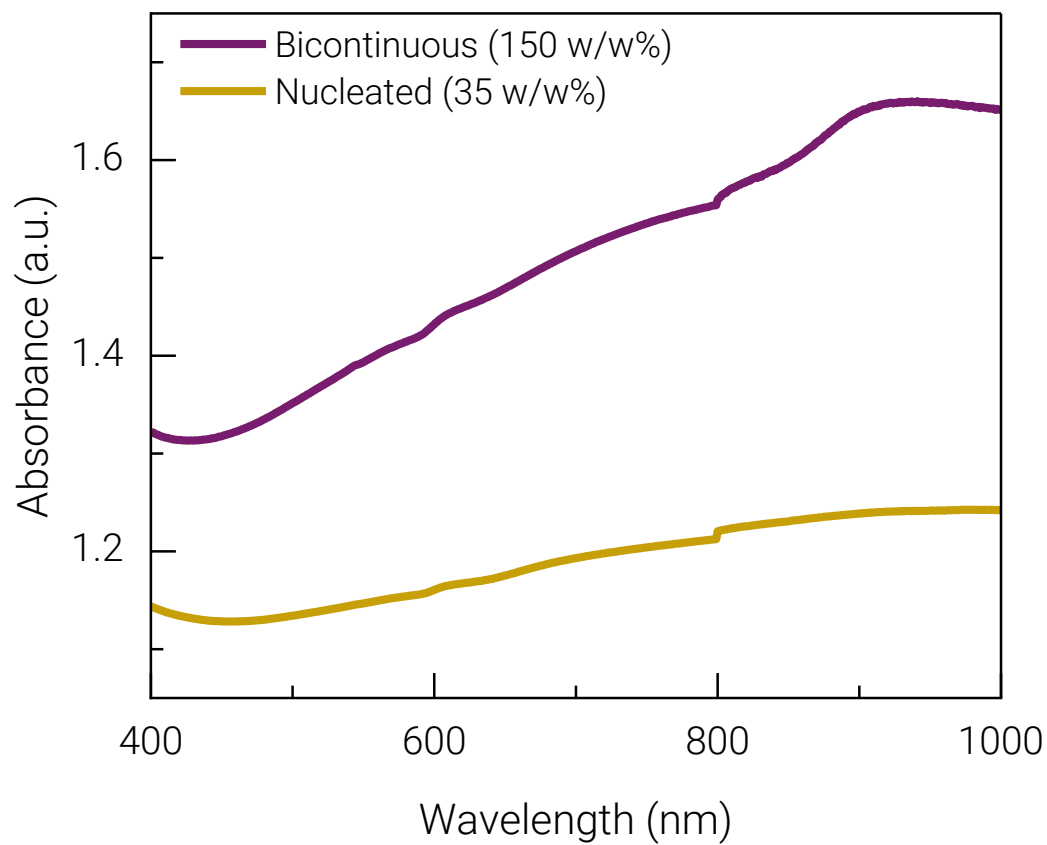


Figure S8: UV-Vis Spectrum of Bicontinuous 150 w/w% and nucleated 35 w/w%.

Supplementary Figure 9

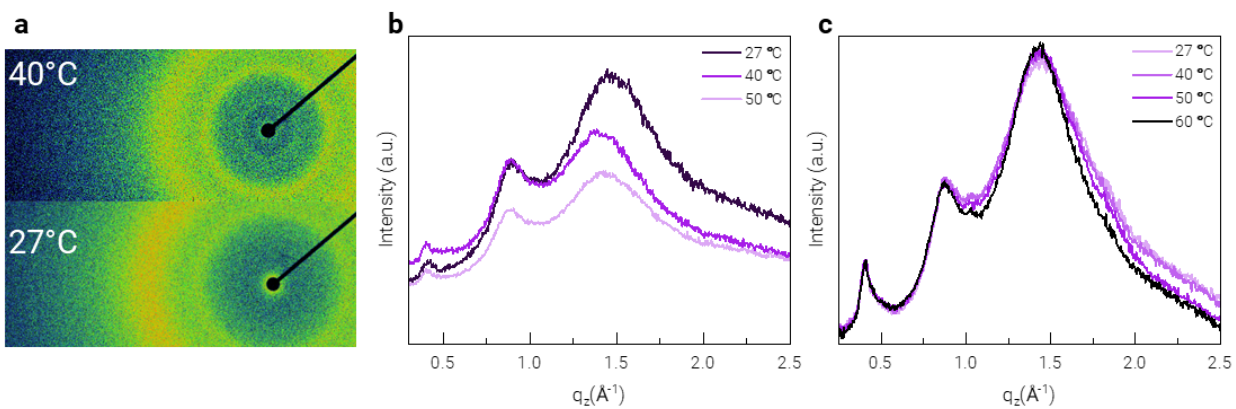


Figure S9: WAXS analysis of (a-b) nucleated 35 w/w% and (c) bicontinuous 100 w/w% across different temperatures.

Supplementary Figure 10

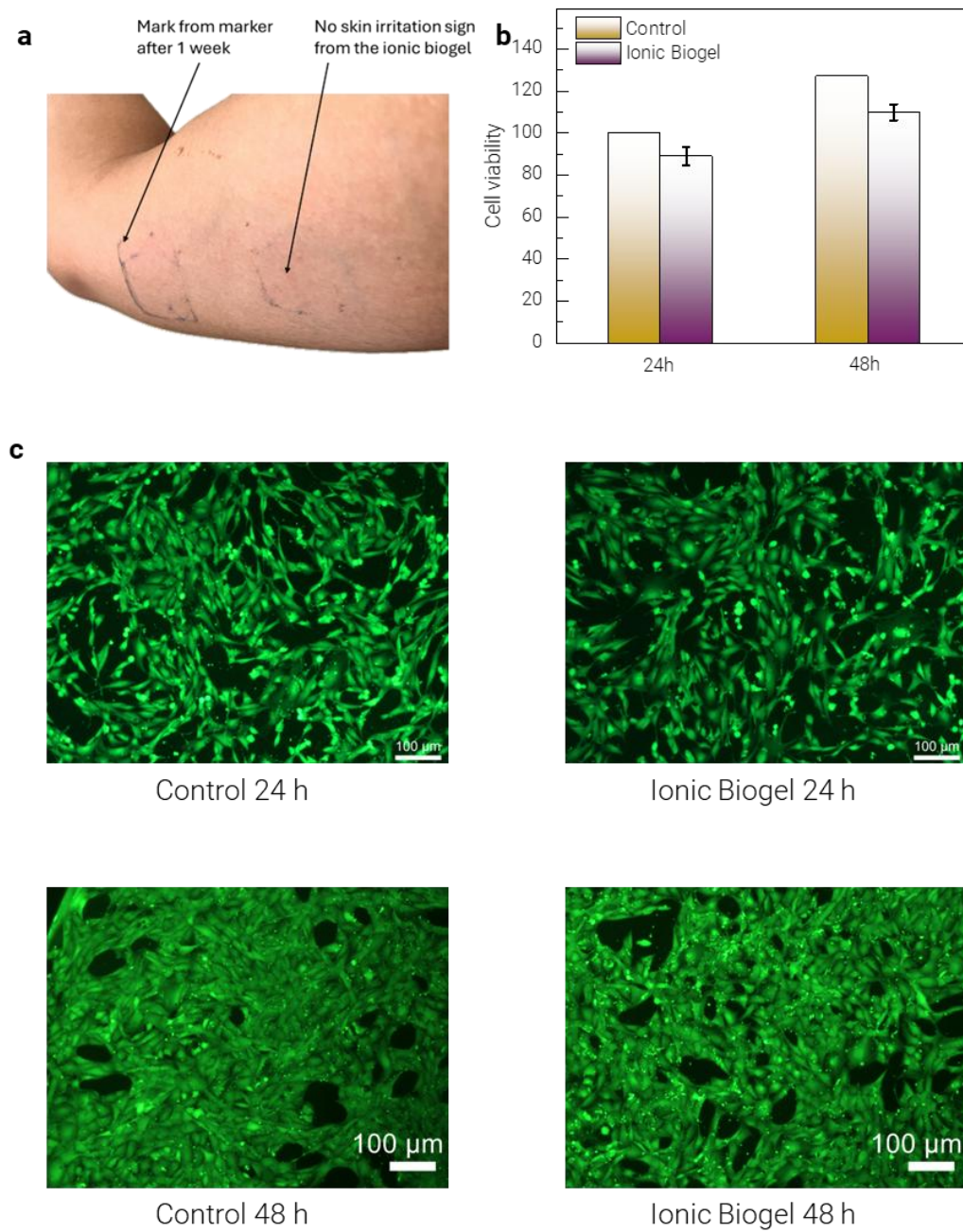


Figure S10: Biocompatibility test of nucleated 35 w/w%. (a) Skin irritation test on a human subject after 7 days of constant contact with the ionic biogel during one of the neurohaptics experiments. (b-c) Cell viability of the ionic biogel against a control for 24 hours and 48 hours.

Supplementary Figure 11

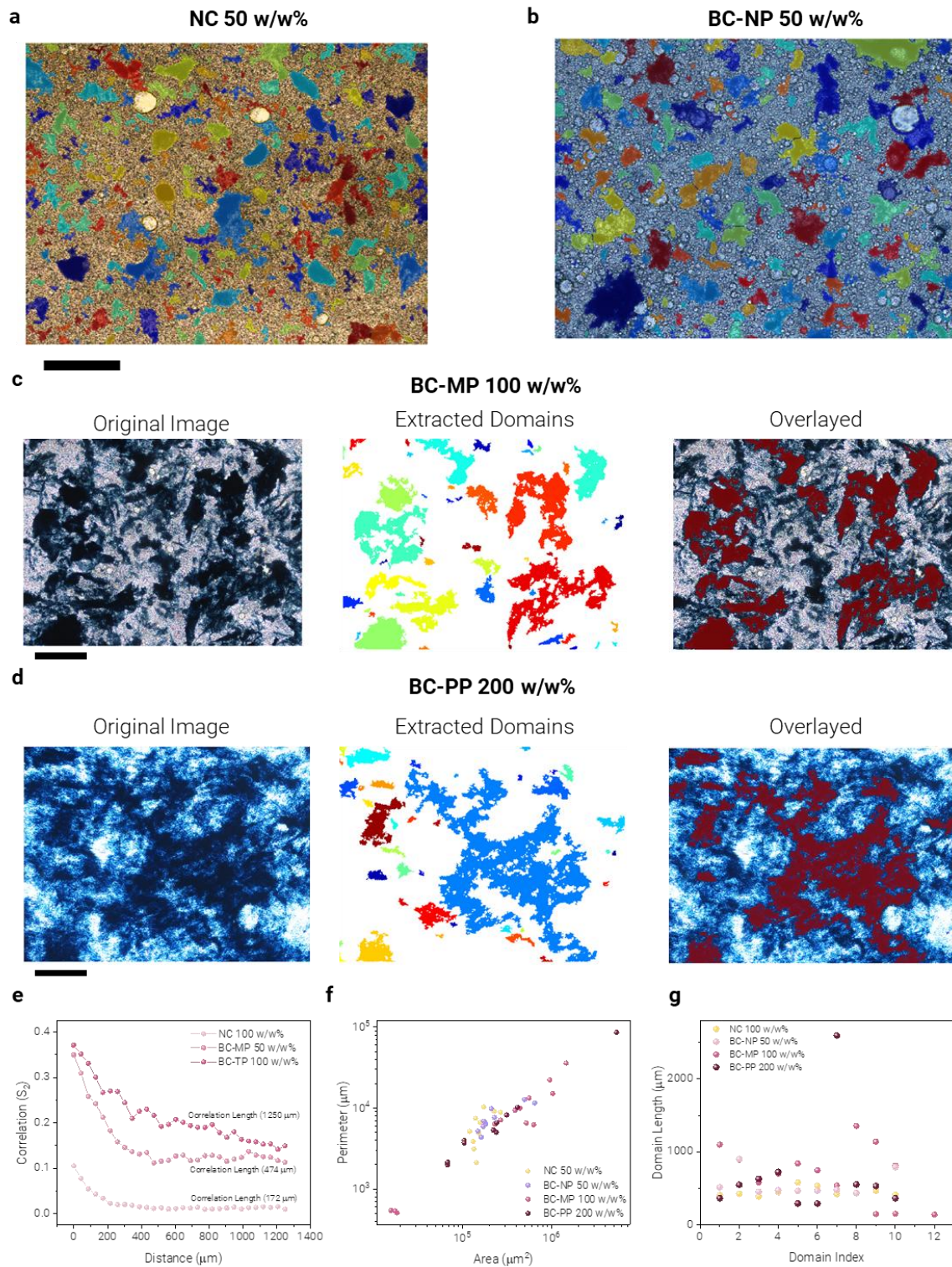


Figure S11: Quantitative image analysis of different phase morphology and overlays of (a) NC 50 w/w%, (b) BC-NP 50 w/w%, (c) BC-MP 100 w/w%, and (d) BC-PP 200 w/w%. (e) 2-point correlation function, (f)

perimeter/area, and **(g)** domain length of NC 100 w/w%, BC-NP 50 w/w%, and BC-MP 100 w/w%. The scale bar for all the images is 0.5 mm.

Supplementary Figure 12

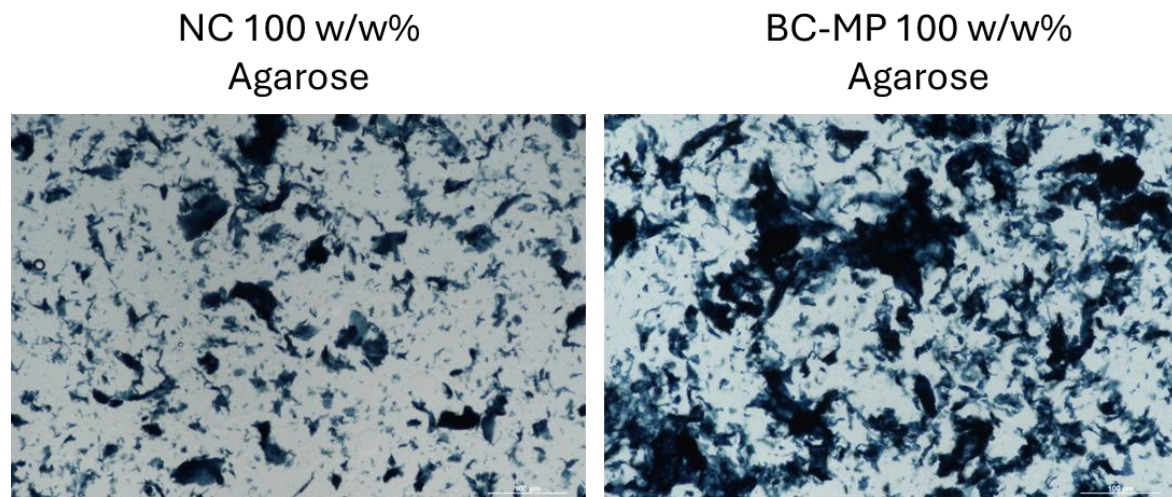


Figure S12: Phase Morphology for Agarose in BC-MP and NC. The scale bar is 100 μm .

Supplementary Figure 13

Bicontinuous System (BC-MP)

Sol Phase ($\tan \delta \sim 1$)
After Heating (80 °C)



Gel Phase ($\tan \delta < 1$)
Room Temperature

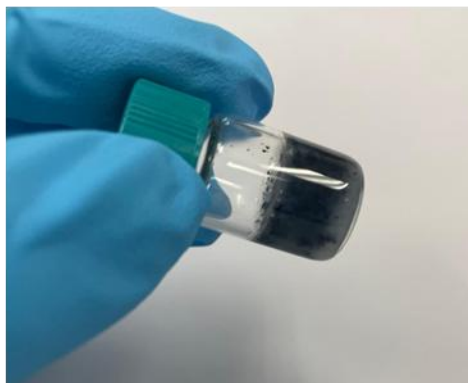


Figure S13: Optical images of bicontinuous system (BC-MP) showing thermoreversibility (inversion test)

Supplementary Figure 14

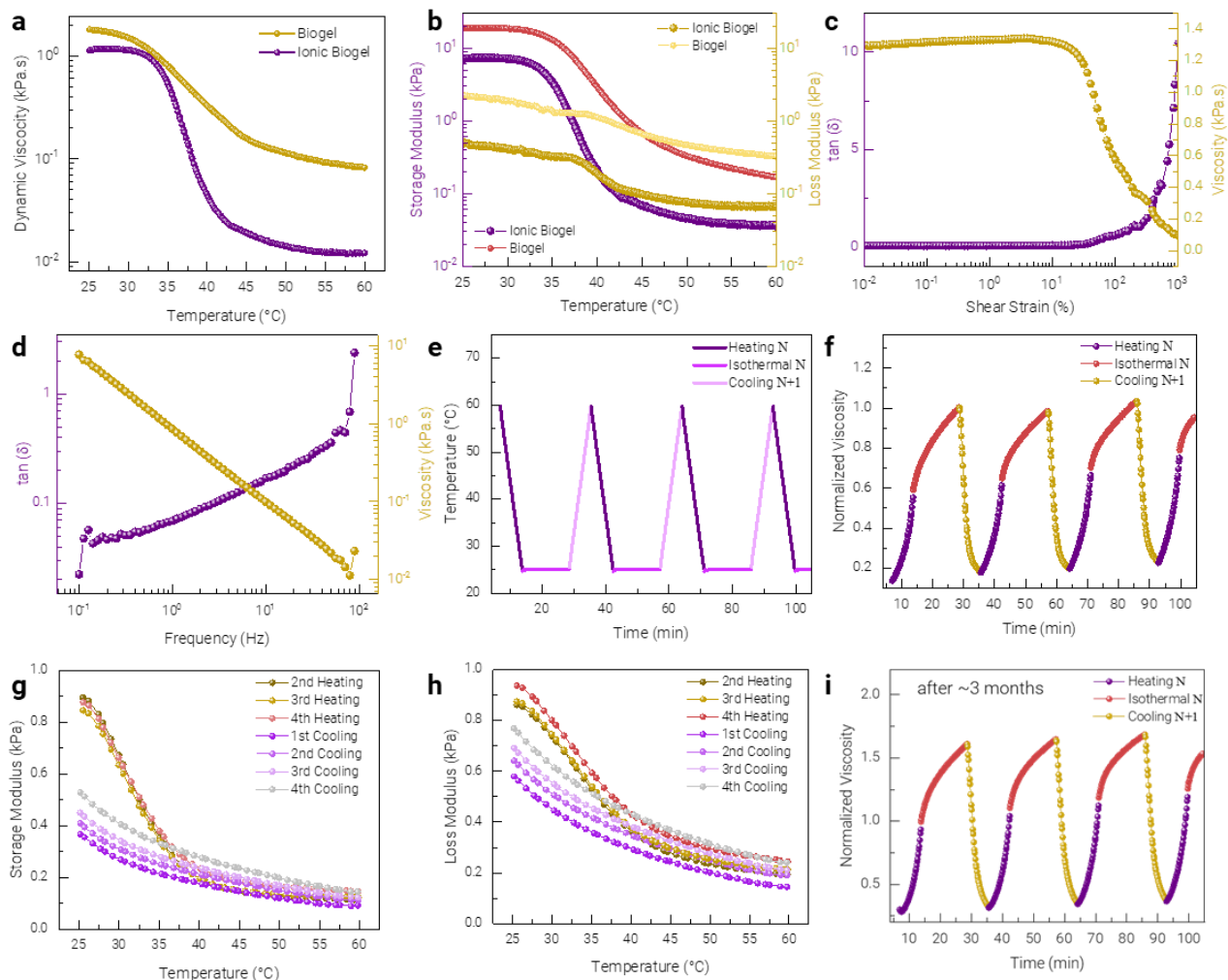


Figure S14: Rheological Analysis of Nucleated 35 w/w%. (a) Dynamic viscosity and (b) Storage modulus of ionic biogel compared to non-ionic biogel. (c) Loss tangent and viscosity as a function of shear strain. (d) Shear-thinning property of the ionic biogel. (e) The temperature profile for the repeated ($n=4$) cycles consisting of heating, cooling at a rate of $5^{\circ}\text{C}/\text{min}$, and followed by an isothermal cycle of 15 mins. (f) Normalized viscosity, (g) storage modulus, and (h) loss modulus of the ionic biogel during these repeated cycles. (i) Normalized viscosity of a ~ 3 -month-old ionic biogel.

