

Thermal-history encoding through ionic-state programming in a monolithic organic thermosynapse

Do Hwan Kim

dhkim76@hanyang.ac.kr

Hanyang University <https://orcid.org/0000-0003-3003-8125>

Zhengyang Kong

Hanyang University

Jin Han Kwon

Korea Advanced Institute of Science and Technology

Ji Hong Kim

Hanyang University

Hanbin Choi

North Carolina State University <https://orcid.org/0009-0008-7561-1128>

Jonghyun Yun

Hanyang University

Seonghwan Yun

Hanyang University

Hyeon Woo Yang

Korea Advanced Institute of Science and Technology

Hyun Joon Shin

Hanyang University

So Young Kim

Hanyang University

Yong Min Kim

Jeonbuk National University

Hong Chul Moon

Korea Advanced Institute of Science and Technology <https://orcid.org/0000-0003-2598-0925>

Article

Keywords:

Posted Date: October 19th, 2026

DOI: <https://doi.org/10.21203/rs.3.rs-11149224/v1>

License: © ⓘ This work is licensed under a Creative Commons Attribution 4.0 International License.
[Read Full License](#)

Additional Declarations: There is **NO** Competing Interest.

Thermal-history encoding through ionic-state programming in a monolithic organic thermosynapse

Zhengyang Kong^{1†}, Jin Han Kwon^{2†}, Ji Hong Kim^{1†}, Hanbin Choi^{3†}, Jonghyun Yun¹,
Seonghwan Yun¹, Hyeon Woo Yang², Hyun Joon Shin¹, So Young Kim¹, Yong Min Kim^{4,5*},
Hong Chul Moon^{2*}, and Do Hwan Kim^{1,6,7*}

¹Department of Chemical Engineering, Hanyang University, Seoul 04763, Republic of Korea.

²Department of Chemical & Biomolecular Engineering, Korea Advanced Institute of Science and Technology (KAIST), Daejeon 34141, Republic of Korea

³Department of Chemical and Biomolecular Engineering, North Carolina State University, Raleigh, NC 27695, USA

⁴School of Chemical Engineering, School of Semiconductor and Chemical Engineering, Jeonbuk National University, Jeonju 54896, Republic of Korea

⁵Clean Energy Research Center, Jeonbuk National University, Jeonju 54896, Republic of Korea

⁶Institute of Nano Science and Technology, Hanyang University, Seoul 04763, Republic of Korea

⁷Clean-Energy Research Institute, Hanyang University, Seoul 04763, Republic of Korea

†These authors contributed equally to this work.

*Corresponding authors' email: yongmink@jbnu.ac.kr (Y.M.K.); hcmoon@kaist.ac.kr (H.C.M); dhkim76@hanyang.ac.kr (D.H.K.)

Abstract

Encoding thermal history directly in soft ionic matter requires a material in which temperature controls not only ion transport but also the persistence of the resulting ionic state. Here we report a bio-inspired monolithic thermosynapse that encodes thermal history through ionic-state programming within a single organic electrochemical transistor. A zwitterionic gel with an upper critical solution temperature regulates ion confinement and release across its phase transition, producing an approximately 922% relative increase in ionic conductivity. Field-induced polarization further stabilizes the written ionic state after bias removal, increasing memory retention by 69.7% relative to a conventional hydrogel and, unusually, making retention more persistent as temperature rises. This coupling of thermally regulated ion transport and post-stimulus state retention allows prior thermal exposure to be stored within the ionic material and translated into history-dependent electronic responses. As a proof of concept, the retained state

regulates robotic recontact, suppressing cumulative heating and thermal drift in a co-located pressure sensor.

Main

In biological systems, protection from thermal stress relies not only on instantaneous temperature sensing but also on neural processes that encode prior exposure, retain thermal-history and use this internal state to regulate subsequent behavior (**Fig. 1a**)¹⁻⁵. Thermosensitive ion channels transduce thermal cues into ionic fluxes and changes in membrane potential, which are integrated across neural circuits to generate protective responses. Repeated or prolonged exposure can further shift response thresholds through activity-dependent synaptic plasticity, allowing previous thermal experience to influence later behavior. Reproducing such history-dependent thermal processing in artificial systems remains challenging. Conventional architecture distributes sensing⁶⁻¹⁰, memory¹¹⁻¹⁹, and control²⁰⁻²² across separate components, requiring repeated signal conversion and interconnects that add latency, energy consumption, and system complexity. More importantly, these architectures rarely retain a local record of prior thermal exposure at the sensing node. This limitation is particularly relevant to densely integrated edge devices, including robots and wearable sensor nodes, in which repeated thermal exposure can cause local heat accumulation and drift in co-located sensors and circuits. Such drift can shift sensor baselines and device operating points, degrading multimodal signal fidelity even when direct electrical crosstalk is minimized. Reliable operation therefore requires not only continuous temperature sensing but also local retention of prior thermal exposure to determine whether sufficient thermal recovery has occurred before subsequent interaction.

Here we report a monolithic thermosynapse in which thermal input programs an internal ionic-state that governs both the immediate channel response and its subsequent evolution. We define a thermosynapse as a monolithic organic electronic device in which thermal input directly regulates synaptic-state writing and retention through the evolving ionic state of the electrolyte. Unlike modular architecture, the device integrates thermal sensing and memory formation within a single ionic-electronic element. Its operation relies on a zwitterionic gel electrolyte exhibiting an upper critical solution temperature (UCST) transition. Below the transition, electrostatic associations within the zwitterionic network confine ions and limit the population of field-

accessible charge carriers. As the temperature approaches the UCST, cooperative network reorganization releases confined ions, increasing ionic availability for electrochemical modulation of organic electrochemical transistors (OECTs) channel²³ and thereby amplifying the immediate response. Under gate bias, the reorganized network also develops field-induced polarization. After bias removal, a persistent polarized ionic-state impedes ion back-diffusion and prolongs retention. Thermal input therefore regulates both the writing and relaxation of the same internal ionic-state, coupling instantaneous sensing to post-stimulus memory within a single device.

By integrating the thermosynapse into a self-adaptive robotic platform (**Fig. 1b**), we show that this retained state can directly govern the timing of subsequent physical interaction. The device state determines both withdrawal from a heated object and the waiting time before recontacting, allowing the robotic hand to regrasp only after sufficient thermal recovery rather than after a fixed externally programmed delay. This state-dependent waiting suppresses cumulative heating during repeated interactions. In a co-located pressure-sensing demonstration, the same thermal-history-dependent operation stabilizes the pressure-sensor baseline and preserves signal fidelity that otherwise drifts as heat accumulates. By contrast, conventional OECT-based controls lacking persistent temperature-dependent memory undergo rapid state relaxation and premature recontact, leading to repeated heating and sensor drift. The monolithic thermosynapse thus extends instantaneous thermal sensing into a thermal-history-dependent sensing-memory-action loop and provides a device-level route towards adaptive edge electronics that use prior thermal exposure to regulate subsequent sensing and control.