Supplementary Figure 15

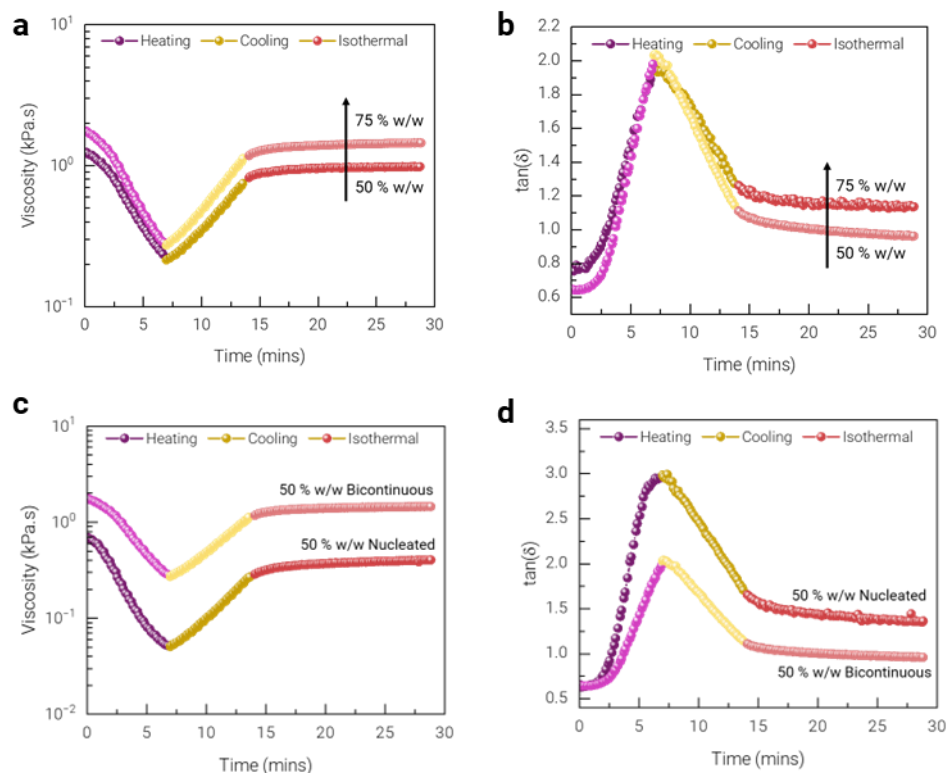


Figure S15: Comparison of Rheology Between Bicontinuous and Nucleated Systems. Comparison of the (a) viscosity and (b) loss tangent of 75 w/w% and 50 w/w% PEDOT:PSS concentration in the bicontinuous system during heating, cooling, and isothermal cycles. Comparison of (c) viscosity and (d) loss tangent of 50 w/w% nucleated and bicontinuous system.

Supplementary Figure 16

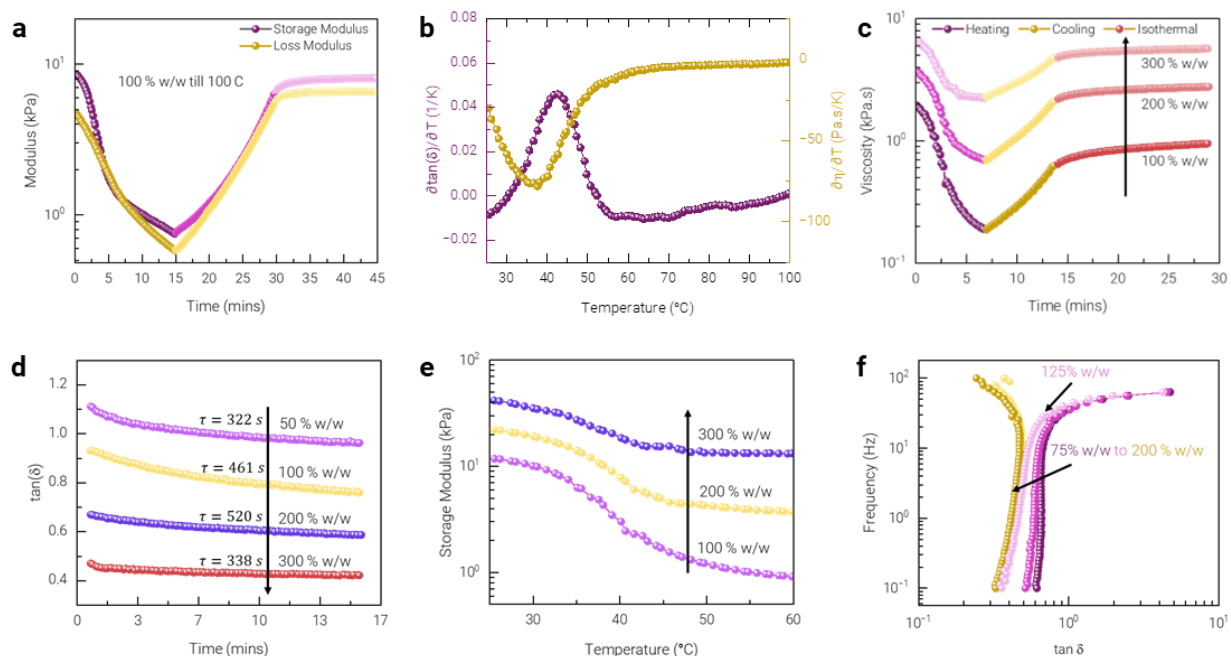


Figure S16: Rheological Analysis of Bicontinuous System. (a) Storage and loss modulus of bicontinuous 100 w/w%, reaching a thermoreversible phase transition briefly. (b) Curvature changes in the loss tangent and viscosity shown by their second-order derivatives. (c) Comparison of the viscosity of 100 w/w%, 200 w/w%, and 300 w/w% bicontinuous systems. (d) Time constant of different concentrations of the bicontinuous system during the isothermal cycle. (e) The change of curvature and comparison of the storage modulus of different concentrations of the bicontinuous systems. (f) Comparison of different concentrations of bicontinuous systems as a function of frequency.

Supplementary Figure 17

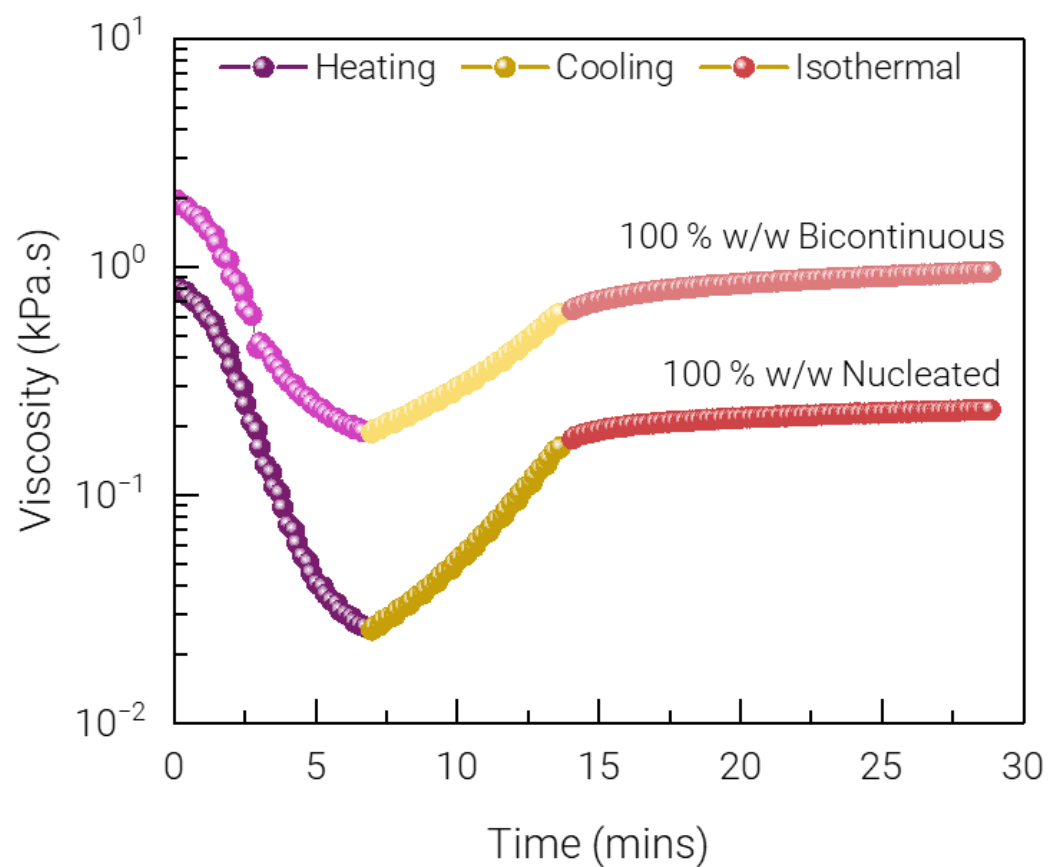


Figure S17: Comparison of Viscosity of 100 w/w% Bicontinuous and Nucleated Systems. The bicontinuous system shows a change in curvature, which is not reflected in the nucleated system.

Supplementary Figure 18

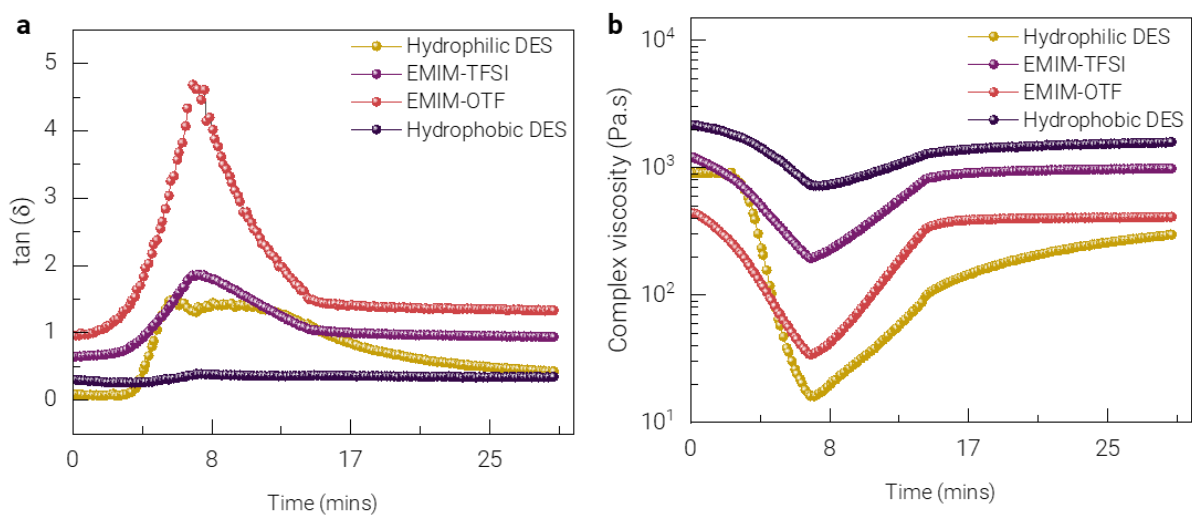


Figure S18: Rheological Analysis of Different Ionic Liquids in the Nucleated System. Comparison of (a) loss tangent and (b) viscosity of different ionic liquids.

Supplementary Figure 19

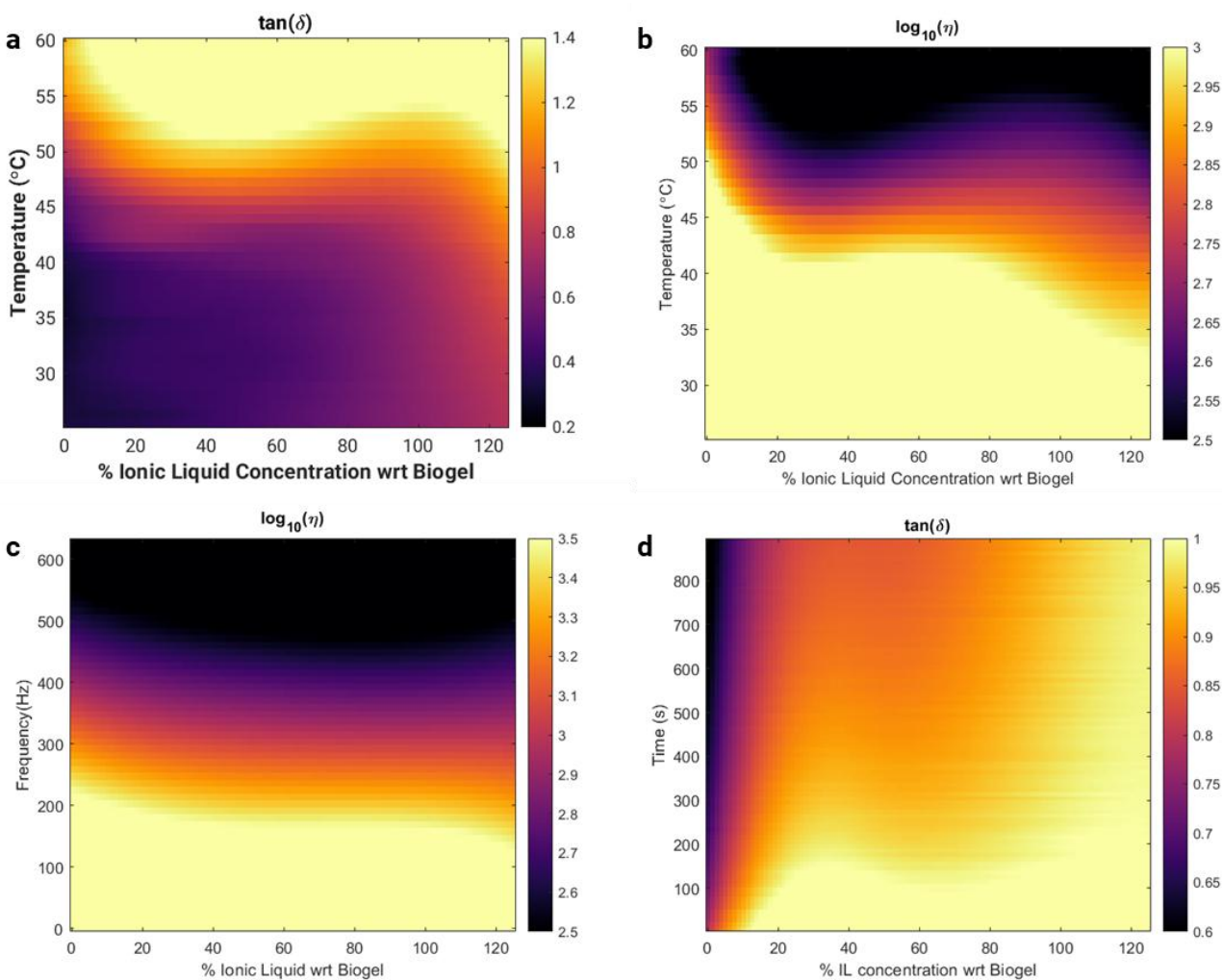


Figure S19: Phase Diagrams w.r.t. the Concentration of Ionic Liquids (with constant PEDOT:PSS concentration) in the Nucleated System. Variation of concentration of EMIM-TFSI in the nucleated system to observe a correlation between temperature and **(a)** loss tangent and **(b)** viscosity. **(c)** Viscosity variation with the concentration of the ionic liquid as a function of frequency or shear rate. **(d)** Change of loss tangent as a function of time in the isothermal relaxation period across different concentrations of the ionic liquid.

Supplementary Figure 20

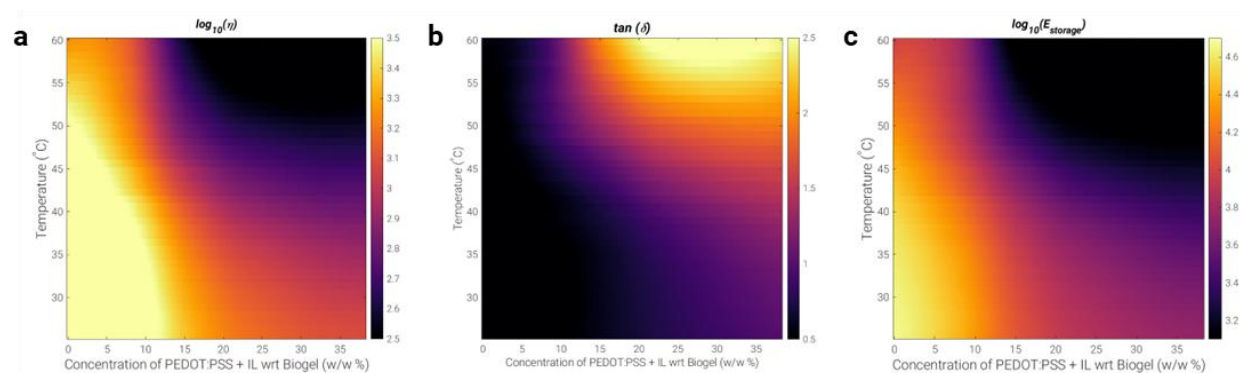


Figure S20: Phase Diagram-Temperature Diagram of Combined Concentration of Ionic Liquids and PEDOT:PSS in the Nucleated System. Variation of combined concentration of EMIM-TFSI and PEDOT:PSS in the nucleated system to observe a correlation between the temperature and **(a)** viscosity and **(b)** loss tangent. **(c)** Variation of storage modulus (in Pa) as a function of temperature across different concentrations.

Supplementary Figure 21

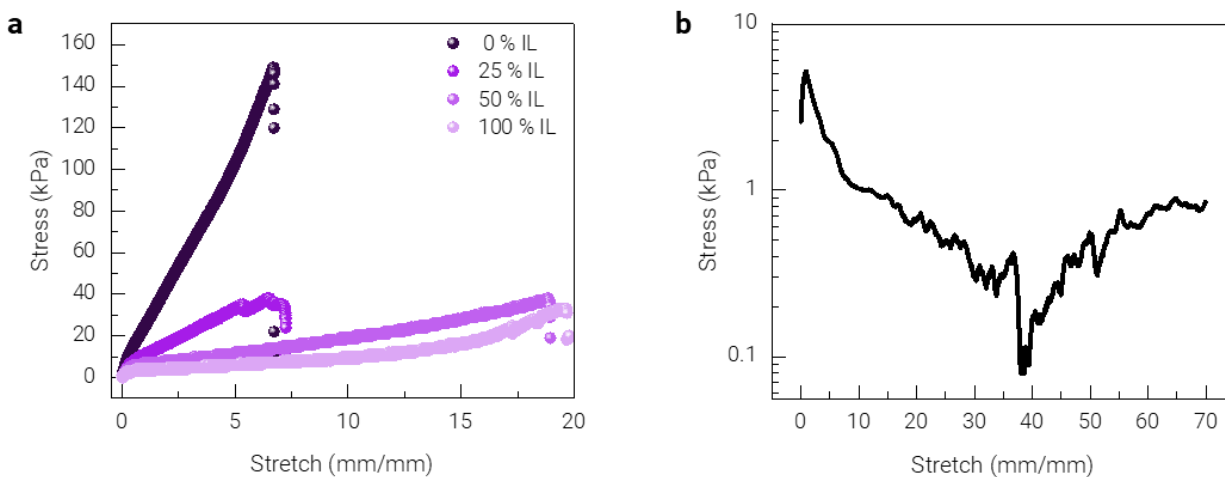


Figure S21: Tensile testing of the Nucleated System with Different Concentrations of Ionic Liquids. Variation of stretchability and Young's modulus as a function of the concentration of EMIM-TFSI: **(a)** 0% IL to 100% IL and **(b)** 125% IL (fluid-like yield stress). The measurements were taken 3 hours after preparation, resulting in ultra-stretchability and liquid-like characteristics with the increase in concentrations.

Supplementary Figure 22

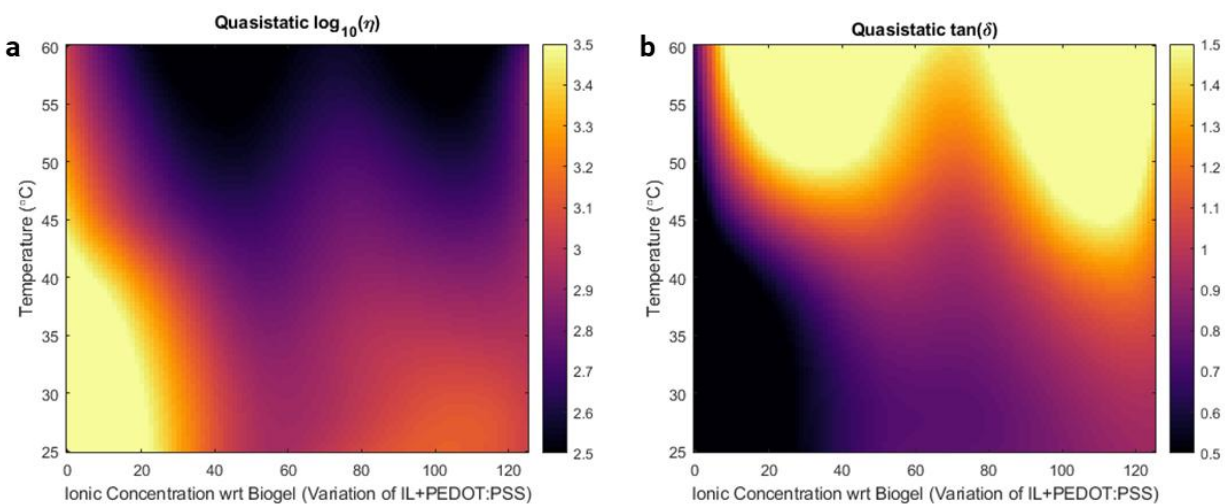


Figure S22: Quasistatic Phase Diagram-Temperature Diagram of Combined concentration of Ionic Liquids and PEDOT:PSS in the Nucleated System. Variation of combined concentration of EMIM-TFSI and PEDOT:PSS in the nucleated system without any temperature rate to observe a correlation between the temperature and (a) viscosity and (b) loss tangent.

Supplementary Figure 23

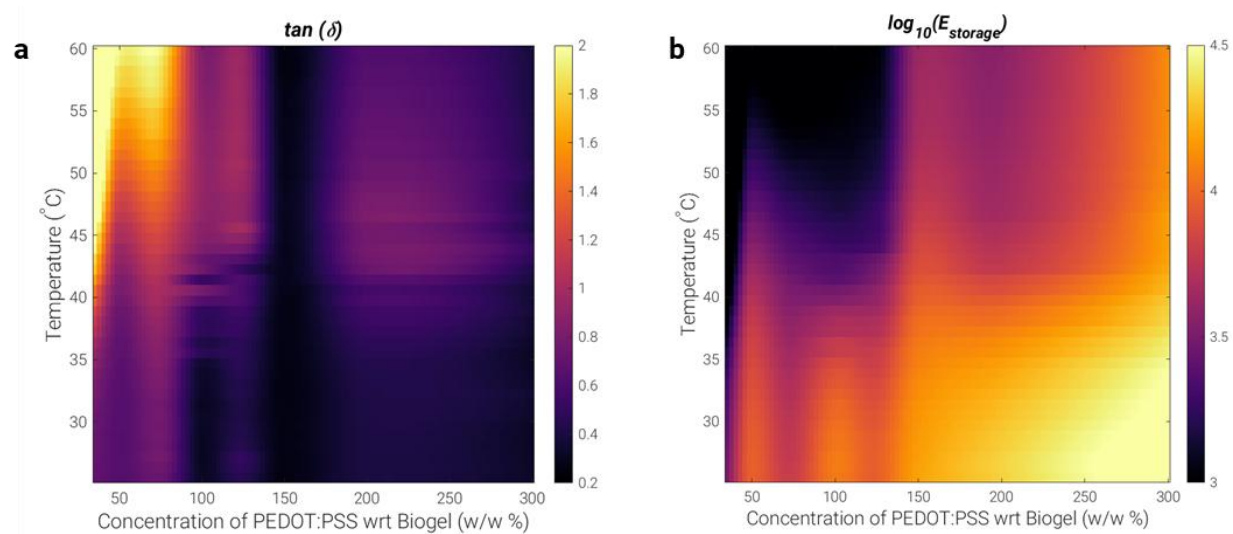


Figure S23: Phase Diagram-Temperature Diagram of PEDOT:PSS Concentration in the Bicontinuous System during Heating. Variation of PEDOT:PSS concentration in the bicontinuous system to observe a correlation between the temperature and (a) loss tangent or (b) storage modulus (in Pa).

Supplementary Figure 24

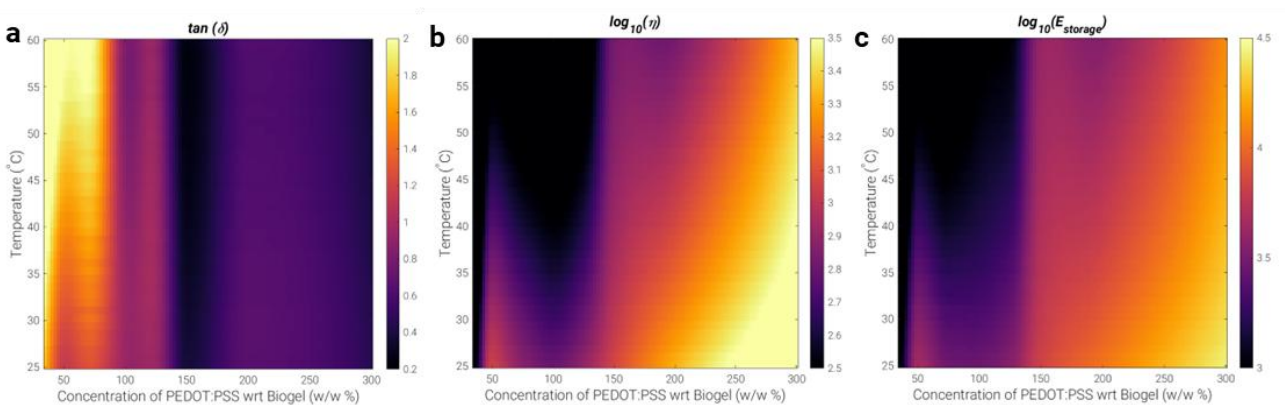


Figure S24: Phase Diagram-Temperature Diagram of PEDOT:PSS Concentration in the Bicontinuous System during Cooling. Variation of PEDOT:PSS concentration in the bicontinuous system to observe a correlation between the temperature and **(a)** loss tangent, **(b)** viscosity and **(c)** storage modulus (in Pa), and the difference from nucleated system shows the loss of thermoreversibility for concentrations 100 – 125 w/w%.

Supplementary Figure 25

Nucleated 35 w/w%

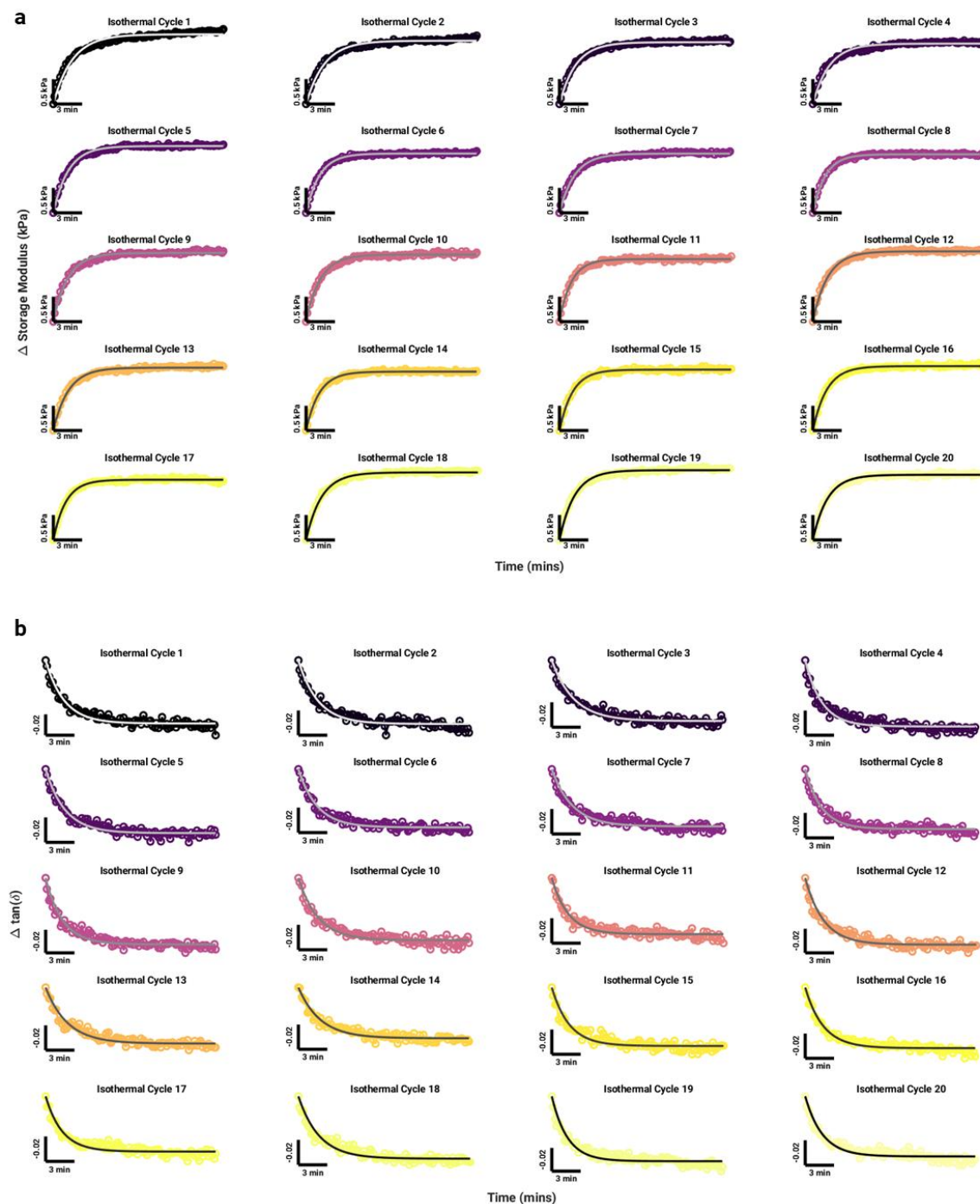


Figure S25: Analysis and Fitting of Each Isothermal Cycle for the Nucleated 35 w/w% System. Variation of (a) storage modulus and (b) loss tangent across different isothermal cycles lasting for 15 mins, after the cooling cycle.

Supplementary Figure 26

Non-ionic Biogel

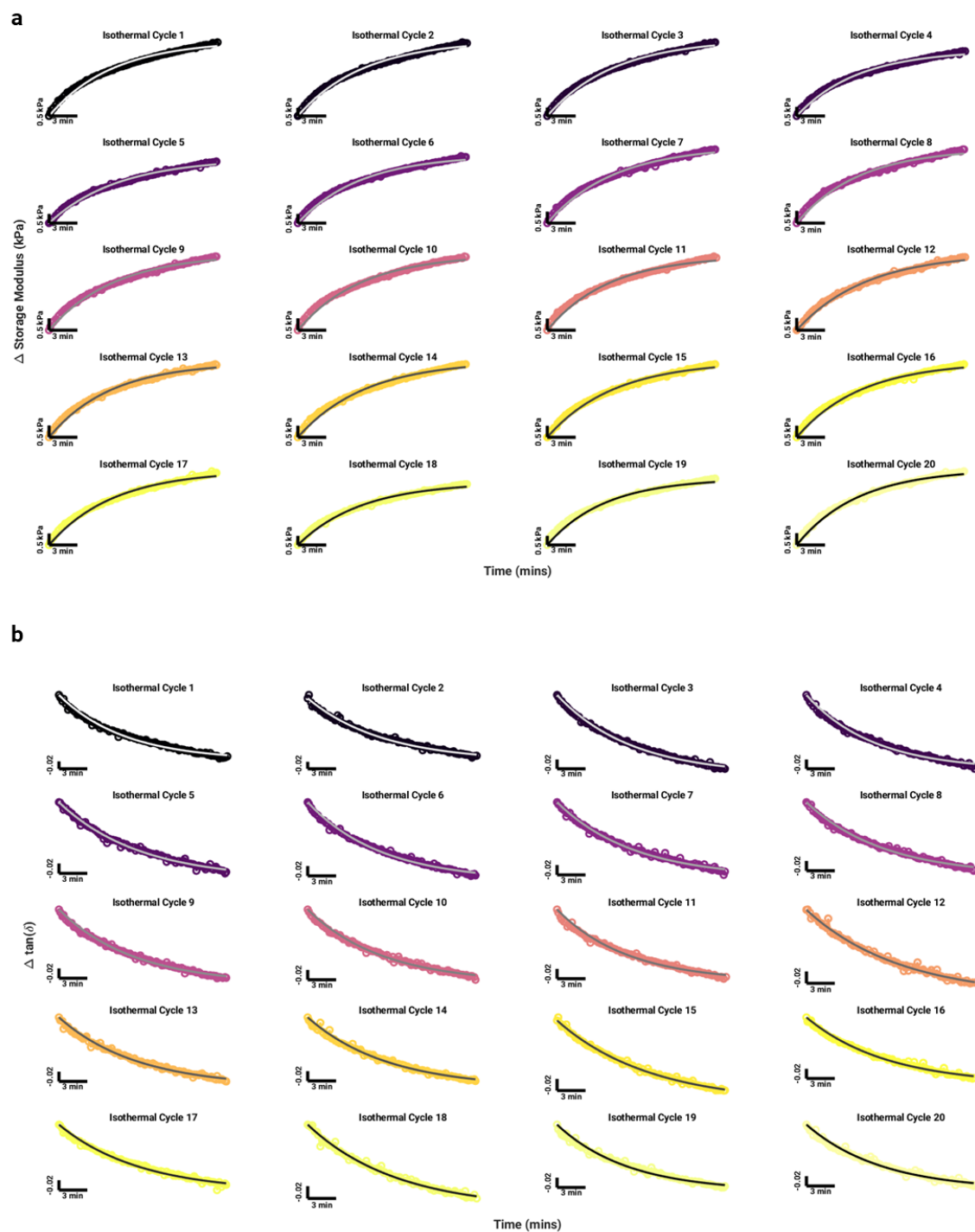


Figure S26: Analysis and Fitting of Each Isothermal Cycle for the Non-ionic Biogel. Variation of (a) storage modulus and (b) loss tangent across different isothermal cycles lasting for 15 mins, after the cooling cycle.

Supplementary Figure 27

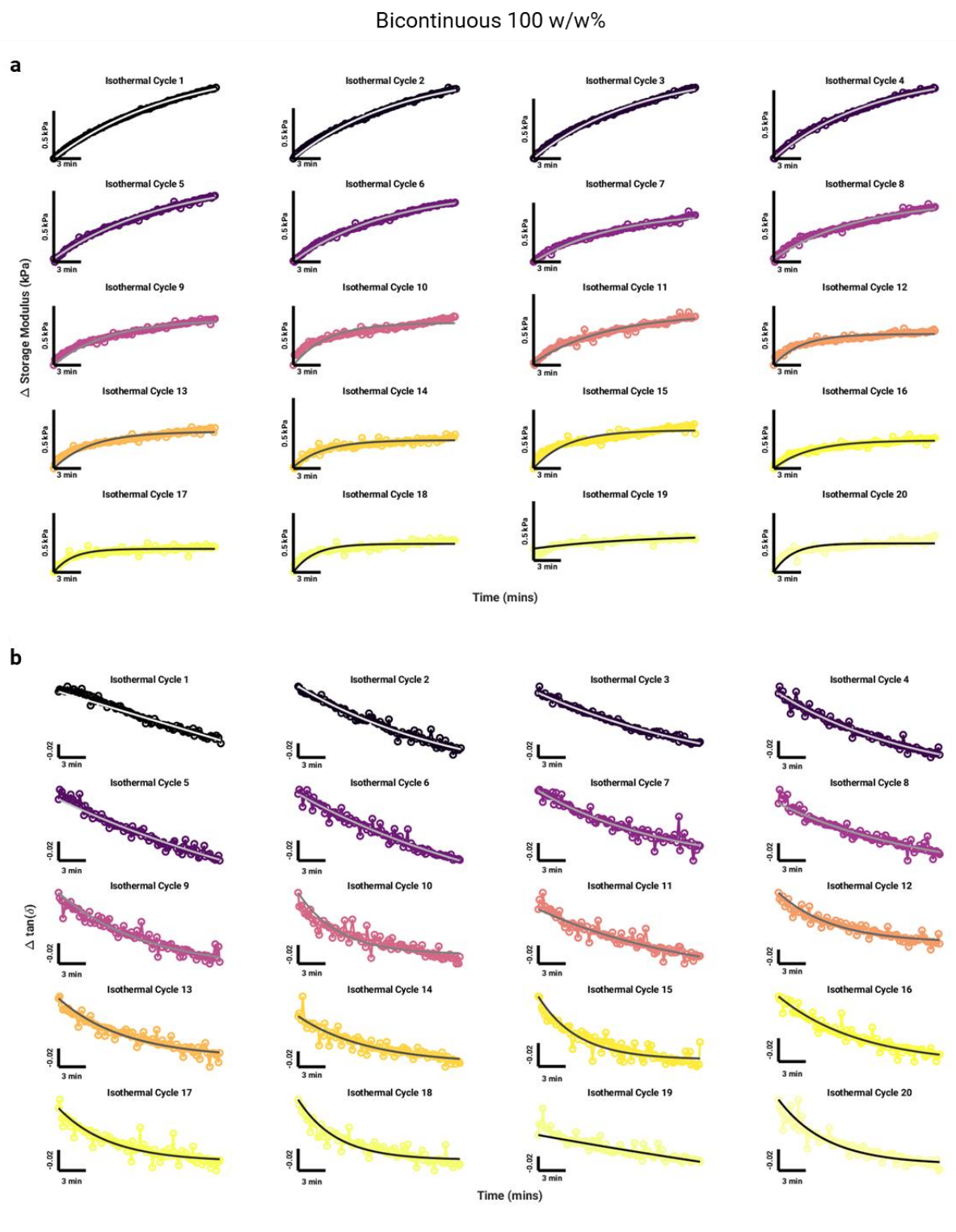


Figure S27: Analysis and Fitting of Each Isothermal Cycle for the Bicontinuous 100 w/w% System. Variation of **(a)** storage modulus and **(b)** loss tangent across different isothermal cycles lasting for 15 mins, after the cooling cycle.

Supplementary Figure 28

Bicontinuous 50 w/w%

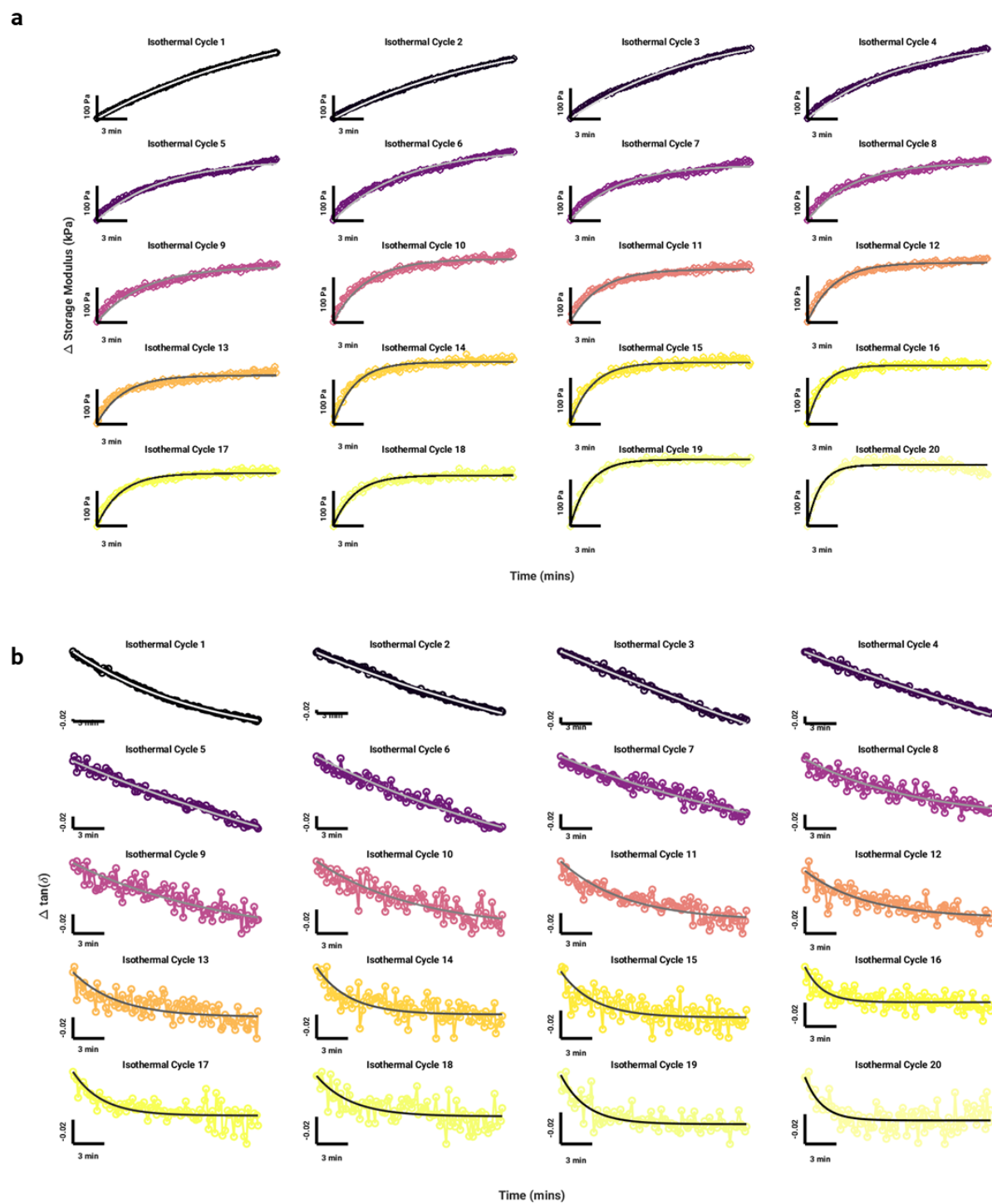


Figure S28: Analysis and Fitting of Each Isothermal Cycle for the Bicontinuous 50 w/w% System. Variation of **(a)** storage modulus and **(b)** loss tangent across different isothermal cycles lasting for 15 mins, after the cooling cycle.