Ionic-state programming for monolithic thermal sensing and memory

To achieve temperature-dependent control over ionic dynamics, we designed a thermoresponsive zwitterionic gel comprising an interpenetrating network of poly[2-(methacryloyloxy)ethyl]-dimethyl-(3-sulfopropyl)ammonium hydroxide (PDMAAPS) and poly(ethylene glycol) diacrylate (PEGDA), containing lithium bis(trifluoromethanesulfonyl)imide (LiTFSI) (Supplementary Note 1 and Supplementary Fig. 1). The PEGDA network provides mechanical integrity while preserving the thermoresponsive behavior of PDMAAPS and suppressing concentration gradients associated with polymer aggregation, thereby enabling stable operation. PDMAAPS exhibits the UCST behavior in aqueous electrolytes, adopting a compact,

associated state at low temperatures owing to strengthened intra- and interchain interactions between its zwitterionic side groups^{24,25}. In this state, LiTFSI is preferentially associated with zwitterion-rich polymer domains, reducing the effective population of ions accessible to an applied electric field (**Fig. 1c**). Upon heating, these domains progressively dissociate as thermal energy weakens the intermolecular associations and increases polymer-segment mobility. This structural reorganization transfers previously confined ions into a more transport-accessible environment, thereby increasing the effective mobile-ion population.

As illustrated in **Fig. 1c-(i)**, this temperature-dependent ion availability regulates the interfacial ionic-state under an applied electric field. Below the UCST, the limited population of field-accessible ions restricts Li⁺ accumulation at the electrolyte-semiconductor interface, resulting in weak channel-current modulation. As the temperature approaches and exceeds the UCST, progressive disruption of the associated zwitterionic domains increases the mobile ion concentration, n_0 , thereby promoting interfacial ion accumulation and shortening the Debye length ($\lambda_D \propto n_0^{-1/2}$)^{26,27}. The resulting enhancement of electrical double layer (EDL) formation strengthens channel modulation²³. By contrast, conventional hydrogels primarily exhibit thermally regulated increases in ion mobility without a comparable change in the population of field-accessible ions and therefore show substantially weaker thermal amplification.

Figure 1c-(ii) illustrates the complementary mechanism underlying ionic-state retention. At elevated temperatures, the reorganized zwitterionic network becomes more susceptible to field-induced dipolar alignment. Following removal of the gate bias, a fraction of this field-induced polarization persists, generating a residual interfacial electrostatic environment that impedes ion back-diffusion and delays relaxation of the bias-written ionic-state²⁸⁻³⁰. In contrast, conventional hydrogels lack a comparably polarizable zwitterionic network and consequently exhibit weaker state retention and faster relaxation at elevated temperatures. This thermoresponsive zwitterionic gel thus couples thermally activated ion release during the formation of a gate-bias-induced ionic-state with post-stimulation ionic-state retention, integrating thermal sensing and memory within a single electrolyte. Consistent with this mechanism, the device response reflects temperature-dependent ion injection into the PEDOT:PSS channel (**Fig. 1d**). At low temperatures, limited ion availability results in weak volumetric dedoping and correspondingly small channel-current

modulation. Near and above the UCST, the increased population of mobile ions promotes ion injections and amplifies cumulative channel modulation. In the conventional OECT, by contrast, heating predominantly increases ion mobility without substantially altering the pool of field-accessible ions, producing a markedly smaller thermal enhancement of the channel response.

UCST-mediated ion confinement and release in a thermoresponsive zwitterionic gel

To position the UCST transition within the target operating window, we systematically varied the gel composition, using the LiTFSI concentration as the principal control parameter (**Fig. 2a**). The UCST behavior of PDMAPS is governed by intra- and interchain associations among its zwitterionic side groups. At low salt concentrations, these electrostatic associations stabilize polymer-rich domains that sequester mobile ions. Increasing the LiTFSI concentration screens the zwitterion-zwitterion interactions and modifies the local ionic-coordination environment through coordination between LiTFSI ions and the polymer network, thereby weakening polymer association and shifting the UCST to lower temperatures. LiTFSI therefore serves not only as an ionic charge carrier but also as a regulator of the ion-confining network. Through this compositional tuning, the transition window was positioned within the 37~50 °C range relevant to low-grade heating range of densely integrated edge devices.³¹ At 0.05 mol% LiTFSI, the UCST transition closely overlapped with this target range. Temperature-dependent transmittance measurements supported the associated-to-dissociated transition, reduced transmittance at lower temperatures was consistent with light scattering from associated polymer domains, whereas heating progressively restored transmittance as these domains dissociated. In contrast, at high salt concentration (10 mol% LiTFSI), which exhibits no UCST behavior within this temperature range, contained few associated domains even at the lowest temperature examined and therefore showed no appreciable change in transmittance with temperature (**Fig. 2b** and Supplementary Fig. 2). Together, these results demonstrate that LiTFSI concentration controls the association-dissociation equilibrium of the zwitterionic network, thereby defining the temperature window for reversible ion confinement and release.

To isolate the contribution of the UCST transition to ion dynamics, we compared two gel compositions. The low-salt gel containing 0.05 mol% LiTFSI, hereafter termed the transition-

active gel, undergoes a UCST transition within the low-grade heating regime. In contrast, the high-salt gel containing 10 mol% LiTFSI, hereafter termed the out-of-transition gel, exhibits a UCST shifted below 0 °C and therefore remains in the dissociated state throughout the low-grade heating regime. The transition-active gel showed a relative increase in ionic conductivity approximately 922% at 47.5 °C, whereas the out-of-transition gel increased by only ~118% at 50 °C (**Fig. 2c** and Supplementary Fig. 3). This pronounced contrast indicates that the transport enhancement originates primarily from UCST-driven network reorganization rather than from conventional thermally activated ion mobility alone (**Fig. 2d**). A corresponding transition-dependent increase was also observed in the EDL capacitance (Supplementary Fig. 4), consistent with an increase in the effective population of field-accessible ions and their interfacial charge storage capacity. To quantify the energetics of ion transport, we analyzed the temperature dependence of ionic conductivity using the Arrhenius relation ^{32,33}:

$$\ln \sigma = \ln \sigma_0 - \frac{E_a}{R} \frac{1}{T}$$

where σ_0 is the pre-exponential factor, E_a is the apparent activation energy for ion transport, R is the gas constant, and T is the absolute temperature. In the out-of-transition gel, E_a remained nearly constant at approximately 5.2 kJ mol⁻¹, indicating that ionic conduction was governed predominantly by conventional thermal activation. By contrast, the transition-active gel exhibited three distinct transport regimes: a low-temperature associated state with a relatively high E_a of approximately 18.1 kJ mol⁻¹, an intermediate transition regime characterized by a rapidly changing transport response, and a high-temperature dissociated state with a substantially lower E_a of approximately 6.9 kJ mol⁻¹ (**Fig. 2e**). This marked reduction in the transport barrier indicates that ion motion becomes increasingly favorable as the associated zwitterionic domains are disrupted across the UCST transition.

Because the conductivity-derived activation energy reflects ion-transport energetics, we next used rheological analysis to probe the polymer network relaxation underlying this transition. Time-temperature superposition analysis showed that both compositions remained gel-like throughout the measurement range, with $G' > G''$ (Supplementary Fig. 5). However, the UCST gel exhibited a pronounced decrease in the segmental-relaxation activation energy, $E_{a,seg}$, from approximately 260.1 to 30.3 kJ mol⁻¹ across the transition (**Fig. 2f** and Supplementary Fig. 6). The out-of-

transition gel showed no comparable change over the same temperature range (Supplementary Fig. 7). These results indicate that the sharp increase in ionic transport is accompanied by substantial reorganization and accelerated relaxation of the zwitterionic polymer network. Temperature-dependent Fourier-transform infrared (FT-IR) spectroscopy further resolved this structural evolution at the molecular level. Near the UCST regime, the sulfonate stretching band in the zwitterionic side chain shifted from 1170.2 to 1168.3 cm^{-1} , consistent with weakened interactions between the side chains of PDMAPS upon heating. Above the transition, the band position changed only slightly, indicating that the network reorganization had largely reached saturation in the dissociated state. Collectively, the transport, rheological, and spectroscopic results establish that UCST-driven reorganization of the zwitterionic network governs reversible ion confinement and release (**Fig. 2g**).