Supplementary Figure 29

Bicontinuous 35 w/w%

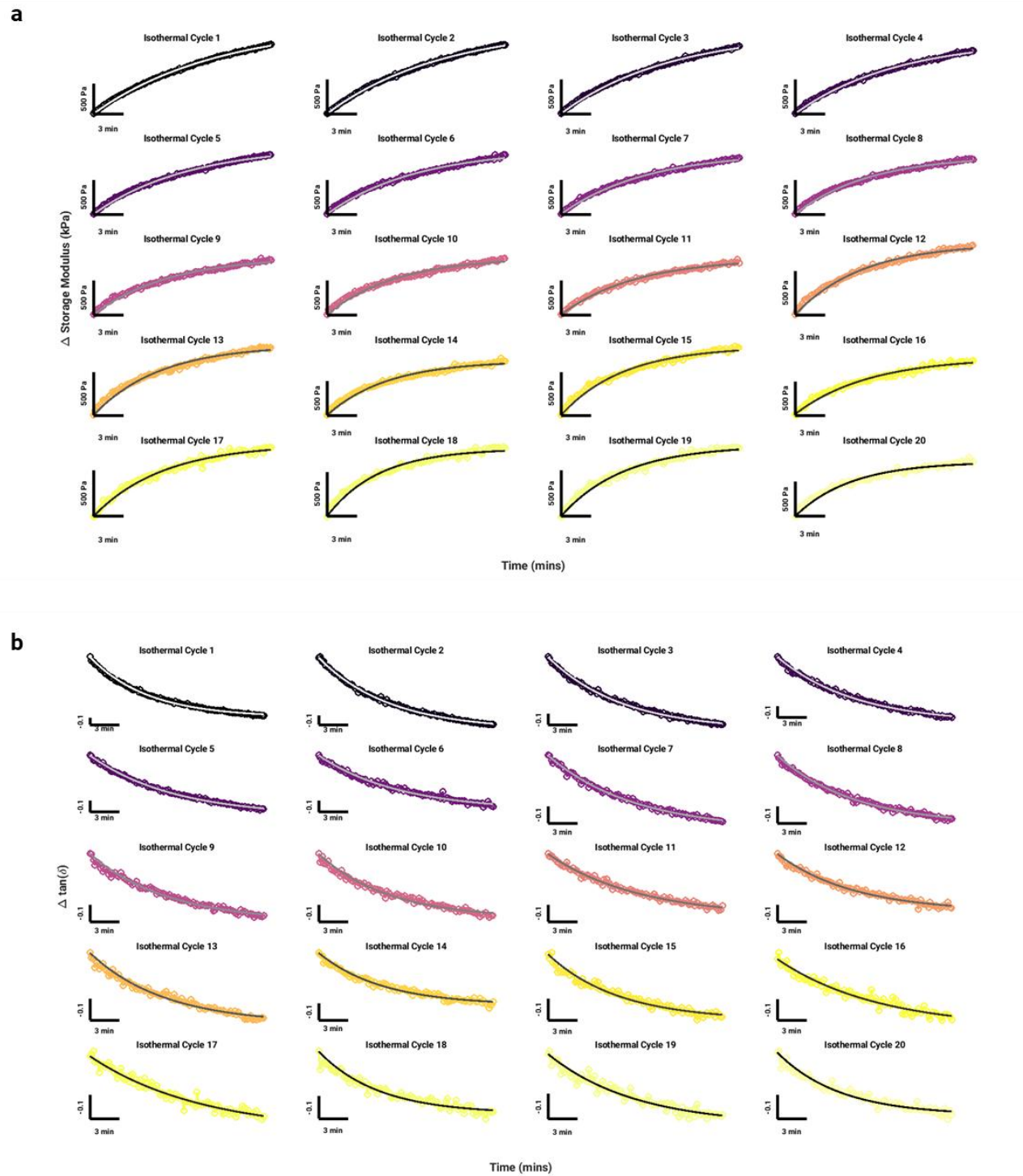


Figure S29: Analysis and Fitting of Each Isothermal Cycle for the Bicontinuous 35 w/w% System. Variation of (a) storage modulus and (b) loss tangent across different isothermal cycles lasting for 15 mins, after the cooling cycle.

Supplementary Figure 30

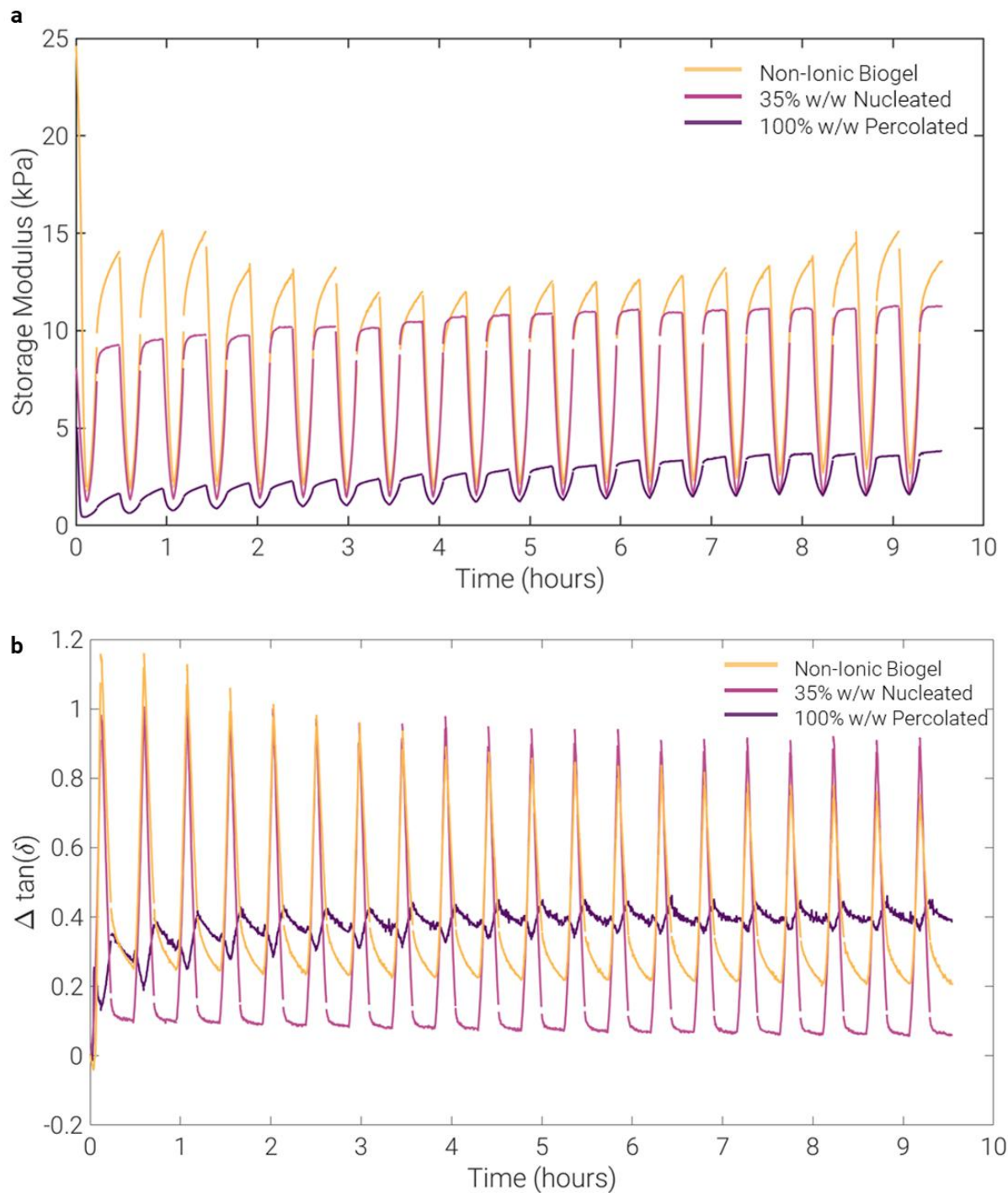


Figure S30: Variation of Rheological Properties across Cycles for Different Material Systems. Variation of (a) storage modulus and (b) changes in loss tangent across different thermal cycles (heating – 5°C/min, cooling – 5°C/min, and isothermal – 15 mins) for non-ionic biogel, 35 w/w% nucleated, and 100 w/w% bicontinuous.

Supplementary Figure 31

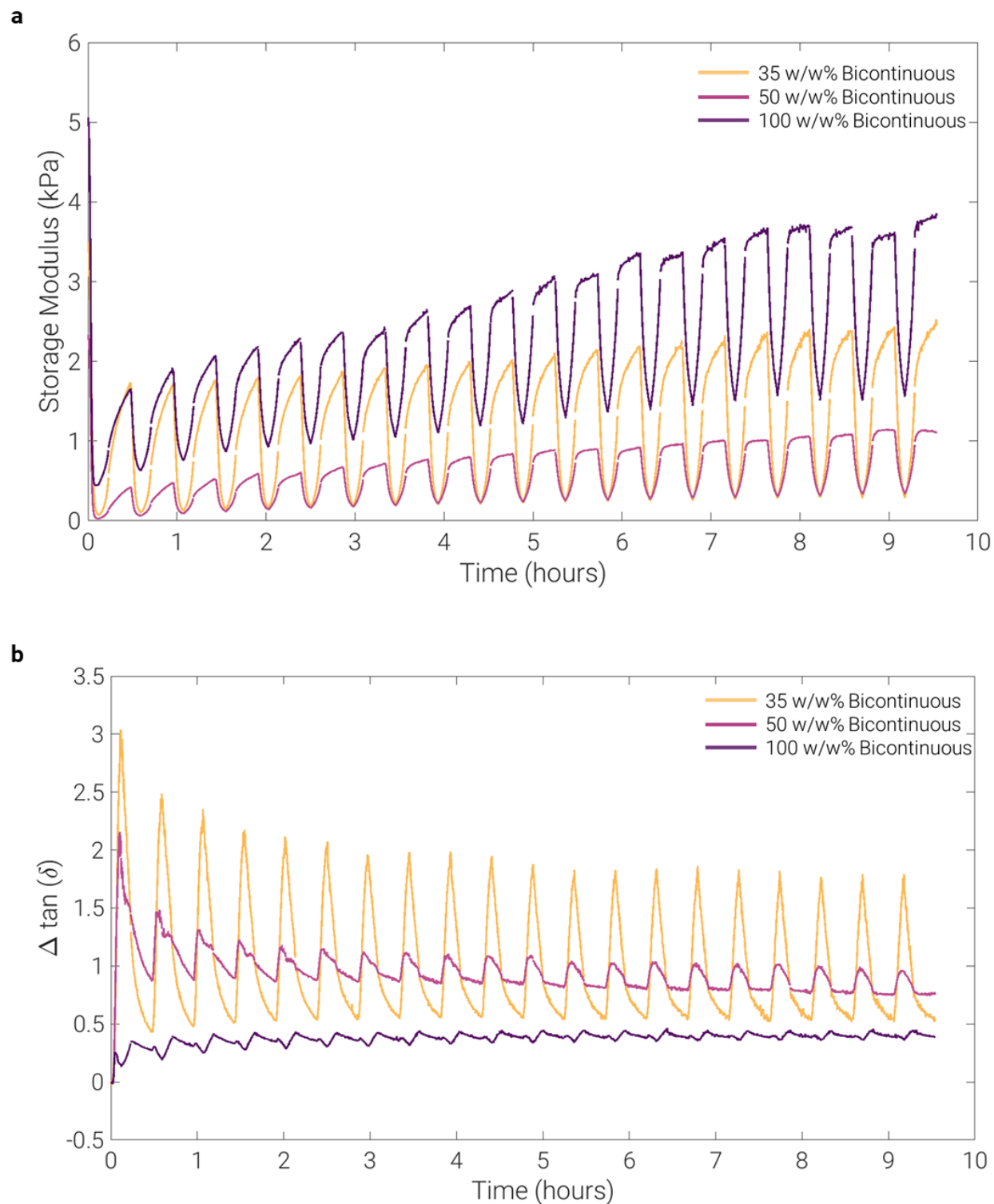


Figure S31: Variation of Rheological Properties across Cycles for Different Bicontinuous Systems. Variation of **(a)** storage modulus and **(b)** changes in loss tangent across different thermal cycles (heating – 5°C/min, cooling – 5°C/min, and isothermal – 15 mins) for 35 w/w%, 50 w/w%, and 100 w/w% bicontinuous.

Supplementary Figure 32

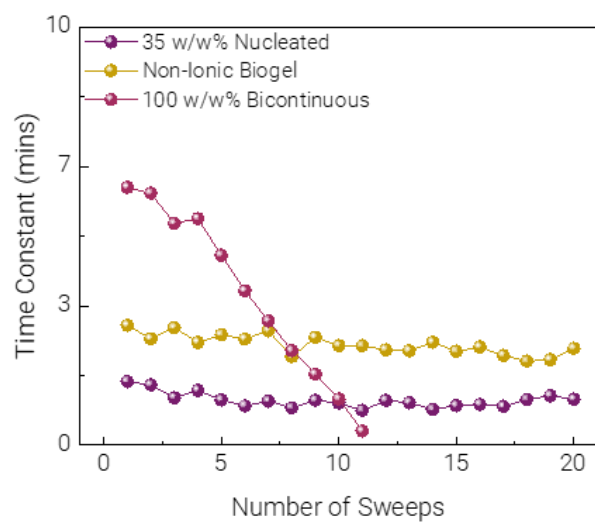


Figure S32: Comparison of the time constant of isothermal relaxation of loss modulus over 20 thermal cycles for nucleated 35 w/w%, non-ionic biogel and bicontinuous 100 w/w%.

Supplementary Figure 33

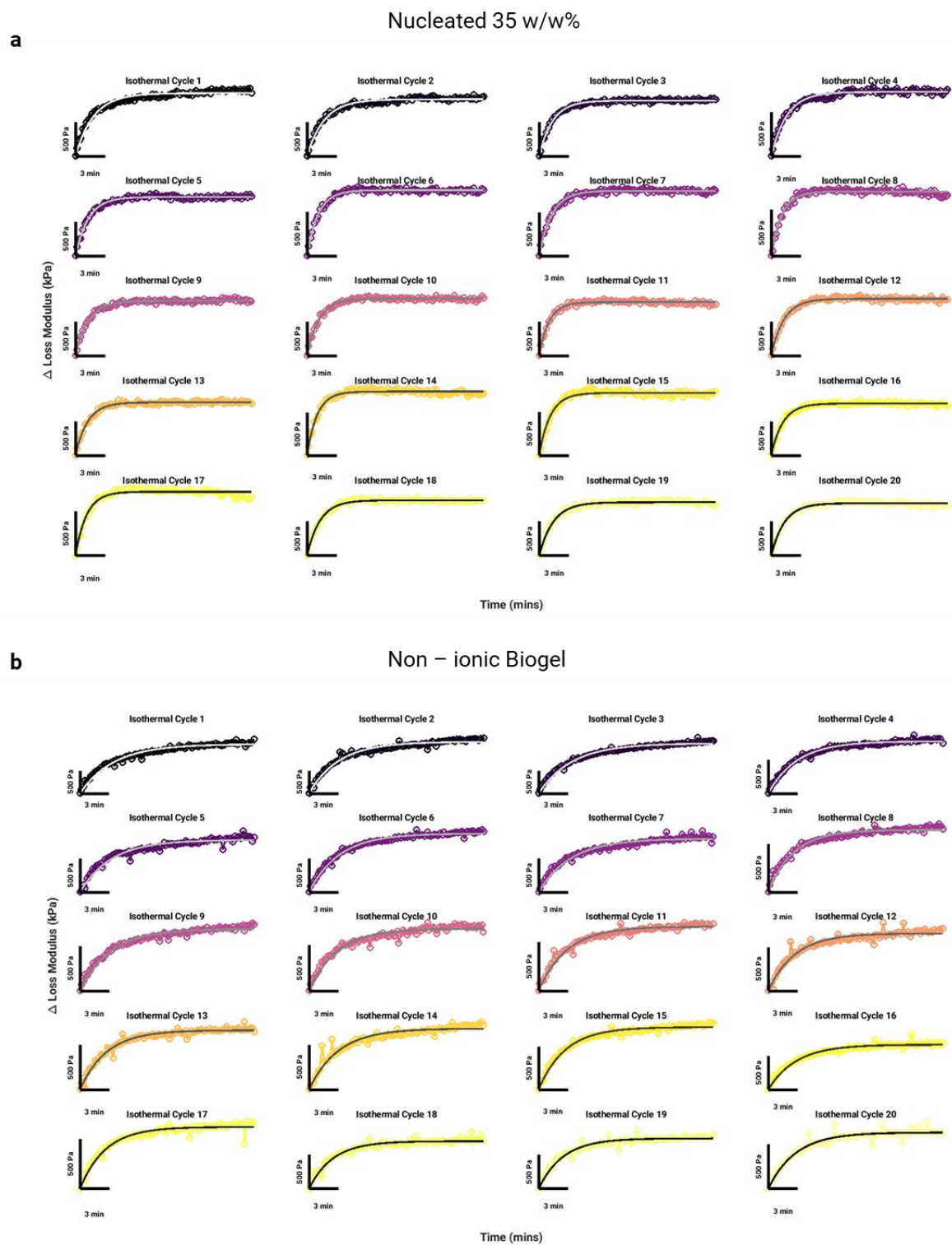


Figure S33: Analysis and Fitting of Loss Modulus for Each Isothermal Cycle. Variation of loss modulus of **(a)** nucleated 35 w/w% and **(b)** non-ionic biogel across different isothermal cycles lasting for 15 mins, after the cooling cycle.

Supplementary Figure 34

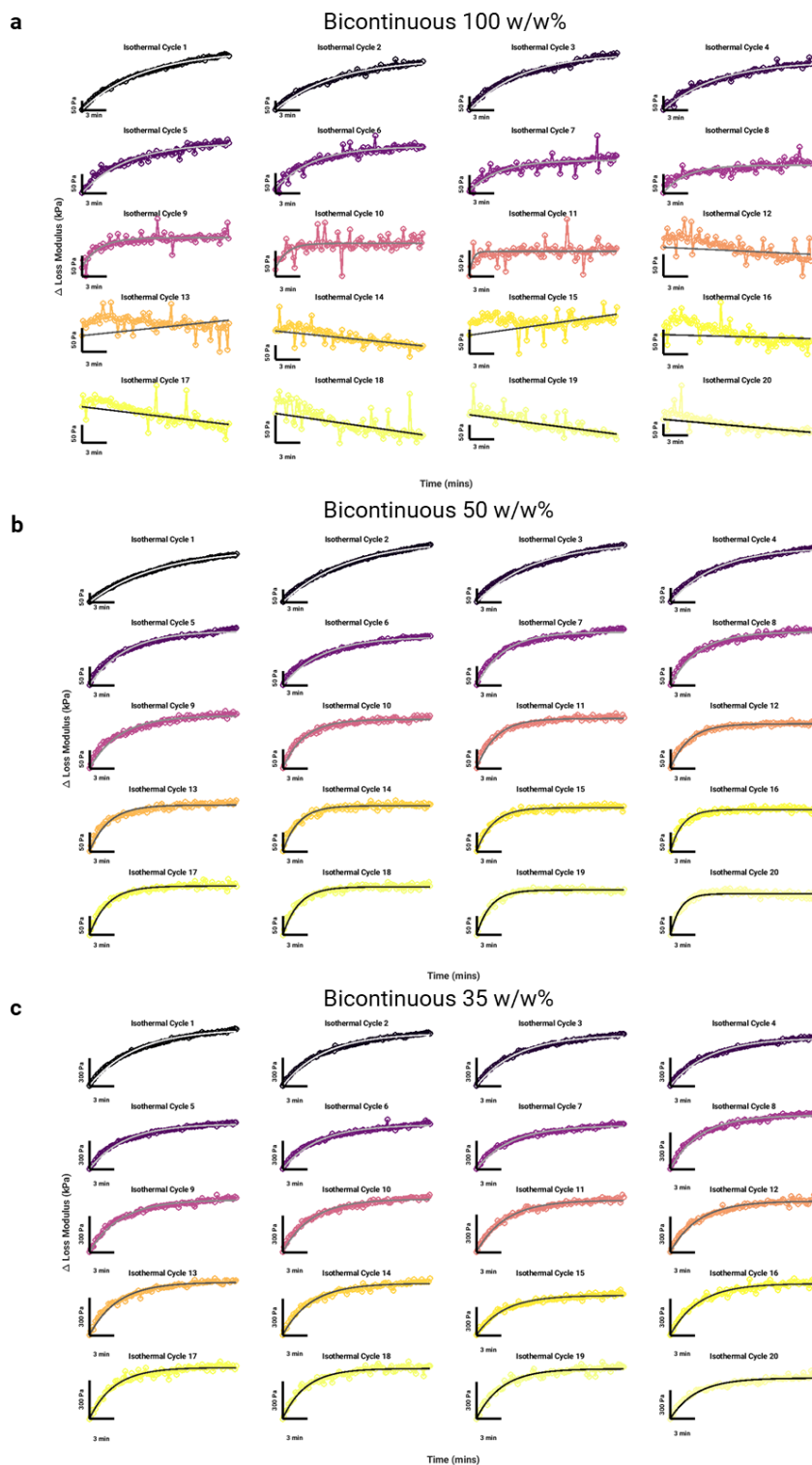


Figure S34: Analysis and Fitting of Loss Modulus for Each Isothermal Cycle. Variation of loss modulus of bicontinuous (a) 100 w/w%, (b) 50 w/w%, and (c) 35 w/w% across different isothermal cycles lasting for 15 mins, after the cooling cycle.

Supplementary Figure 35

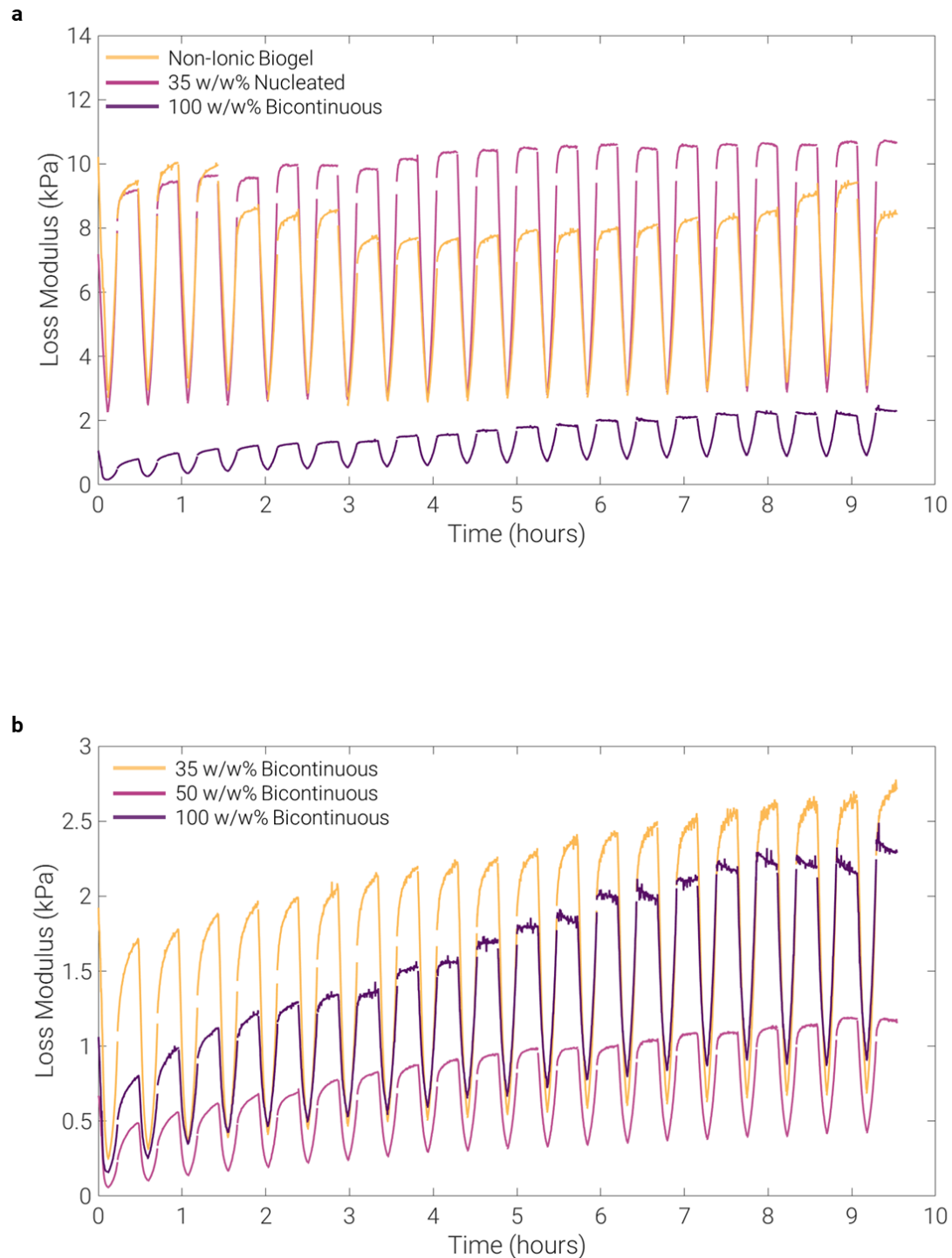


Figure S35: Variation of Loss Modulus across Cycles for different bicontinuous systems. Comparison of loss modulus for **(a)** non-ionic biogel, nucleated 35 w/w%, and bicontinuous 100 w/w%, and **(b)** bicontinuous 35 w/w%, 50 w/w%, and 100 w/w%.

Supplementary Figure 36

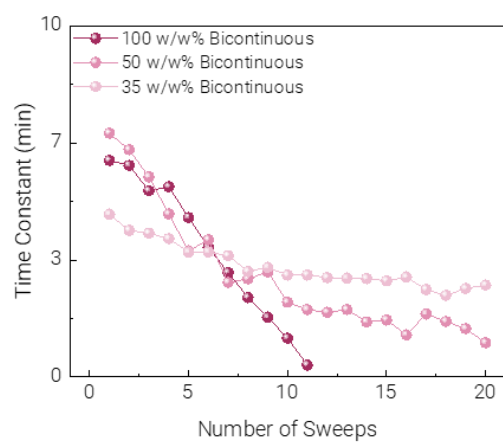


Figure S36: Comparison of the time constant of isothermal relaxation of loss modulus over 20 thermal cycles for bicontinuous 35 w/w%, 50 w/w%, and 100 w/w%.

Supplementary Figure 37

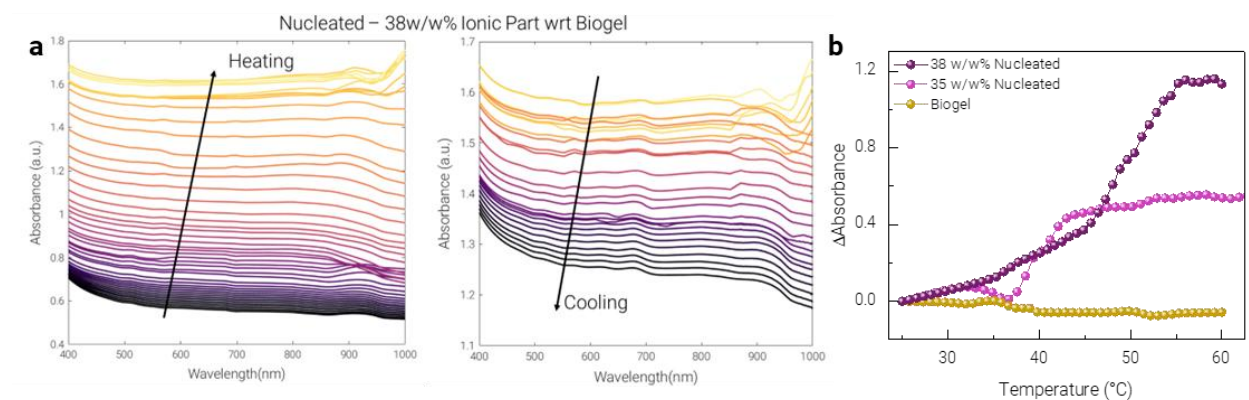


Figure S37: Variation of UV-Vis Absorbance with Temperature for Different Material Systems. (a) UV-Vis absorbance during heating and cooling for nucleated 38 w/w%. **(b)** The onset of turbidity in nucleated 35 w/w% and 38 w/w% compared to non-ionic biogel.

Supplementary Figure 38

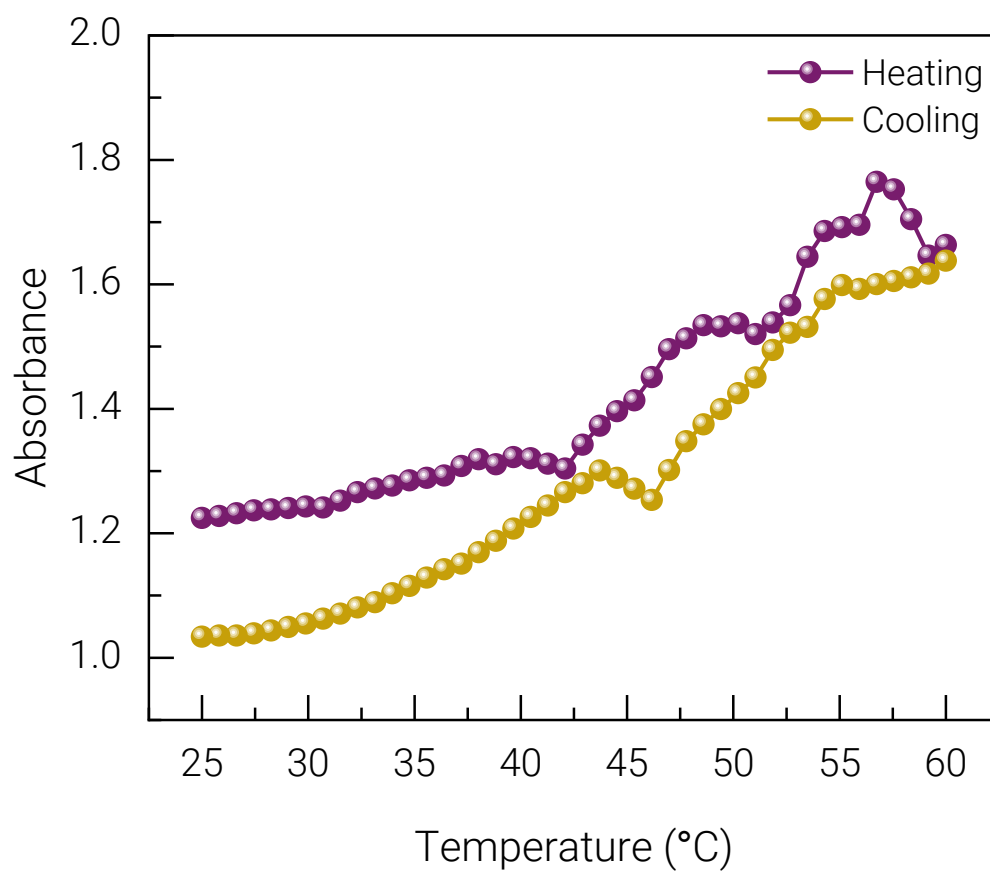


Figure S38: UV-Vis Temperature Turbidity test of BC-NP 50 w/w%.

Supplementary Figure 39

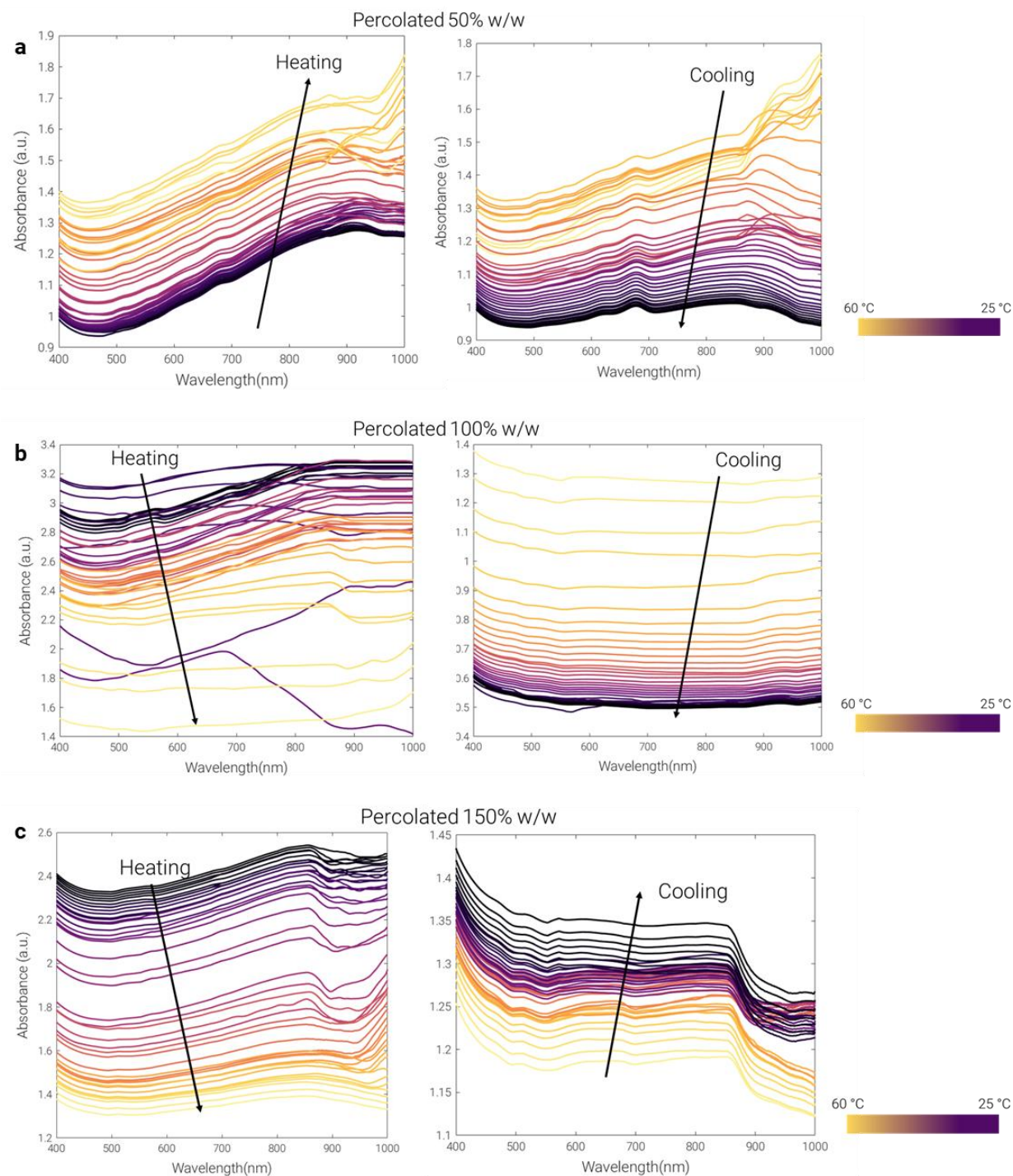


Figure S39: Variation of UV-Vis Absorbance with Temperature for the Bicontinuous System. UV-Vis absorbance during heating and cooling for bicontinuous (a) 50 w/w%, (b) 100 w/w%, and (c) 150 w/w%.

Supplementary Figure 40

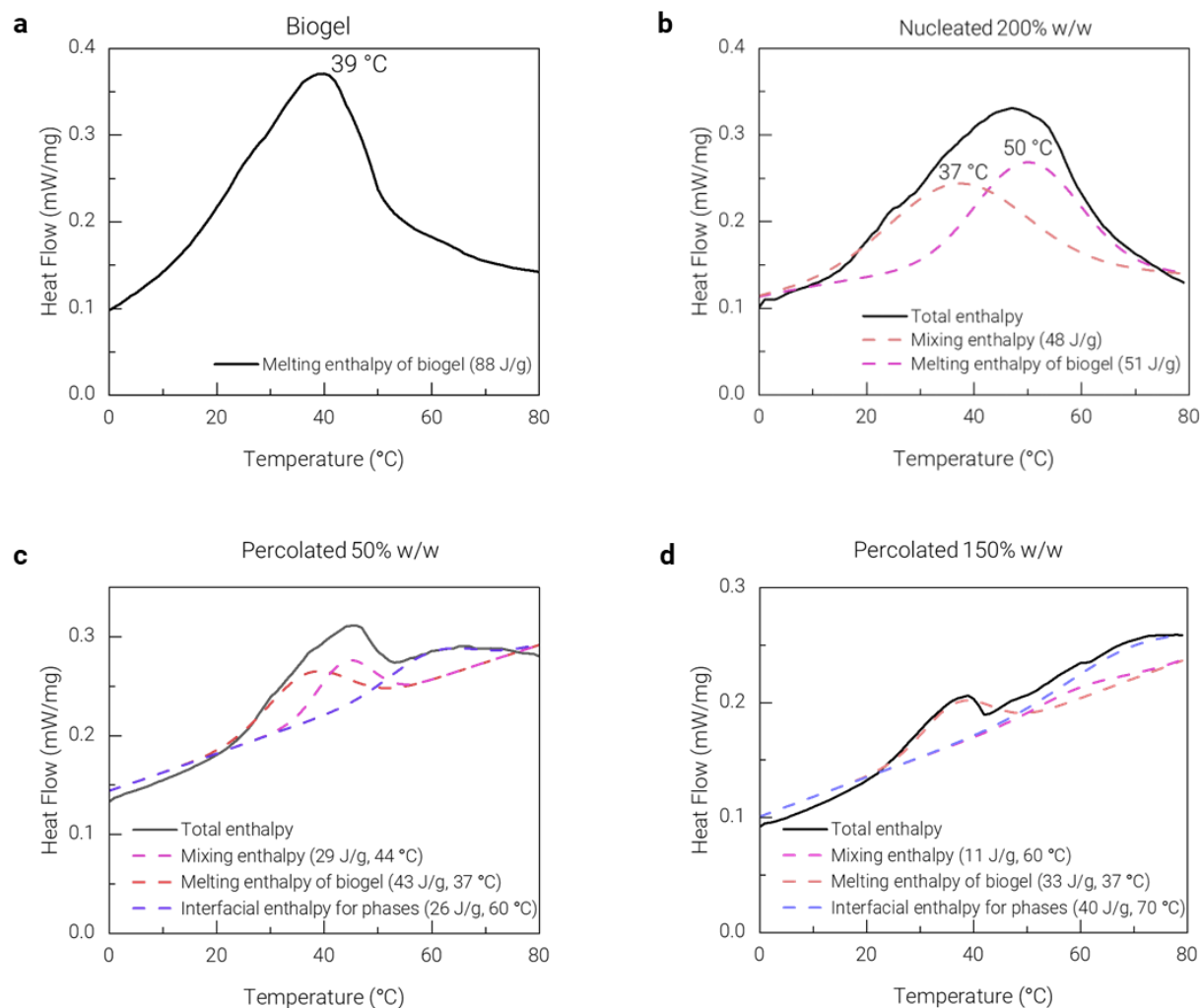


Figure S40: Differential Scanning Calorimetry (DSC) analysis of different materials systems. Thermal analysis showing different enthalpies such as melting, mixing, and interfacial enthalpy for **(a)** biogel (only melting), **(b)** nucleated 200 w/w% (mixing and melting), and bicontinuous **(c)** 50 w/w% and **(d)** 150 w/w%.

Supplementary Figure 41

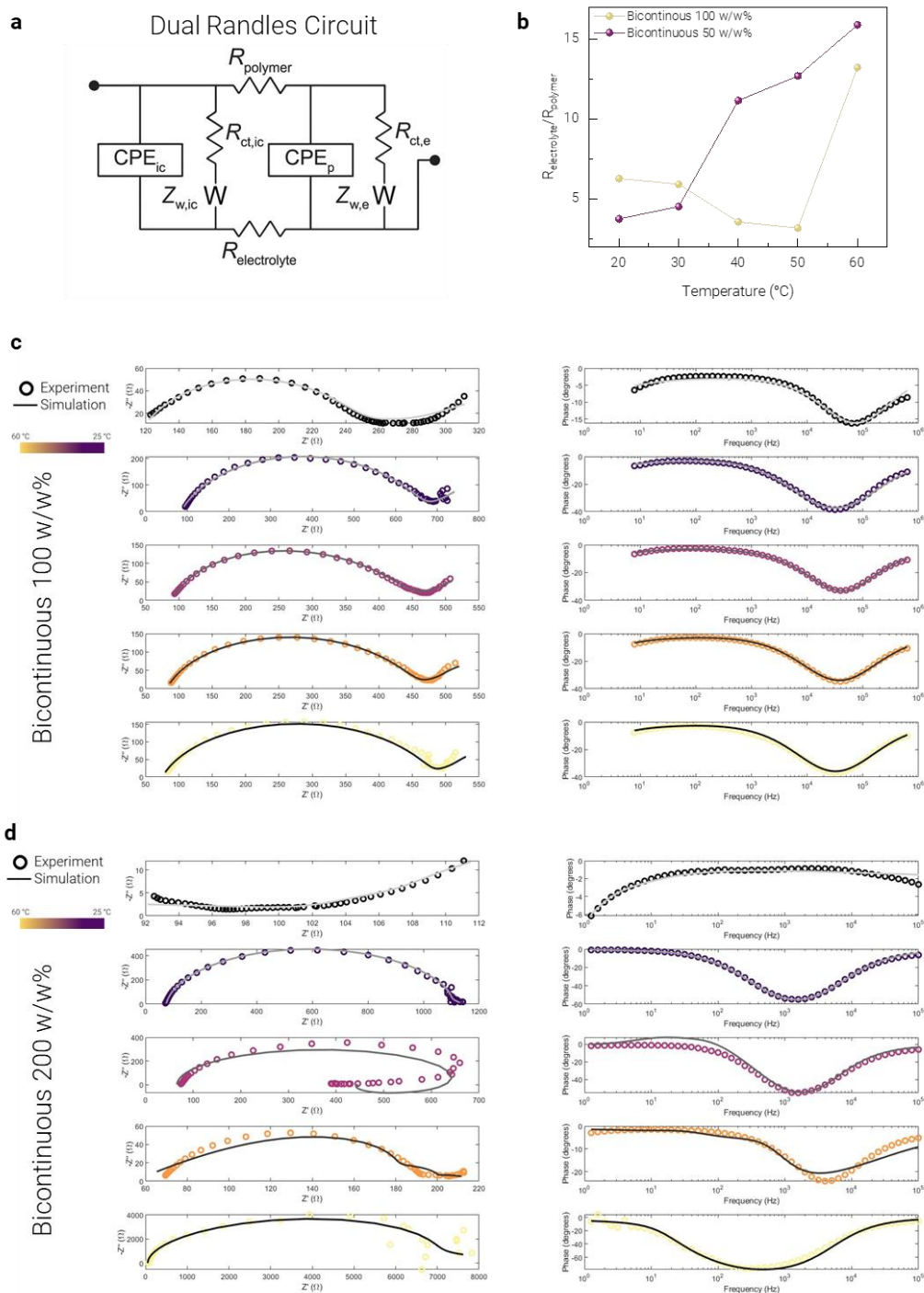


Figure S41: Variation of Electrochemical Impedance Spectroscopy with Temperature for Bicontinuous System. (a) Schematic of the Dual Randles model to simulate EIS and extract parameters with (b) extracted parameter $R_{\text{electrolyte}}/R_{\text{polymer}}$ as a function of temperature. Variation of EIS as a function of temperature (25°C, 30°C, 40°C, 50°C, and 60°C) for bicontinuous (c) 100 w/w% and (d) 200 w/w%.