Thermally amplified sensing and synaptic modulation in a monolithic thermosynapse

To translate UCST-regulated ionic-state reorganization into an electronic response, we used the transition-active gel, functioning as a thermoresponsive zwitterionic gel, as the gate electrolyte in a PEDOT:PSS-based OECT, thereby forming a monolithic thermosynapse. Cyclic voltammetry (CV) of the gel showed no pronounced redox features within the operating-voltage window, suggesting minimal parasitic Faradaic reactions at the electrolyte-electrode interface (Supplementary Fig. 8)²⁷. Voltage-dependent ultraviolet-visible (UV-vis) spectroscopy and electrochemical impedance spectroscopy (EIS) of a gel/PEDOT:PSS capacitor revealed the onset of PEDOT:PSS dedoping above approximately 1.5 V and an accompanying increase in capacitance consistent with ion penetration into the bulk channel (Supplementary Figs. 9 and 10). Notably, the effective volumetric capacitance increased markedly between 30 and 47.5 °C, mirroring the temperature-dependent conductivity transition and indicating enhanced ionic access to the PEDOT:PSS channel upon reorganization of the zwitterionic network (Supplementary Fig. 11). At room temperature, the device exhibited stable output and transfer characteristics, together with pulse-number-, amplitude- and duration-dependent plasticity, providing the baseline for investigating thermally regulated synaptic responses (Supplementary Note 2).

To prove the role of the UCST-responsiveness, we compared our OECT device with a conventional OECT gated by a PEO-based hydrogel lacking ion-polarizable units and UCST behavior (Supplementary Fig. 12). With increasing temperature, the transfer curves of the thermoresponsive device shifted towards lower gate voltages while retaining their overall form, indicating more efficient channel dedoping without a change in the operating mode (Supplementary Fig. 13). We therefore selected $V_G = 1.5$ V, at which the temperature-dependent current responses were most clearly resolved. A single gate pulse generated an inhibitory postsynaptic current (IPSC), the amplitude of which increased progressively with temperature (**Fig. 3a**). Using the initial current at 25 °C as a reference, we defined the normalized response as $\Delta I/I_{25^\circ\text{C}}$. Between 30~47.5 °C, both devices exhibited increased current modulation, but the enhancement was substantially greater in the monolithic thermosynapse. This contrast shows that the UCST transition amplifies thermal sensing beyond the conventional temperature dependence of ion mobility alone.

The influence of the phase transition became more pronounced under consecutive gate-pulse stimulation. In the monolithic thermosynapse, the IPSC accumulated with pulse number, and the separation between responses at different temperatures progressively increased (**Fig. 3b**). Elevated temperature therefore enhanced both the response to an individual pulse and the cumulative modulation produced by repeated stimulation. By contrast, the conventional OECT showed substantially weaker temperature dependence under the same conditions (**Fig. 3c**). These results indicate that the amplified response arises predominantly from UCST-regulated changes in the accessible ionic population rather than solely from thermally accelerated ion transport. Because the channel conductance reflects the electrochemical doping state of PEDOT:PSS^{23,34,35}, we next examined how temperature affects the gate-induced conductance change. Consecutive positive gate pulses progressively decreased the conductance, corresponding to long-term depression (LTD), whereas subsequent reverse pulses restored the conductance through long-term potentiation (LTP) (**Fig. 3d** and Supplementary Fig. 14). The amplitudes of both LTD and LTP increased with temperature and reached their largest values at 50 °C, thereby expanding the accessible conductance window. Consistent with this trend, the dynamic range $G_{\text{max}}/G_{\text{min}}$, defined as the ratio between the maximum and minimum conductance states, increased monotonically with

temperature (**Fig. 3e**). The UCST-regulated ionic-state therefore determines not only the instantaneous thermal response but also the range over which synaptic weights can be updated.

Finally, the device maintained reversible conductance modulation over 900 alternating LTD and LTP pulses at 40 °C (**Fig. 3f**), with comparable cycling behavior observed at 35 and 50 °C (Supplementary Fig. 15). The conductance swing increased at elevated temperatures without compromising the reversibility of the updates. Collectively, these results show that UCST-driven network reorganization expands the accessible ionic and electronic state space of the OECT, coupling thermally amplified sensing to robust and tunable synaptic modulation.

Thermal-history encoding through retained ionic-states

A defining requirement of a thermosynapse is not only to detect an instantaneous thermal stimulus but also to retain a readable state that reflects prior exposure. We therefore examined whether temperature could regulate both the writing and post-stimulus retention of the electrolyte ionic-state. As illustrated in **Fig. 4a**, heating weakens local associations within the zwitterionic network and increases the orientational susceptibility of its dipolar side groups under an applied electric field³⁶⁻³⁸. Following removal of the gate bias, a fraction of the field-induced polarization may persist, creating a residual interfacial electrostatic environment that impedes ion back-diffusion from the PEDOT:PSS channel and stabilizes the written ionic-state^{39,40}. Polarization-electric field (P-E) measurements revealed increasingly open hysteresis loops at lower LiTFSI concentrations, suggesting that the ionic composition of the gel influences polarization retention (Supplementary Fig. 16)⁴¹. Moreover, both the loop opening and remanent component increased with temperature in the zwitterionic gel (**Fig. 4b**), whereas the PEO-based hydrogel exhibited only weak hysteresis over the same temperature range (**Fig. 4c**). Because P-E hysteresis in ionic soft materials can contain both dipolar and mobile ion contributions, these results do not uniquely identify the microscopic origin of the polarization but are consistent with the formation of a more persistent polarized ionic-state at elevated temperatures.

We next investigated how this thermally regulated polarized state affects ionic-state retention in the OECT. During gate-pulse stimulation, both the monolithic thermosynapse and conventional OECTs exhibited larger current modulation as the temperature increased (**Fig. 4d** and Supplementary Fig. 17). Their responses diverged markedly, however, after removal of the gate

bias. In the thermoresponsive device, the residual current and retention time progressively increased with temperature, with a 69.7% increase in the retained memory level and a pronounced enhancement emerging above approximately 30 °C, near the onset of the UCST transition. By contrast, the conventional OECT exhibited thermally accelerated relaxation, with the written state decaying more rapidly at higher temperatures (**Fig. 4d** and Supplementary Fig. 18). This inversion of the conventional temperature-retention relationship suggests that the persistent interfacial polarization generated in the zwitterionic gel counteracts thermally accelerated ion back-diffusion and thereby stabilizes the bias-written ionic-state.

To distinguish temperature-dependent state writing from post-stimulus retention, we implemented two complementary temperature-modulation protocols. In the first protocol, temperature was varied only during gate-pulse stimulation, whereas the subsequent retention condition was held constant (**Fig. 4e**). Increasing the writing temperature enhanced both the stimulation current and the residual current after bias removal, indicating that UCST-regulated ion availability enables a larger ionic-state to be written into the PEDOT:PSS channel. In the second protocol, the writing temperature was held constant, and temperature was varied only during the field-off retention period (**Fig. 4f**). The initially written states were therefore nearly identical, but their subsequent relaxation depended strongly on the retention temperature. The residual current decayed more rapidly at lower temperatures, whereas elevated temperature prolonged the bias-written state.

Under both protocols, the conventional OECT exhibited the opposite temperature dependence, with elevated temperatures accelerating relaxation and reducing the residual current (Supplementary Fig. 19). Together, these results show that the zwitterionic gel couples temperature-dependent ionic-state writing with enhanced field-off retention. Thermal history is therefore encoded not simply in the magnitude of an instantaneous response, but in the subsequent evolution of the material's internal ionic-state, allowing prior thermal exposure to shape later device outputs. Having established the distinctive temperature-enhanced memory retention of our monolithic thermosynapse, we next quantified its retention time at different temperatures under a low operating voltage of 1 V and benchmarked the results against previously reported conventional synapses (**Fig. 4g**, Supplementary Fig. 20 and Supplementary Table 1)⁴²⁻⁴⁶. The retention time increased monotonically with temperature, reaching approximately 1,800 s at 55 °C and

substantially exceeding those of conventional monolithic and integrated synapses. Notably, this positive temperature dependence contrasts with the behavior of conventional synapses, which generally exhibit shorter retention and accelerated memory decay at elevated temperatures. By allowing the duration of the retained state to increase with the level of thermal hazard, this temperature-retention relationship provides a critical basis for adaptive thermal protection. Additionally, the temperature-dependent IPSC responses were highly reproducible across six repeated measurements at each temperature from 20 to 60 °C (Supplementary Fig. 21), and an artificial neural network trained on these pre- and post-synaptic signals classified the eight temperature states with an accuracy of 97.2% when both signals were combined, exceeding the 91.7% and 94.4% obtained from pre- or post-synaptic signals alone (Supplementary Fig. 22). These results confirm that the thermosynapse encodes temperature into reliable, well-separated states suitable for closed-loop operation.

Thermal-history-dependent system integrity under repeated heating

Having established temperature-dependent ionic-state writing and retention, we next examined whether the stored state could regulate physical action in a manner that preserves the integrity of co-located electronics. Densely integrated edge systems, including robots and wearable devices, can experience repeated local heating in the 37~55 °C range, which can accumulate when heat dissipation is insufficient and perturb neighboring sensor signals. We therefore integrated the monolithic thermosynapse with a robotic hand through a differential amplifier and an Arduino-based motor-positioning system, in which thermal exposure writes an internal ionic-state and its subsequent evolution controls the servomotors through predefined current thresholds (**Fig. 5a,b**). The release threshold, I_{th1} , was defined from the current reached after 6 s of exposure to a 55 °C solid heat source. Crossing I_{th1} triggered release of the object, whereas subsequent relaxation of the written state determined when recontact was permitted. Thus, the waiting interval was not imposed as a fixed delay but emerged from the relaxation dynamics of the thermosynapse itself. A porcine-skin layer was placed at the contact interface as a tissue surrogate for monitoring cumulative surface heating (**Fig. 5c**). Unlike modular thermal-reflex architectures in which sensing and memory are implemented in separate components, the present device senses thermal exposure and retains its history through the evolving ionic-state of the same OECT, allowing prior thermal exposure to regulate the timing of subsequent physical interaction.

We first evaluated whether the thermosynapse could distinguish benign from elevated thermal conditions using a common actuation threshold. In the monolithic thermosynapse, ion confinement at low temperature suppresses cumulative channel modulation, whereas heating across the UCST transition increases the population of field-accessible ions and strongly amplifies the inhibitory postsynaptic current (IPSC). Consequently, the state trajectories at 25 and 55 °C diverged markedly under repeated stimulation. The conventional OECT-based system exhibited substantially weaker temperature discrimination because mobile ions remained readily accessible even at low temperatures. Consistent with this difference, the mean temperature-dependent paired-pulse facilitation (PPF) index increased by 239% in the monolithic thermosynapse, compared with 29% in the conventional OECT-based system (Supplementary Fig. 23), producing a substantially wider temperature-dependent threshold window (**Fig. 5d**). This separation translated directly into temperature-selective actuation (**Fig. 5e**, Supplementary Fig. 24 and Supplementary Videos 1 and 2). At 25 °C, the IPSC of the monolithic thermosynapse remained below the release threshold and the robotic hand maintained its grip, whereas at 55 °C the enhanced ionic response drove the IPSC across the same threshold and triggered release (**Fig. 5e(i)**). By contrast, the conventional OECT-based system underwent progressive channel modulation even at 25 °C and eventually crossed the threshold under both conditions, resulting in loss of temperature-selective actuation (**Fig. 5e(ii)**). UCST-regulated ion confinement and release therefore translate temperature into distinct state trajectories rather than merely accelerating an otherwise similar ionic response.

We next asked whether retention of the written state could regulate the timing of repeated interactions and thereby preserve the fidelity of a neighboring sensor. A capacitive pressure sensor was co-located with the thermosynapse at the contact interface, and the IPSC, porcine-skin temperature and pressure-sensor response were monitored simultaneously during repeated grab-release-regrasp cycles with a heated object (**Fig. 5f** and Supplementary Video 3). In the conventional OECT-based system, the written state relaxed rapidly after release, causing the control signal to return to the recontact condition before the contact interface had sufficiently cooled. Repeated premature recontact led to progressive heat accumulation. This accumulated heat also perturbed the co-located pressure sensor: under an identical applied pressure of 10 kPa, its capacitive response exhibited a progressively shifted baseline across successive cycles. Thus, the

same mechanical input generated different electrical outputs as the local thermal state evolved, directly demonstrating loss of pressure-sensing fidelity induced by cumulative heating.

The monolithic thermosynapse exhibited a markedly different temporal response. The persistent polarized ionic-state after bias removal slowed relaxation of the written state and prolonged the interval before recontacting. Because complete relaxation to the initial current level ($\Delta I = 0$) would require an impractically long waiting time, we defined an operational recontact criterion at 18% of the retained memory level for the robotic demonstration. This criterion preserved the intrinsic dependence of recontact timing on the relaxation dynamics of the device while providing a practical operating window. As the thermal exposure increased, the written ionic-state became more persistent, consistent with the temperature-dependent retention observed in **Fig. 4** and Supplementary Fig. 25. The resulting waiting interval therefore increased with thermal exposure without requiring a separately programmed timer. This memory-governed delay allowed the contact temperature to recover towards its initial value before regripping, limiting cumulative heating and maintaining a stable pressure-sensor response over repeated cycles. Infrared thermography further visualized the contrasting thermal trajectories (**Fig. 5f(i,ii)** and Supplementary Fig. 26): the conventional OECT-based system exhibited progressive heat accumulation, whereas the monolithic thermosynapse allowed the contact interface to recover close to its initial thermal state between successive interactions. Crucially, the timing of recontact was determined not by a separate memory element or a fixed delay but by relaxation of the temperature-programmed ionic-state within the thermosynapse. The device therefore converts prior thermal exposure into an adaptive waiting interval that regulates subsequent interaction while preserving the fidelity of co-located sensing, establishing a thermal-history-dependent sensing-memory-action loop within an integrated electronic system.