Supplementary Figure 42

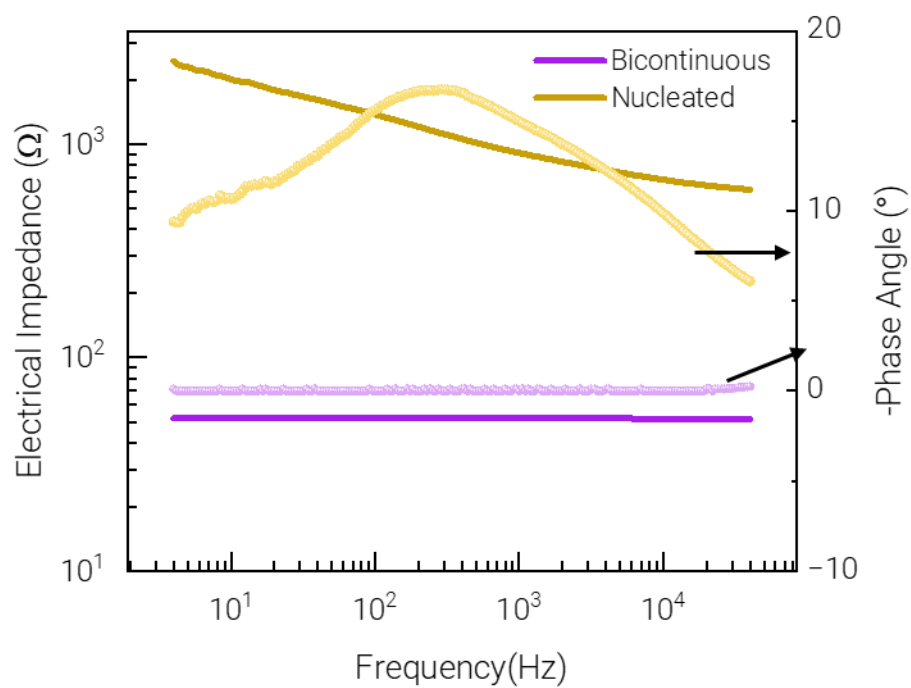


Figure S42: Comparison of Electrical Impedance Spectra of Agarose-based ionic biogel for BC-MP (100 w/w%) and NC (100 w/w%).

Supplementary Figure 43

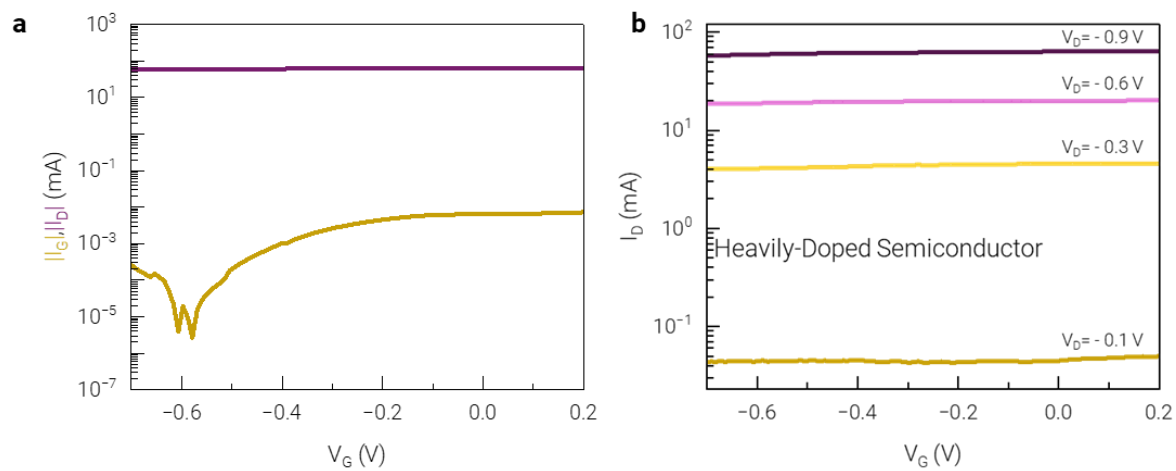


Figure S43: Highly Doped Semiconducting Characteristics of Layered 100 w/w%. (a) Transfer characteristics with a bias of -0.9 V and (b) transfer characteristics showing the typical ohmic-like electrical transport analogous to a degenerately doped semiconductor

Supplementary Figure 44

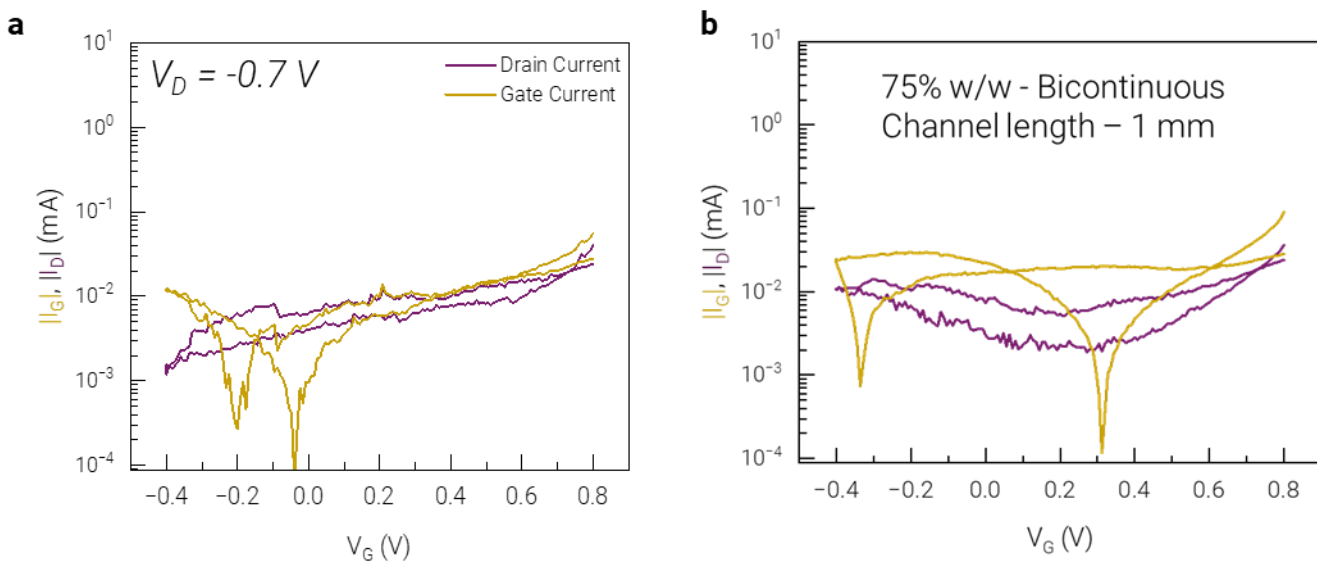


Figure S44: Transfer Characteristics of gate leakage-dominated OECT with mobile ions acting as charge carriers using the (a) Nucleated 100 w/w% and (b) Bicontinuous 75 w/w% channel material. The drain bias is -0.7 V.

Supplementary Figure 45

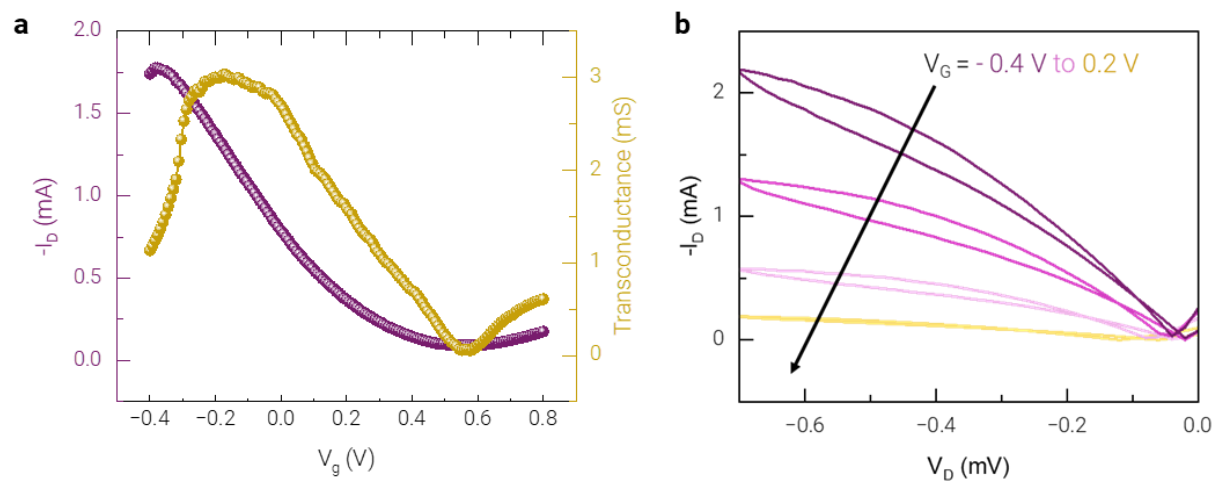


Figure S45: Characteristics of a semiconducting channel based on Bicontinuous 100 w/w% with proper electrostatic modulation by the gate. (a) Transfer characteristics and transconductance with a bias of -0.7 V and (b) output characteristics.

Supplementary Figure 46

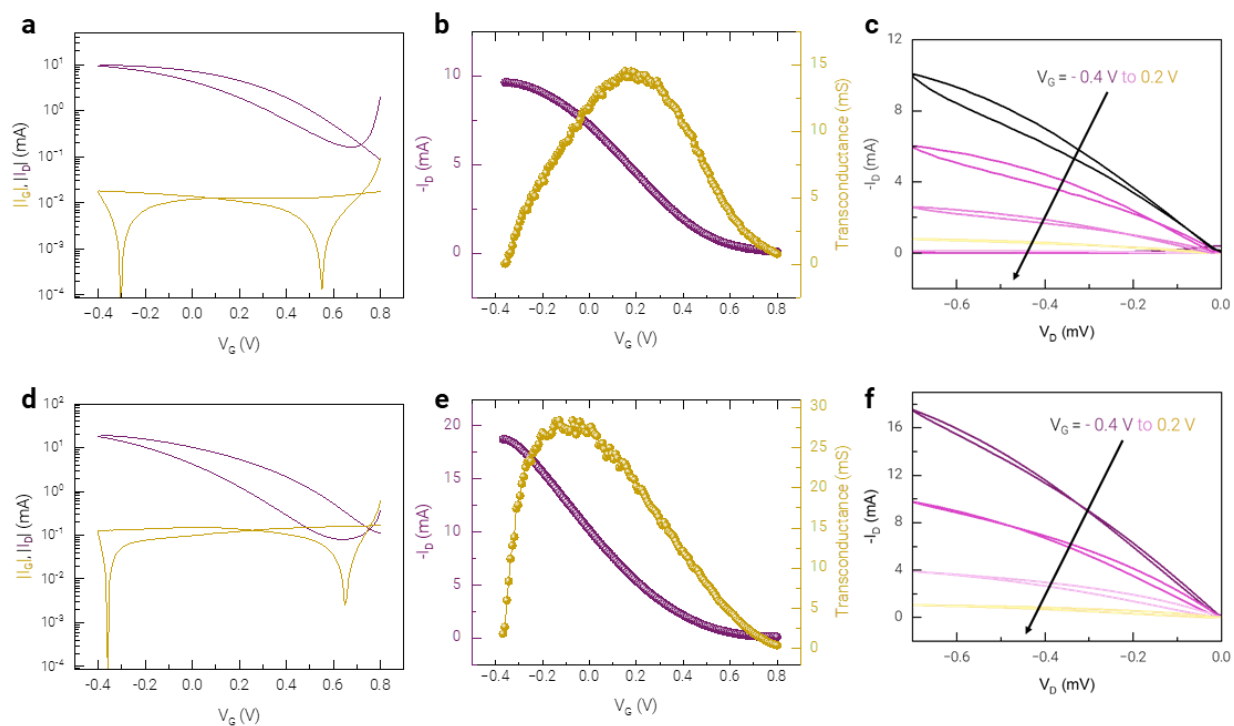


Figure S46: Semiconducting Characteristics of the Bicontinuous System. (a,d) Transfer characteristics, (b,e) transconductance, and (c,f) output characteristics of bicontinuous (a-c) 200 w/w% and (d-f) 300 w/w%. The drain bias is -0.7 V.

Supplementary Figure 47

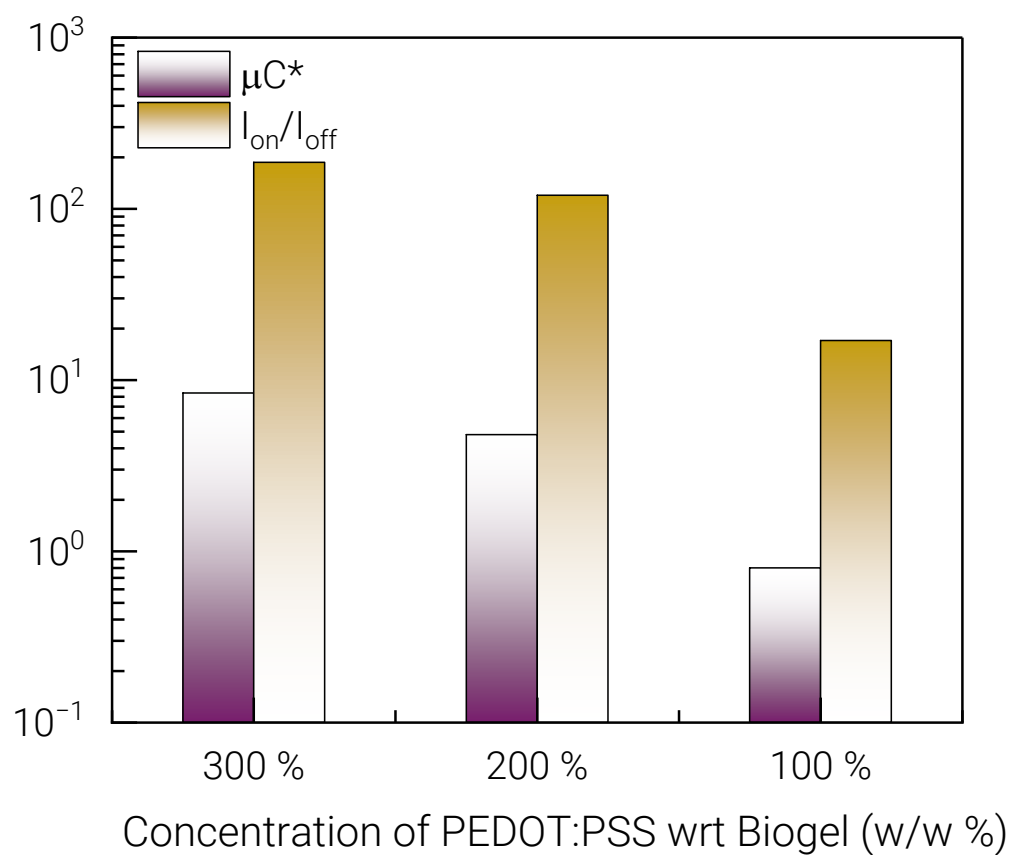


Figure S47: Variation of on-off ratio and figure of merit of BC-MP 100 w/w%, BC-PP 200 w/w%, and 300 w/w%. The semiconducting property improves with increases in the concentration of PEDOT:PSS due to the formation of more coherent charge transport.

Supplementary Figure 48

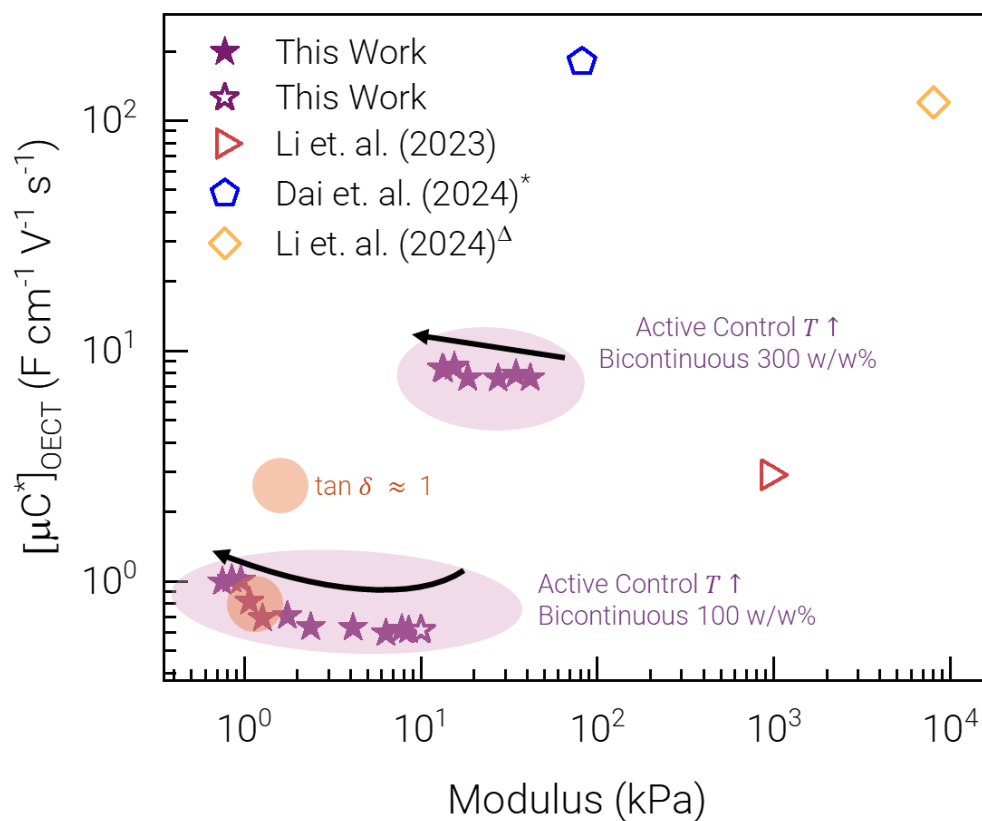


Figure S48: Comparison of Figure of Merit in Semiconducting Hydrogels. This work enables the active control of modulus using temperature due to its stimuli-responsive and physically crosslinking nature.

*Here $[\mu]_{OECT}[C^*]_{OECT}$ is extracted from the literature instead of $[\mu C^*]_{OECT}$.(8)

^ΔN-type semiconducting hydrogels, whereas the rest are p-type hydrogels.(11)

Supplementary Figure 49

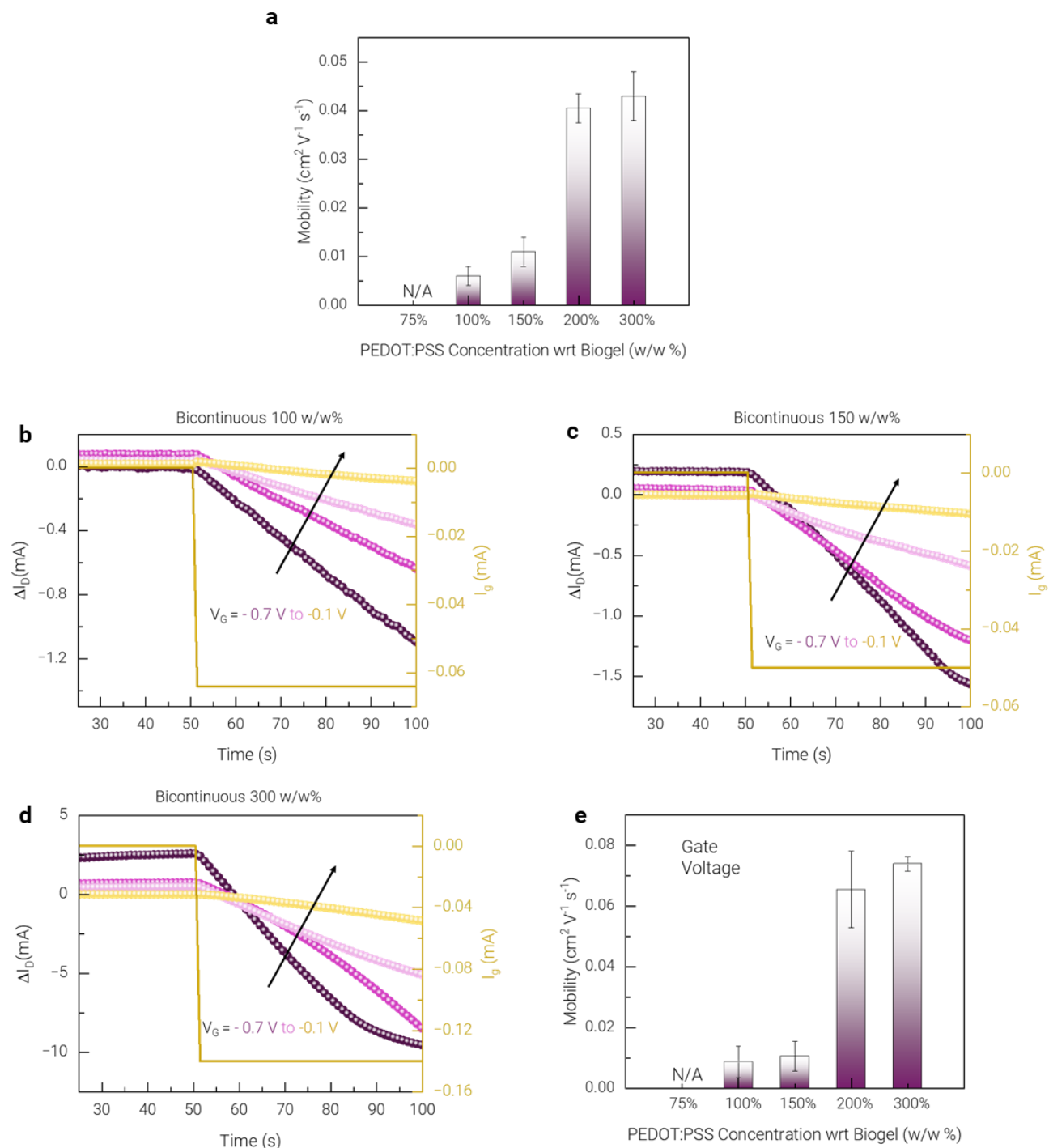


Figure S49: Mobility of the Bicontinuous System. (a) Mobility calculated using constant gate current for bicontinuous (b) 100 w/w%, (c) 150 w/w%, and (d) 300 w/w%. (e) Mobility calculated using sinusoidal gate voltage.