Outlook

In conclusion, we establish a bio-inspired monolithic thermosynapse in which thermal input directly programs the evolution and retention of ionic-states within a single organic electrochemical transistor. In the UCST-type zwitterionic electrolyte, temperature-dependent network reorganization regulates ion confinement and release, thereby controlling the population of mobile ions available for channel modulation. Under gate bias, field-induced polarization

contributes to the formation of a persistent polarized ionic-state after bias removal, suppressing ion back-diffusion and stabilizing the written state. Together, these coupled processes integrate immediate thermal sensing with history-dependent memory within a single ionic-electronic device. In contrast to a conventional OECT, in which elevated temperature accelerates ionic relaxation, the monolithic thermosynapse retains information about prior thermal exposure and uses this internal state to regulate subsequent device responses.

As a proof of concept, integration with a robotic platform allowed the retained ionic state to determine the waiting interval before renewed contact, thereby providing sufficient time for thermal recovery and suppressing cumulative heating at the contact interface. This state-dependent recovery also preserved the fidelity of a co-located pressure sensor by limiting heat-induced baseline drift under repeated operation. The thermosynapse therefore converts prior thermal exposure into a persistent internal state that regulates both subsequent physical interaction and the reliability of neighboring sensing functions. More broadly, our results establish temperature-gated ionic-state programming as a device strategy for embedding thermal-history directly into adaptive electronics. By linking ionic-state evolution, memory retention and physical action within a monolithic device, this work provides a route towards electronic systems that respond not only to instantaneous temperature but also to the history of prior thermal exposure.

Methods

Materials

[2-(Methacryloyloxy)ethyl]dimethyl-(3-sulfopropyl) ammonium hydroxide (DMAPS), 2,2'-azobis(isobutyronitrile) (AIBN), poly(ethylene glycol) diacrylate (PEGDA), 2-hydroxy-2-methylpropiophenone (HMPP), 2,2,2-trifluoroethanol (TFE), and lithium bis(trifluoromethanesulfonyl)imide (LiTFSI) was purchased from Sigma-Aldrich. PEDOT:PSS (Clevios™ PH 1000) was purchased from Ossila.

Synthesis of PDMAPS for zwitterionic gel

DMAPS (17 g, 0.061 mol), AIBN (0.001 g, 0.0061 mmol), and TFE (85 mL) were used for polymerization (at 70 °C, 24 h). The polymer was precipitated three times in methanol. Number

average molecular weight (M_n) and polydispersity (D) of the obtained PDMAPS were measured using size exclusion chromatography (SEC, LC-4500, JASCO) with a refractive index detector (RI-4030, JASCO) calibrated with standard poly (ethylene oxide) (calibration kit, Scientific Polymer Products, Inc.). The obtained PDMAPS had an M_n of 126,700 g mol⁻¹ and D of 1.44 (Fig. S1).

Fabrication of zwitterionic gel

PDMAPS (0.5g), PEGDA ($M_n \approx 700$ g/mol) (0.05g), HMPP (1 mg) were dissolved in 0.05~10 mol% LiTFSI aqueous solution (0.75 mL) and stirred for 2 h at 60 °C. This solution was injected into the VHB tape template on the glass or gold electrodes. Subsequently, UV irradiation was performed for 300 s at 60 °C. Finally, the opaque thermoresponsive zwitterionic gel films were obtained. The control hydrogel without UCST behavior was also fabricated using a similar procedure.

Characterizations of zwitterionic gel

Ionic conductivities of zwitterionic gel and control hydrogel were measured using electrochemical impedance spectroscopy (EIS; IM6, Zahner) at an amplitude of 10 mV and in the frequency region from 10⁶ to 10⁻¹ Hz. The temperature was controlled by a constant-temperature chamber (J-RHC3-LCD-T, JISCO). The dynamic mechanical behaviors were characterized by an oscillatory rheometer (MCR-92, Anton Paar) with 8 mm parallel plates. The sample gap was fixed at ~1 mm and the strain was set to a small strain amplitude of 1% for frequency-dependent measurements. To examine polymer dynamics in the zwitterionic and control gels, master curves were constructed using the time-temperature superposition principle, derived from isothermal frequency sweep measurements performed over a temperature range of 0 to 70 °C, with 20 °C as the reference temperature. Attenuated total reflection Fourier-transform infrared (ATR-FTIR) spectra were recorded in absorption mode using a Bruker Optics GmbH spectrometer equipped with a ZnSe crystal. Temperature-dependent ATR-FTIR measurements were performed over the temperature range of 25~60 °C to monitor thermally induced structural changes in the zwitterionic gel.

Fabrication and characterization of organic electrochemical transistors

Highly doped n-type Si/SiO₂ wafers with a 3000 Å SiO₂ layer were sequentially cleaned with acetone, isopropyl alcohol and deionized water by ultrasonication, followed by drying with N₂ gas.

Source and drain electrodes were formed by thermal evaporation of Cr/Au (3/47 nm) through a shadow mask under high vacuum. PEDOT:PSS (PH1000) was used as the organic semiconductor channel. The PEDOT:PSS solution was filtered through a PVDF membrane, spin-coated at 1000 rpm for 60 s and annealed at 100 °C for 60 min. The channel geometry was defined by the shadow-mask design, with a channel length and width of 100 μm and 6000 μm , respectively. A VHB tape frame was then placed around the PEDOT:PSS channel region, and the zwitterionic gel precursor solution was introduced into the frame. The zwitterionic gel-based electrolyte was formed by ultraviolet irradiation. Finally, an Au-coated electrode was placed on top of the VHB frame as the gate electrode and to encapsulate the hydrogel, suppressing rapid water evaporation from the electrolyte. For comparison, conventional OECTs were fabricated using a PEO-based hydrogel precursor solution under the same device geometry and measurement conditions. The output and transfer characteristics of the OECTs were measured under ambient conditions using a semiconductor parameter analyzer (Keithley 4200). For temperature-dependent measurements, the device temperature was controlled using a heating stage and monitored with an infrared thermal camera. Gate-pulse stimulation measurements were performed by applying voltage pulses to the gate electrode while recording the drain current at a fixed drain bias. The inhibitory postsynaptic current (IPSC), conductance modulation, and memory level were extracted from the drain-current responses. UV-vis-NIR absorption spectra of the electrode/ zwitterionic gel /PEDOT:PSS/electrode samples were recorded using a UV-vis-NIR spectrophotometer (V-770, JASCO) over a wavelength range of 400~2400 nm, with a spectral resolution of 0.5 nm and a scan rate of 1000 nm min⁻¹. EIS measurements of the electrode/ zwitterionic gel /PEDOT:PSS/electrode samples were performed using a PGSTAT302N electrochemical workstation (Metrohm Autolab) over a frequency range of 0.1 Hz to 1 MHz with an AC amplitude of 10 mV.

ANN-based temperature classification

To evaluate the information encoded in the temperature-dependent signals, the measured pre-synaptic and post-synaptic responses were preprocessed and used as inputs to an artificial neural network (ANN). The input data were configured in three ways: (i) pre-synaptic signals only, (ii) post-synaptic signals only, and (iii) combined pre- and post-synaptic signals. Each sample was assigned to one of eight temperature classes ranging from 20 to 60 °C. Classification performance

for each input configuration was evaluated using confusion matrices and the corresponding overall classification accuracy.

Design of the closed-loop robotic system with a monolithic thermosynapse

The robotic control system was constructed by integrating the monolithic thermosynapse with an Arduino-based motor-positioning system. Because the servomotors of the robotic hand were voltage-controlled, the drain-current output of the thermosynapse was first converted into a voltage signal and amplified before being transmitted to the controller. The control circuit comprised a current-to-voltage conversion resistor, a differential/inverting amplifier, and an Arduino-based motor-positioning unit (**Fig. 5a**). The drain current generated by the thermosynapse was converted into a voltage across the load resistor, amplified by the differential/inverting amplifier and supplied to the A0 analogue-input pin of the Arduino. The Arduino subsequently controlled the servomotors of the robotic hand through its digital output pins. The system operated through two sequential processes, thermal sensing and memory formation, followed by memory-guided robotic actuation. During thermal sensing and memory formation, thermal stimulation modulated the drain current of the thermosynapse according to its temperature-dependent ionic-state dynamics. The resulting current signal was converted and amplified to generate the input voltage for the motor-positioning system. During memory-guided robotic actuation, the Arduino compared the input voltage with two predefined threshold current, I_{th1} and I_{th2} . When the input voltage reached I_{th1} , high-level output signals were generated at the D5 and D6 pins, actuating the servomotors to release the object held by the robotic hand. As the retained device state subsequently relaxed and the input voltage reached I_{th2} , the servomotors were actuated again to regrasp the object. Because complete relaxation to the initial current level required an impractically long waiting time, the operational recontact threshold I_{th2} was defined as the voltage corresponding to 18% of the retained memory level. The interval between I_{th1} and I_{th2} was used to evaluate memory-guided protective operation. This interval represents the period for which the robotic hand remained in the released state following thermal stimulation and was determined by relaxation of the retained thermosynaptic signal. A shorter interval therefore corresponded to faster state relaxation and earlier recontact, whereas a longer interval reflected more persistent memory and provided additional time for thermal recovery before regrasping.

Data availability

All relevant data that supports the results of this study are available within the article and its supplementary information files. Further data is available from the corresponding authors upon request.

References

1. Vriens, J., Nilius, B. & Voets, T. Peripheral thermosensation in mammals. *Nat. Rev. Neurosci.* **15**, 573–589 (2014).
2. Jung, S. et al. A forebrain neural substrate for behavioral thermoregulation. *Neuron* **110**, 266–279 (2022).
3. Yarmolinsky, D. A. et al. Coding and plasticity in the mammalian thermosensory system. *Neuron* **92**, 1079–1092 (2016).
4. Ambroziak, W. et al. Thermally induced neuronal plasticity in the hypothalamus mediates heat tolerance. *Nat. Neurosci.* **28**, 346–360 (2025).
5. Jouandet, G. C. et al. Rapid threat assessment in the drosophila thermosensory system. *Nat. Commun.* **14**, 7067 (2023).
6. Sharma, S. et al. A miniaturized ingestible temperature sensor for continuous internal monitoring. *Nat. Electron.* **9**, 956–968 (2026).
7. Zhang, X. et al. Flexible temperature sensor with high reproducibility and wireless closed-loop system for decoupled multimodal health monitoring and personalized thermoregulation. *Adv. Mater.* **36**, 2407859 (2024).
8. Shin, J. et al. Sensitive wearable temperature sensor with seamless monolithic integration. *Adv. Mater.* **32**, 1905527 (2020).
9. Baines, R. et al. Multi-modal deformation and temperature sensing for context-sensitive machines. *Nat. Commun.* **14**, 7499 (2023).
10. Yang, L. et al. Thermoelectric porous laser-induced graphene-based strain-temperature decoupling and self-powered sensing. *Nat. Commun.* **16**, 792 (2025).
11. Zhu, C. et al. Stretchable temperature-sensing circuits with strain suppression based on carbon nanotube transistors. *Nat. Electron.* **1**, 183–190 (2018).

- 526 12. Kim, J. S. et al. Intrinsically stretchable subthreshold organic transistors for highly
527 sensitive low-power skin-like active-matrix temperature sensors. *Adv. Funct. Mater.* **34**,
528 2305252 (2024).
- 529 13. Boahen, E. K., Yoo, H. & Kim, D. H. Sensing on-chip temperature with two-dimensional
530 iontronic transistors. *Nat. Sens.* **1**, 204-205 (2026).
- 531 14. Sim, K. et al. An epicardial bioelectronic patch made from soft rubbery materials and
532 capable of spatiotemporal mapping of electrophysiological activity. *Nat. Electron.* **3**, 775-
533 784 (2020).
- 534 15. Zhao, C. et al. Skin-like drift-free biosensors with stretchable diode-connected organic
535 field-effect transistors. *Nat. Electron.* **8**, 981-993 (2025).
- 536 16. Huang, T. et al. Closed-loop haptic–thermal perception with memristor-based spiking
537 neurons for embodied neuromorphic intelligence. *Adv. Funct. Mater.* **36**, e23270 (2026).
- 538 17. Shi, J. et al. Adaptive processing enabled by sodium alginate based complementary
539 memristor for neuromorphic sensory system. *Adv. Mater.* **36**, 2314156 (2024)
- 540 18. Li, Z. et al. Crossmodal sensory neurons based on high-performance flexible memristors
541 for human-machine in-sensor computing system. *Nat. Commun.* **15**, 7275 (2024).
- 542 19. Kim, K. K. et al. Unisensory processing of interleaving memristive nanowires enabling
543 multimodal sensing at human-scale resolution. *Nat. Mater.* **25**, 463-471 (2026).
- 544 20. Li, J. et al. AI-embodied multi-modal flexible electronic robots with programmable sensing,
545 actuating and self-learning. *Nat. Commun.* **16**, 8818 (2025).
- 546 21. Han, M. et al. Catheter-integrated soft multilayer electronic arrays for multiplexed sensing
547 and actuation during cardiac surgery. *Nat. Biomed. Eng.* **4**, 997-1009 (2020).
- 548 22. Ji, J. et al. Multisensory electronic skin with decoupled pressure–temperature-sensing
549 capabilities for similar object recognition. *Proc. Natl. Acad. Sci. U. S. A.* **122**, e2519693122
550 (2025)
- 551 23. Rivnay, J. et al. Organic electrochemical transistors. *Nat. Rev. Mater.* **3**, 17086 (2018).
- 552 24. Lei, Z. & Wu, P. Zwitterionic skins with a wide scope of customizable functionalities. *ACS*
553 *Nano* **12**, 12860-12868 (2018).
- 554 25. Fu, W. et al. UCST-type thermosensitive hairy nanogels synthesized by RAFT
555 polymerization-induced self-assembly. *ACS Macro Lett.* **6**, 127-133 (2017).