Supplementary Figure 50

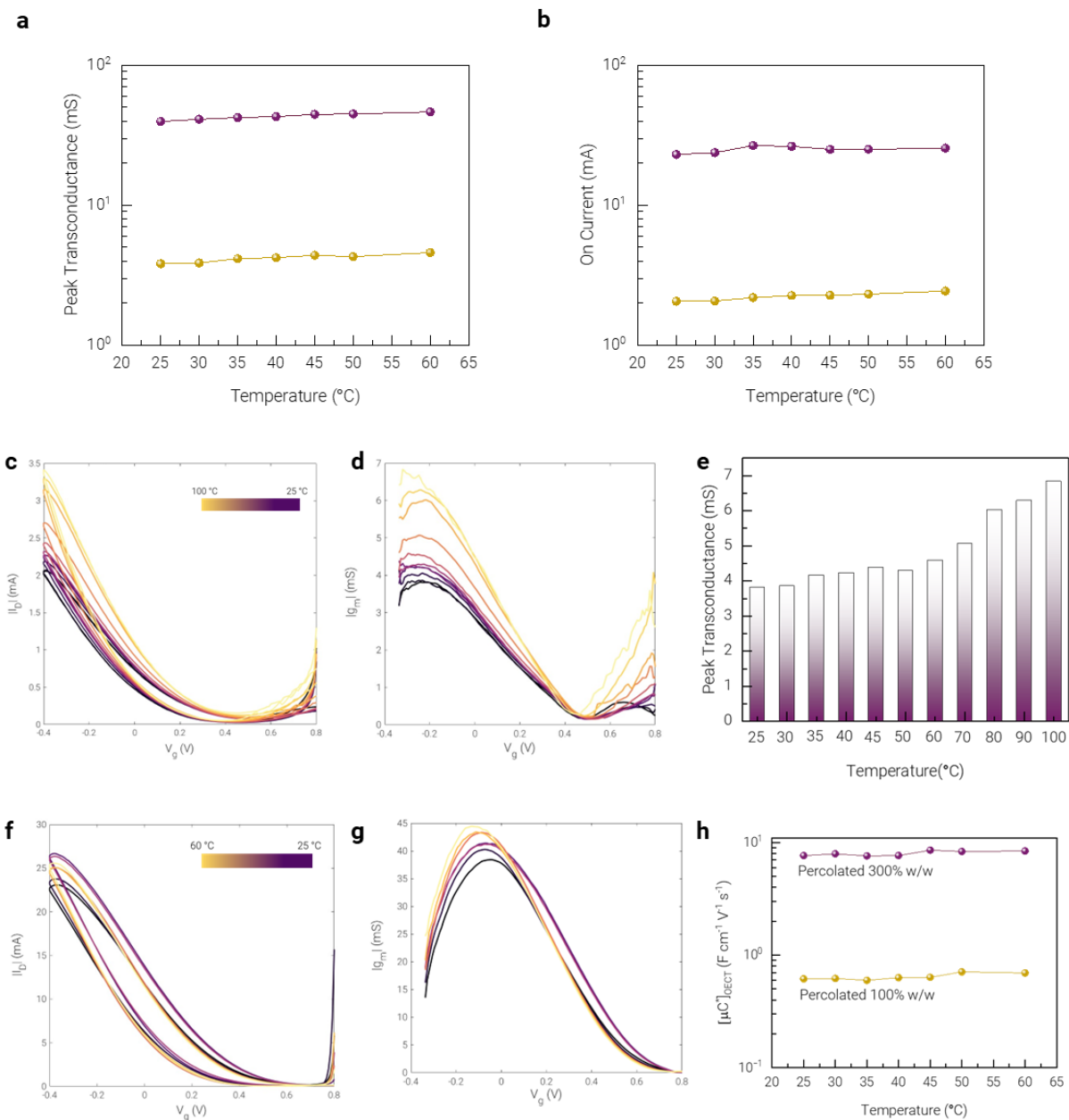


Figure S50: Semiconducting Properties of the Bicontinuous System as a Function of Temperature. (a) Peak Transconductance and (b) On Current as a function of Temperature. (c) Transfer curve and (d) transconductance as a function of gate voltage, and (e) peak transconductance as a function of temperature of bicontinuous 100 w/w%. (f) Transfer curve and (g) transconductance as a function of gate voltage for bicontinuous 300 w/w%. (h) Figure of merit of bicontinuous 300 w/w% and 100 w/w% as a function of temperature. The drain bias is -0.7 V.

Supplementary Figure S1

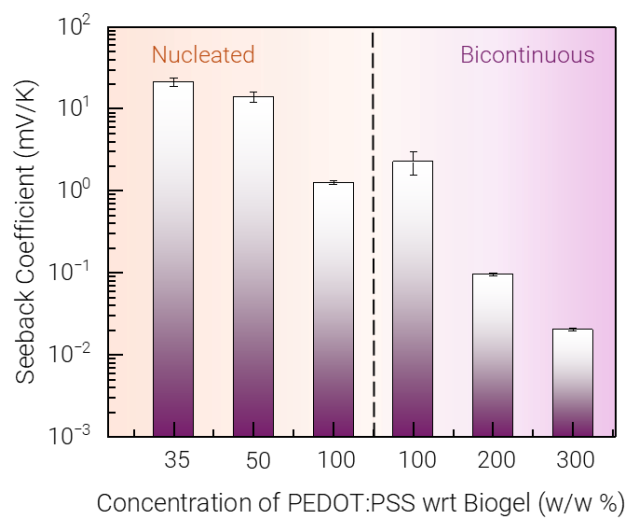


Figure S51: Variation of Seeback coefficient for NC and BC-MP and BC-PP systems with different PEDOT:PSS loadings.

Supplementary Figure 52

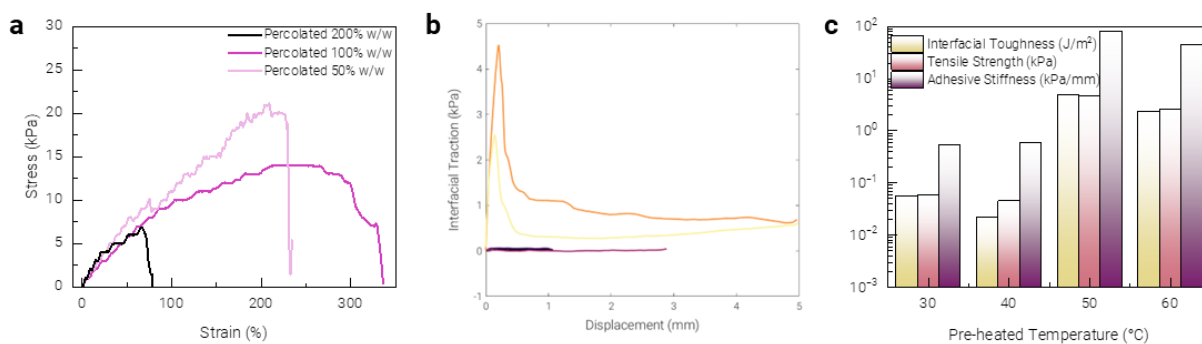


Figure S52: Mechanical and Adhesion Properties of the Bicontinuous System. (a) Tensile testing of bicontinuous 200w/w%, 100 w/w%, and 50 w/w%. (b-c) Tensile adhesion testing of bicontinuous 100 w/w% as a function of temperature.

Supplementary Figure S3

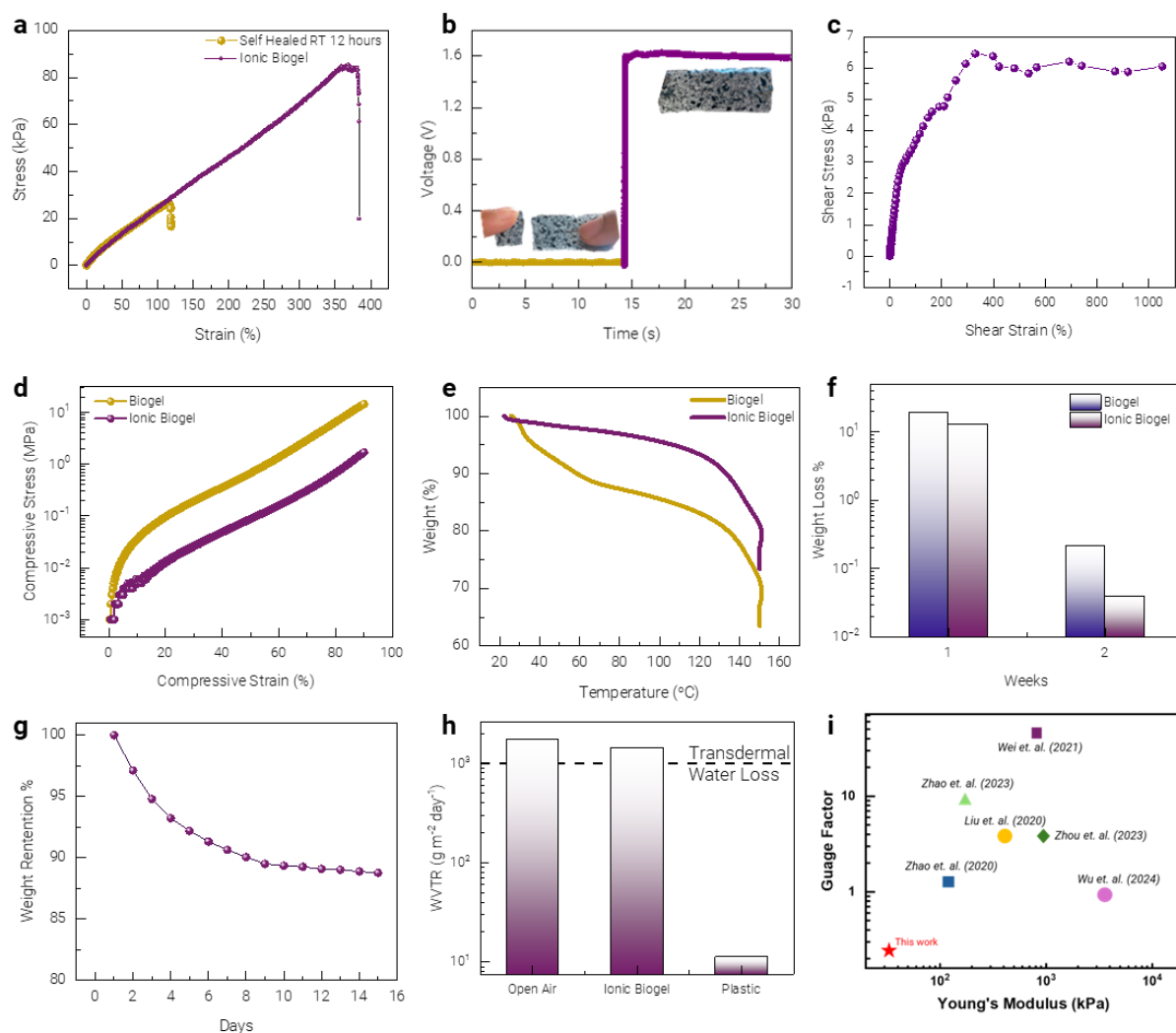


Figure S53: Mechanical Properties of the Nucleated 35 w/w% System. (a) Tensile testing of freshly prepared nucleated 35 w/w% and 12-hour self-healed ionic biogel. (b) Demonstration of self-healing ionic biogel using the immediate onset of electrical conductivity. (c) Shear test, (d) compression test, (e) thermogravimetric analysis (TGA), and (f) weight loss or (g) retention of nucleated 35 w/w% compared to non-ionic biogel. (h) Water vapor transmission rate (WVTR) of ionic biogel compared to open air and plastic. (i) Record strain-insensitivity of ionic biogel compared to other literature reports.

Supplementary Figure 54

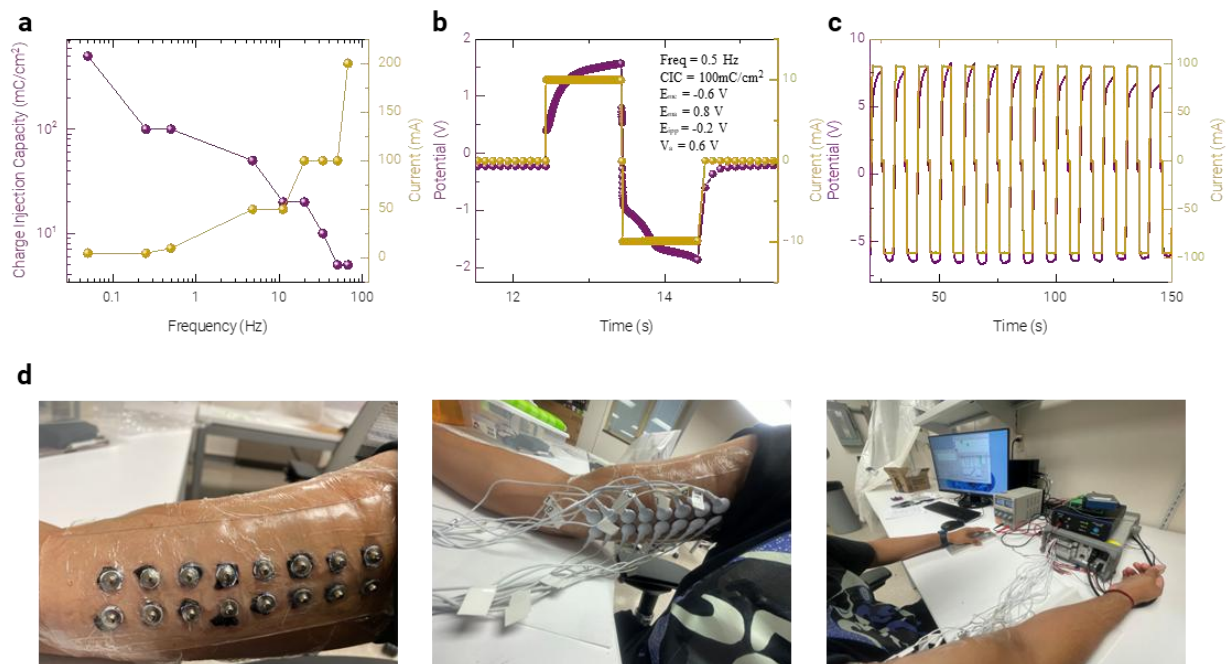


Figure S54: Electrical Stimulation Characterization of the Nucleated 35 w/w% System. (a) Charge injection capacity and current injected as a function of frequency. Voltage transient with (b) 10 mA current at a frequency of 0.5 Hz and (c) 100 mA current at a frequency of 0.1 Hz. (d) Wearable stimulator array and the electrical stimulation experimental setup.

Supplementary Figure S55

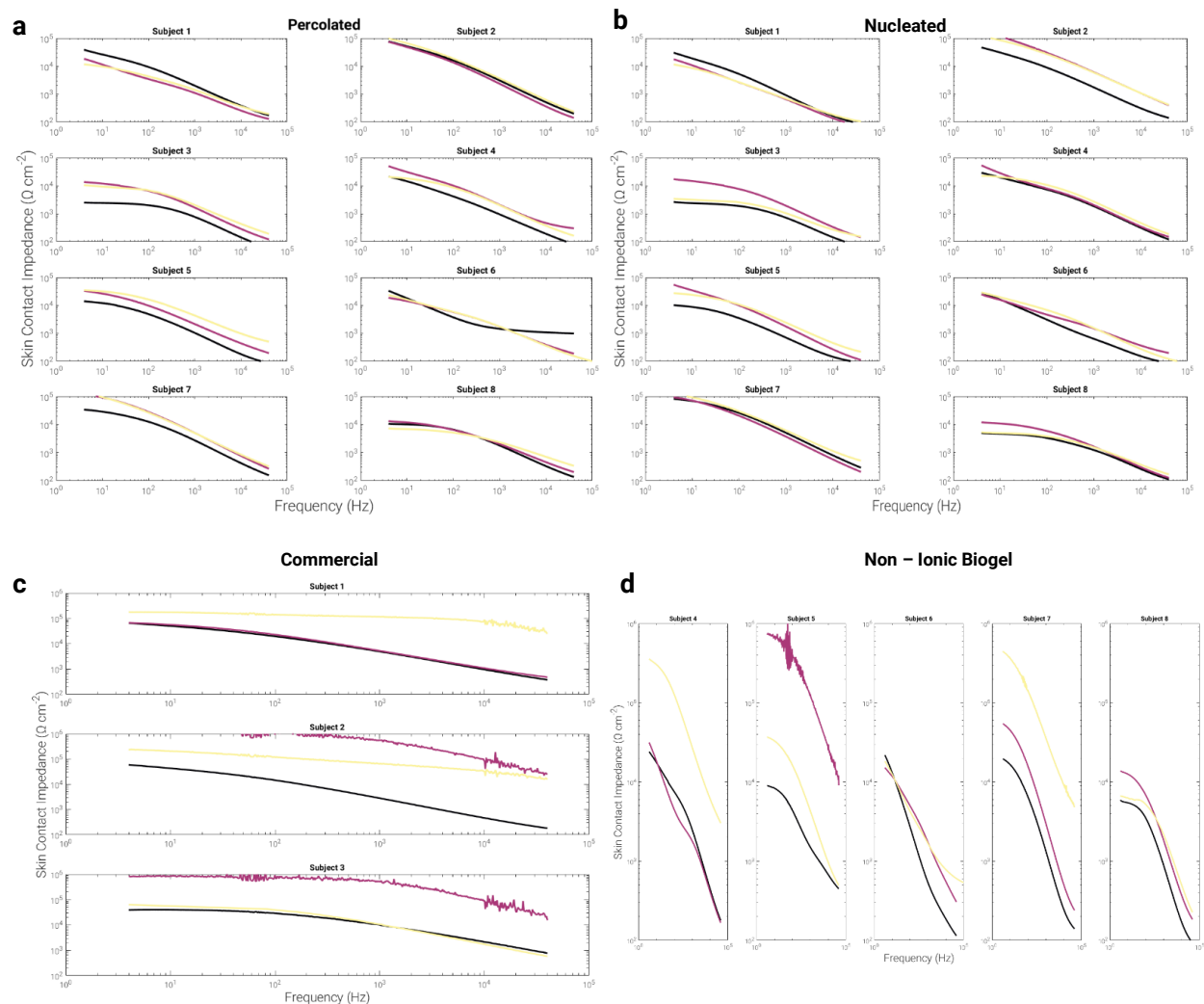


Figure S55: Skin-contact Impedance Spectra across Different Subjects for Different Material Systems. Skin-contact impedance across different subjects and days for (a) bicontinuous 100 w/w%, (b) nucleated 100 w/w%, (c) commercial, and (d) non-ionic biogel.

Supplementary Figure 56

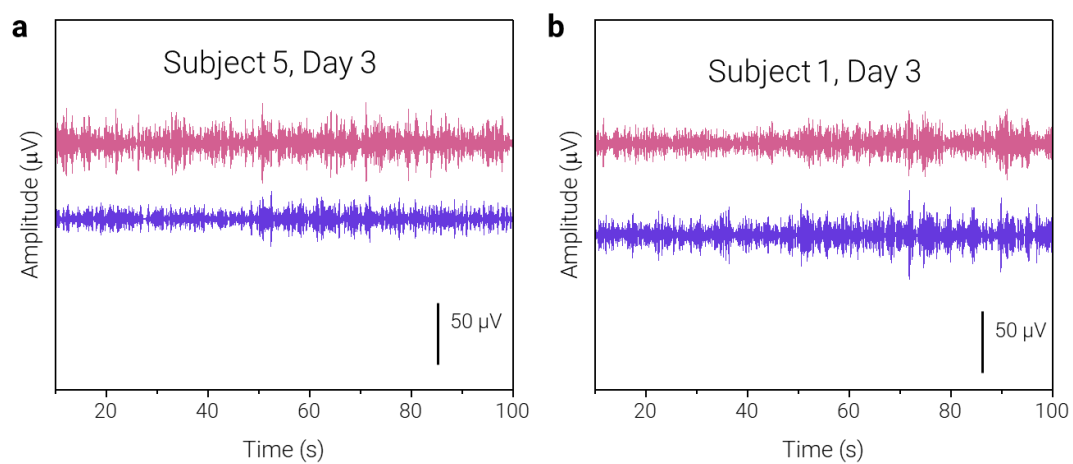


Figure S56: Signal fidelity (alpha band) under difficult physiological conditions. Day 3 of (a) Subject 5 after workout, accumulating sweat and salt, and (b) Subject 1 with accumulation of skin oil by not taking a shower.

Supplementary Figure 57

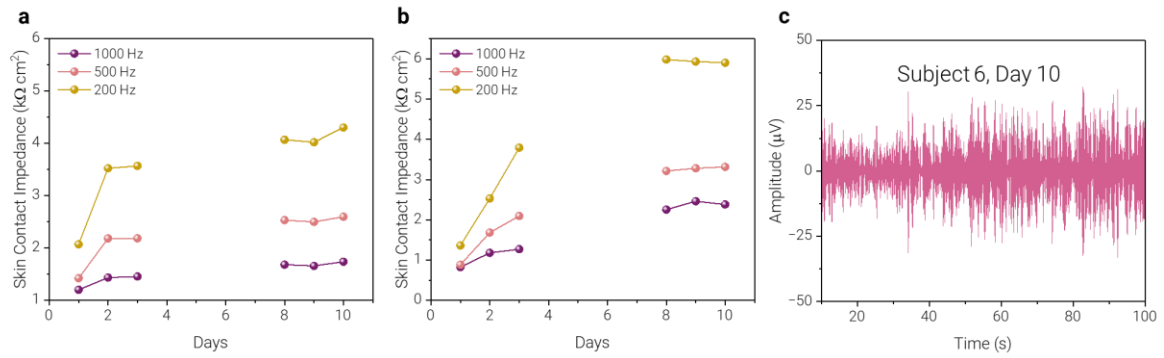


Figure S57: Skin contact impedance of Subject 6 after 10 days of reusing the EEG electrodes with (a) BC-MP and (b) BC-NP. (c) The alpha band signal of Subject 6 at Day 10 using BC-MP.

Supplementary Figure 58

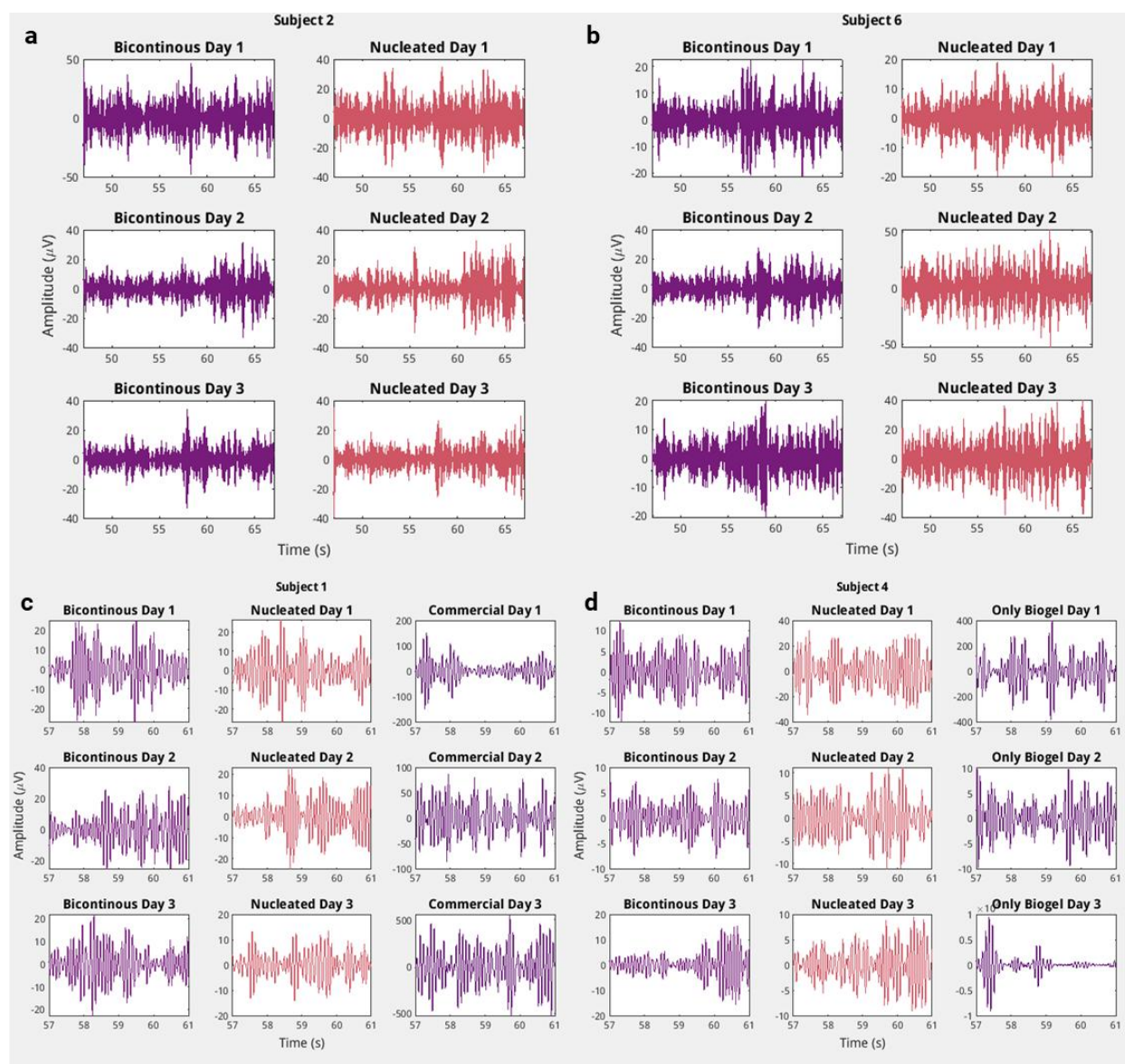


Figure S58: EEG Signal from Various Subjects across Days using Different Material Systems. EEG signal from (a) subject 2 and (b) subject 6 using bicontinuous 100 w/w% and nucleated 100 w/w% across days 1, 2, and 3. (c) EEG signal from subject 1 compared to commercial across three days. (d) EEG signal from subject 4 compared to non-ionic biogel across three days.

Supplementary Figure 59

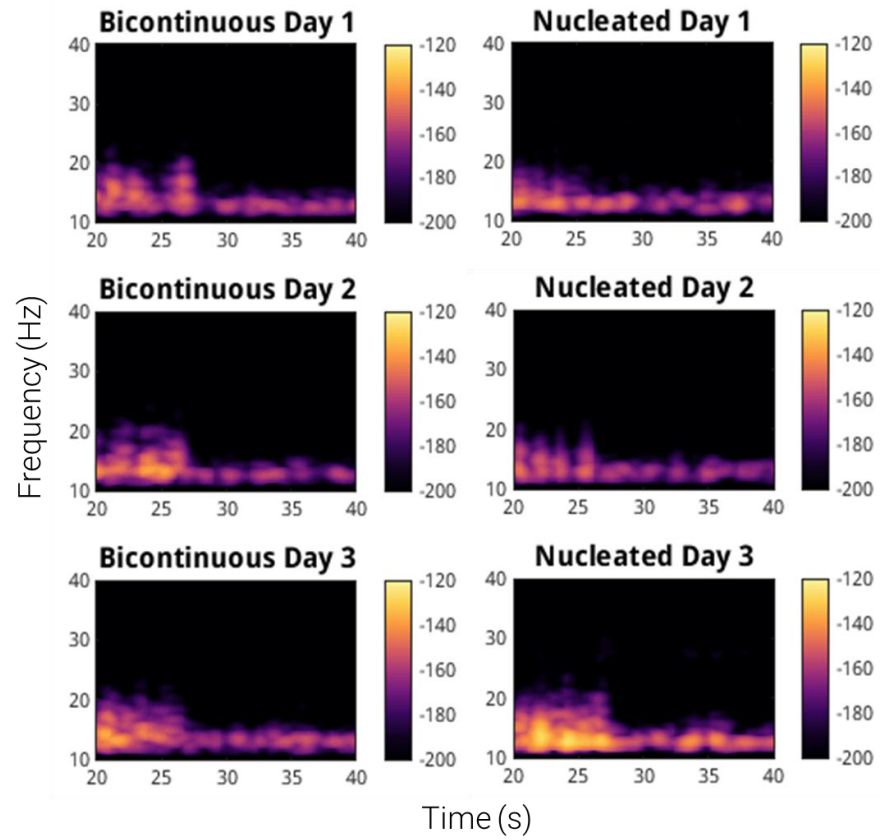


Figure S59: EEG spectrogram showing Event Related Desynchronization (ERD) using Bicontinuous and Nucleated material systems across days 1, 2, and 3.

Supplementary Figure 60

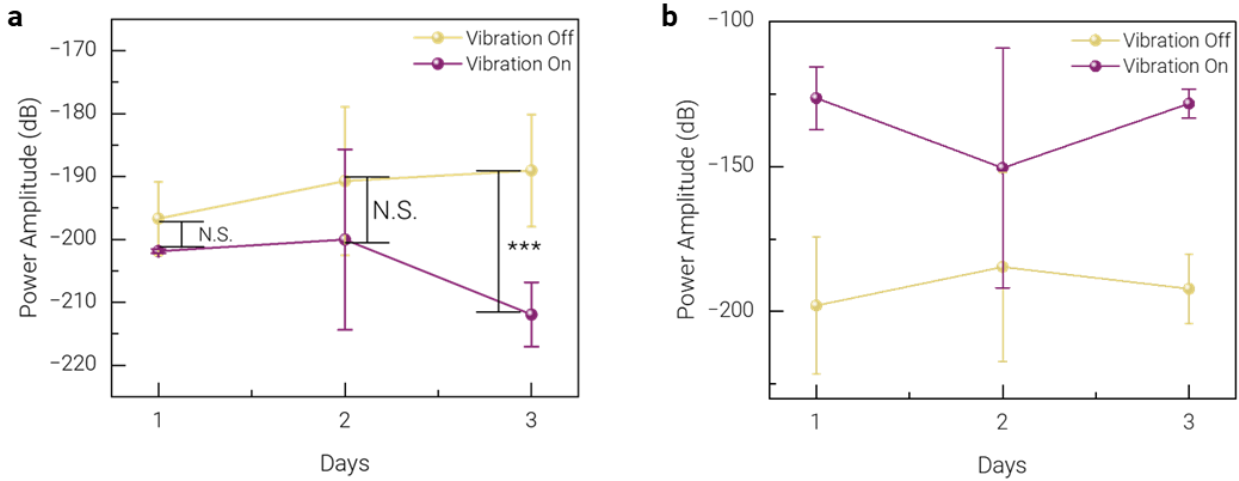


Figure S60: Power amplitude showing event-related desynchronization (ERD) during sensory mechanical vibration using (a) nucleated electrodes and (b) commercial electrodes over 3 days.

Supplementary Figure 61

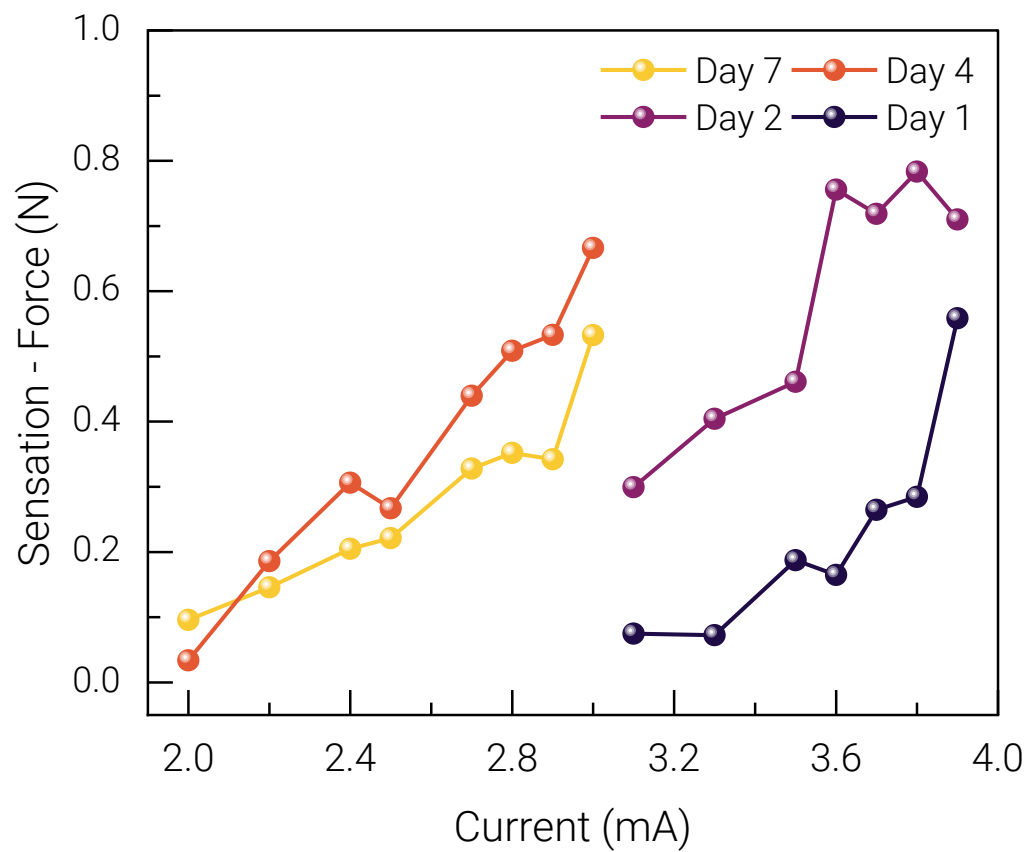


Figure S61: Perceived Sensation of the Subject across Different Days of Stimulation.

Supplementary Figure 62

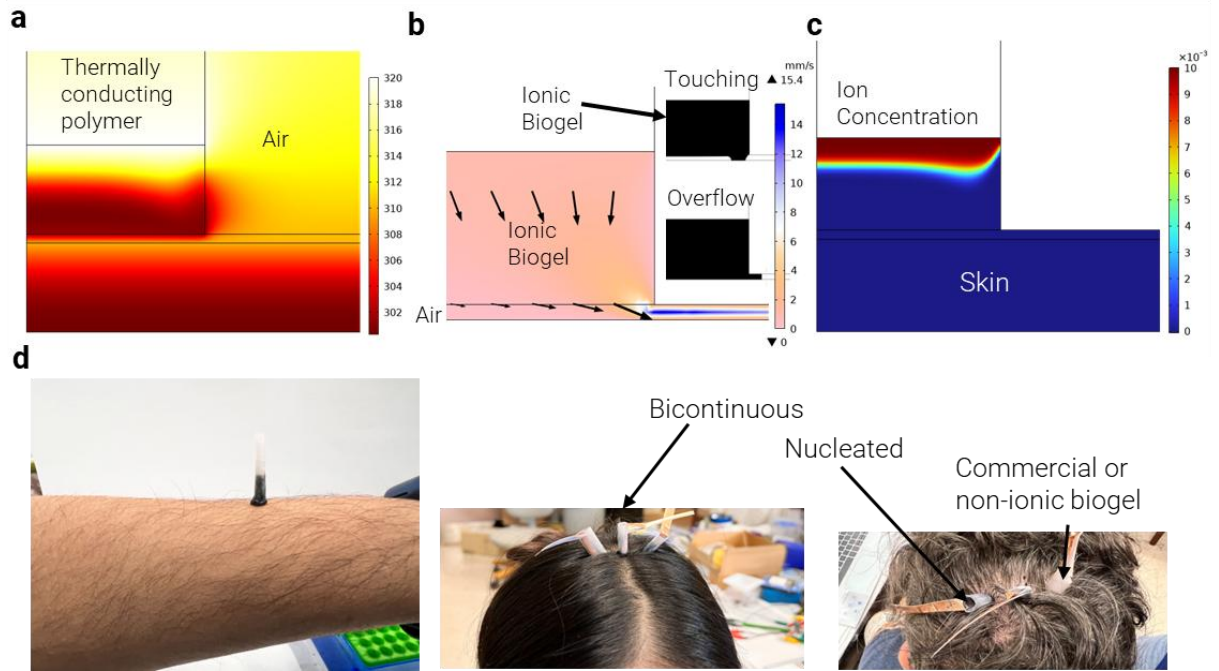


Figure S62: Deployment and Analysis of the Self-balancing EEG Electrode. COMSOL simulation for **(a)** temperature distribution, **(b)** phase field flow and distribution, and **(c)** ion concentration distribution of the ionic biogel. **(d)** Optical images of the deployment of EEG electrodes on the arm (left) and on the scalp with hair (right).

Supplementary Figure 63

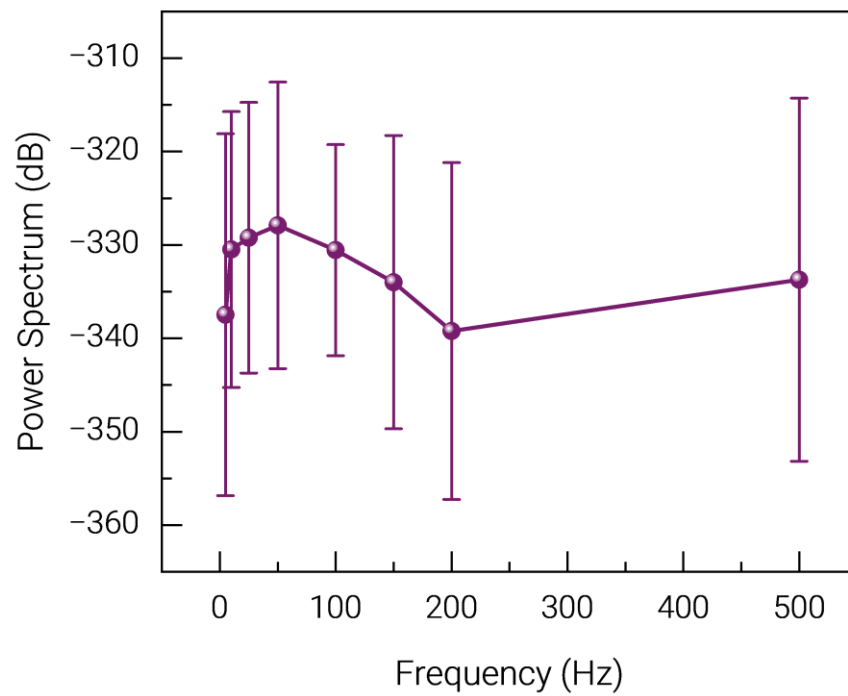


Figure S63: Power spectrum of EEG during haptic sensation applied at different frequencies (5 Hz – 500 Hz) on day 7.

Supplementary Figure 64

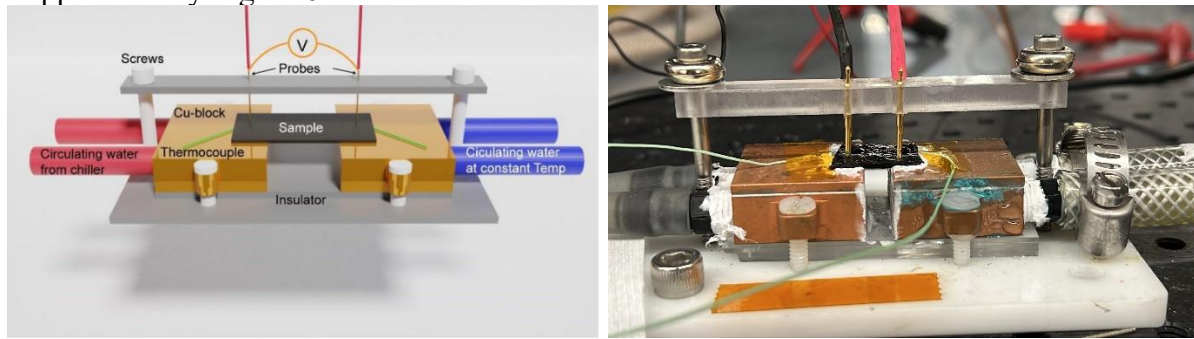


Figure S64: Schematic diagram and optical image of the custom-built system to measure the Seebeck coefficient.

Supplementary Table 1

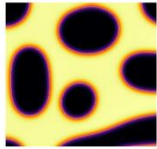
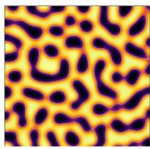
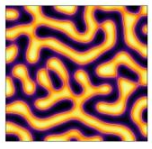
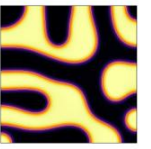
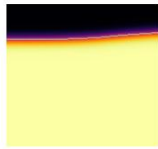
Controlling Phases Morphology using Mixing Strategy					
Nucleated <i>Complete Mixing</i>	Bicontinuous <i>Incomplete Mixing</i>			Layered <i>No Mixing</i>	
Nucleated (NC)	Non-Percolated (BC-NP)	Marginally Percolated (BC-MP)	Permanent Percolated (BC-PP)		Phase Morphology
					Phase-Separation
~ 5 – 10 μm	~ 100 μm	~ 1000 μm	> 2000 μm	> 5 mm	Phase Length
No Doping	No Doping	Moderate Doping	Good Doping	Degenerate Doping	Semiconducting
~ 10 mS/cm	~ 0.1 S/cm	~ 10 S/cm	~ 30 S/cm	> 120 S/cm-	Conductivity
Phase - Transition	Phase - Transition	Phase - Transition	No Phase - Transition		Rheology
Onsets Turbidity	Onsets Turbidity	Turbidity	No Turbidity		UV-Vis
Entropy- Dominated	Enthalpy- Dominated	Enthalpy- Dominated	- Enthalpy- Dominated		DSC

Table S1: Controlling the phase morphology using mixing strategy: Nucleated, Non-Percolated Bicontinuous, Marginally Percolated Bicontinuous, Permanently Percolated Bicontinuous and Layered System.