- 556 26. Kweon, H. et al. Ion trap and release dynamics enables nonintrusive tactile augmentation
557 in monolithic sensory neuron. *Sci. Adv.* **9**, eadi3827 (2023).
- 558 27. Romele, P., Ghittorelli, M., Kovács-Vajna, Z. M. & Torricelli, F. Ion buffering and interface
559 charge enable high performance electronics with organic electrochemical transistors. *Nat.*
560 *Commun.* **10**, 3044 (2019)
- 561 28. Jeong, B., Gkoupidenis, P. & Asadi, K. Solution-processed perovskite field-effect transistor
562 artificial synapses. *Adv. Mater.* **33**, 2104034 (2021).
- 563 29. Fabiano, S. et al. Ferroelectric polarization induces electronic nonlinearity in ion-doped
564 conducting polymers. *Sci. Adv.* **3**, e1700345 (2017).
- 565 30. Lee, D. et al. Controlling long-term plasticity in neuromorphic computing through
566 modulation of ferroelectric polarization. *ACS Appl. Mater. Interfaces* **16**, 58940–58951
567 (2024).
- 568 31. Simin, Wang, et al. Data center temperature prediction and management based on a Two-
569 stage self-healing model. *Simul. Model. Pract. Theory* **132**, 102883 (2024).
- 570 32. Quartarone, E. & Mustarelli, P. Electrolytes for solid-state lithium rechargeable batteries:
571 recent advances and perspectives. *Chem. Soc. Rev.* **40**, 2525-2540 (2011).
- 572 33. Li, H. et al. A skin-mimicked polymer gel electrolyte for stabilizing lithium metal batteries.
573 *Adv. Energy Mater.* **15**, 2405365 (2025).
- 574 34. Gkoupidenis, P., Schaefer, N., Garlan, B. & Malliaras, G. G. Neuromorphic functions in
575 PEDOT:PSS organic electrochemical transistors. *Adv. Mater.* **27**, 7176–7180 (2015).
- 576 35. Bernards, D. A. & Malliaras, G. G. Steady-state and transient behavior of organic
577 electrochemical transistors. *Adv. Funct. Mater.* **17**, 3538–3544 (2007).
- 578 36. Pickett, P. D., Ma, Y. & Prabhu, V. M. Polyzwitterion fast and slow mode behavior are
579 coupled to phase separation as observed by dynamic laser light scattering. *J. Chem. Phys.*
580 **159**, 104902 (2023).
- 581 37. Mei, W. et al. Zwitterions raise the dielectric constant of soft materials. *Phys. Rev. Lett.* **127**,
582 228001 (2021).
- 583 38. Owusu, F., Tress, M., Nüesch, F. A., Lehnert, S. & Opris, D. M. Synthesis of polar
584 polynorbornenes with high dielectric relaxation strength as candidate materials for
585 dielectric applications. *Mater. Adv.* **3**, 998–1006 (2022).
- 586 39. Li, X. et al. Polymer electrets and their applications. *J. Appl. Polym. Sci.* **138**, 50406 (2021).

40. Guo, Z. et al. Organic molecular and polymeric electrets toward soft electronics. *Mol. Syst. Des. Eng.* **7**, 537–552 (2022).
41. Bhunia, R. et al. Ferroelectric-assisted ion dynamics for prolonged tactile cognizance in a biomimetic memory-in-sensor system. *Adv. Electron. Mater.* **11**, 2400550 (2025).
42. Fu, Y. M. et al. Sputtered electrolyte-gated transistor with temperature-modulated synaptic plasticity behaviors. *ACS Appl. Electron. Mater.* **4**, 2933–2942 (2022).
43. Cui, D. et al. A low-voltage stretchable synaptic transistor array for temperature perception, facilitated associative learning, and neuromorphic computing. *ACS Appl. Mater. Interfaces* **17**, 68136–68146 (2025).
44. Lu, S. et al. Thermally tunable photonic synaptic transistor with bioinspired temporal dynamics for task-adaptive neuromorphic vision. *ACS Appl. Mater. Interfaces* **18**, 32931–32942 (2026).
45. Chang, Y. et al. Robust biodegradable synapse with sub-biological energy and extended memory for intelligent reflexive system. *Nat. Commun.* **16**, 10610 (2025).
46. Wang, Y. et al. Stretchable temperature-responsive multimodal neuromorphic electronic skin with spontaneous synaptic plasticity recovery. *ACS Nano* **16**, 8283–8293 (2022).

Acknowledgements

This work was supported by the National Research Foundation of Korea (NRF) grant funded by the Korea government (MSIT) (RS-2024-00405818, RS-2025-00515479), the Pioneer Research Center Program through the National Research Foundation of Korea funded by the Ministry of Science, ICT & Future Planning (RS-2022-NR067540), and Korea Basic Science Institute (National research Facilities and Equipment Center) grant funded by the Ministry of Education (RS-2024-00436346). This research was also supported by Creative Materials Discovery Plus Program through the National Research Foundation of Korea (NRF) funded by Ministry of Science and ICT (RS-2026-25607756).

Author information

D.H.K., H.C.M. and Y.M.K. supervised the project. D.H.K., H.C.M., Y.M.K., Z.K., J.H.Kw., and H.C. conceived the concept and designed the experiments. Z.K. and J.H.Kw. conducted the main experiments and performed the material characterization. H.C., H.W.Y and H.J.S. assisted with the experimental procedures. J.Y., S.Y., Z.K., and J.H.Ki. designed and performed robotic hand experiments. Z.K., J.H.Kw., J.H.Ki. and H.C. wrote the manuscript. D.H.K., H.C.M., and Y.M.K. revised the manuscript. All authors discussed the results and commented on the manuscript.

Ethics declarations

Competing interests

The authors declare no competing interests.

Additional information

Supplementary information

Correspondence and requests for materials should be addressed to Yong Min Kim, Hong Chul Moon and Do Hwan Kim.

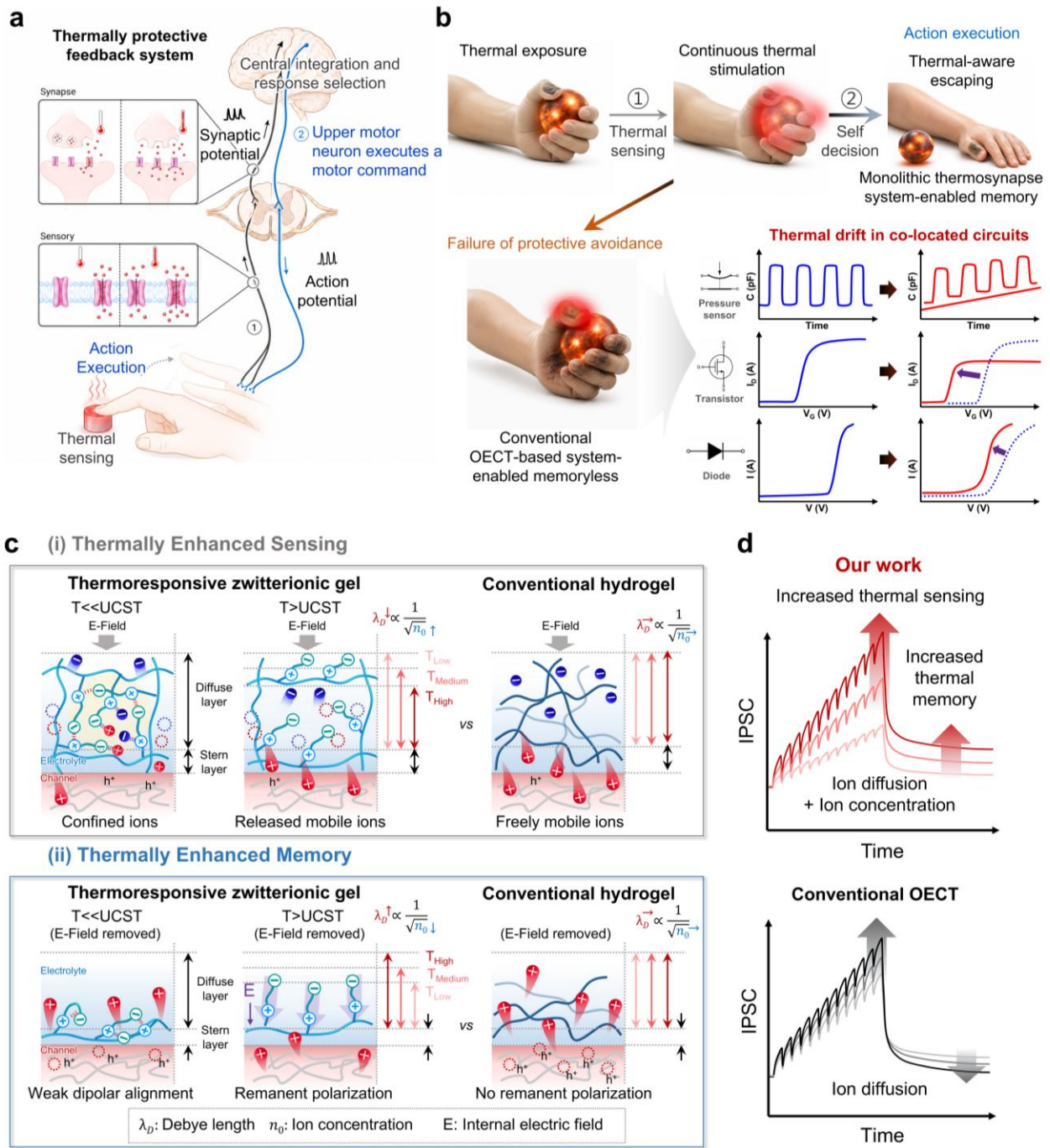


Fig. 1 | Bioinspired thermal-history encoding in a monolithic thermosynapse.
a, Biological thermoprotective feedback, in which peripheral thermal sensing, synaptic plasticity, central integration, and motor output coordinate protective responses during repeated thermal contact. **b**, Concept of a self-adaptive robotic system controlled by a monolithic thermosynapse. The retained device state triggers thermal-aware withdrawal and delays recontact, whereas a conventional OECT-based system lacking persistent thermal memory undergoes repeated contact with the heated object, leading to cumulative heating and drifting in co-located sensor signals. **c**,

Molecular mechanisms underlying thermally enhanced sensing (i) and memory (ii). Below the upper critical solution temperature (UCST), zwitterionic associations confine field-accessible ions. Heating across the transition reorganizes the network, increases the mobile-ion population and strengthens interfacial ionic modulation. After removal of the electric field, a persistent polarized ionic-state impedes ion back-diffusion, whereas the conventional hydrogel shows no comparable retained state. **d**, Schematic inhibitory postsynaptic current (IPSC) responses illustrating concurrent thermal amplification of sensing and memory in the monolithic thermosynapse, in contrast to thermally accelerated relaxation in the conventional OECT.

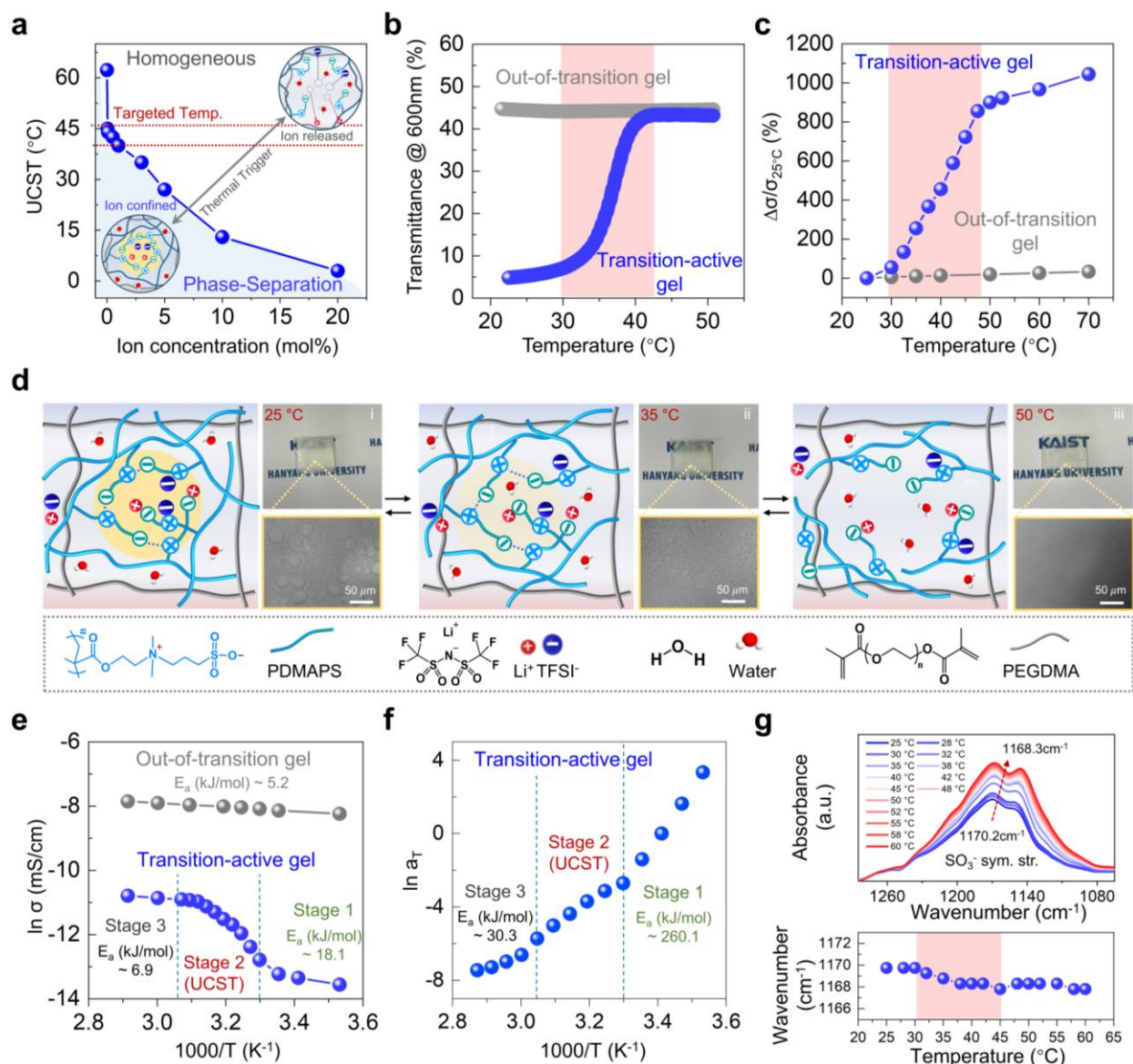


Fig. 2 | UCST-mediated ion transport and network reorganization in a zwitterionic gel. **a**, UCST as a function of LiTFSI concentration in the thermoresponsive zwitterionic gel. **b**, Optical changes in transmittance at 600 nm for transition-active and out-of-transition gels. **c**, Relative ionic conductivity as a function of temperature for transition-active and out-of-transition gels within the measured temperature range. **d**, Schematic and optical characterization of the temperature-induced transition from an associated, ion-confining state to a dissociated, ion-accessible state. **e**, Arrhenius plots of ionic conductivity for the transition-active gel and the out-of-transition gel, revealing distinct ion-transport regimes across the phase transition. **f**, Temperature dependence of the horizontal shift factor, a_T , for the transition-active gel, showing the change in polymer-network relaxation across the transition. **g**, Temperature-dependent Fourier-transform infrared spectra of the transition-active gel (top) and corresponding shift of the sulfonate stretching band (bottom), indicating weakened interactions of zwitterionic side chains upon heating.

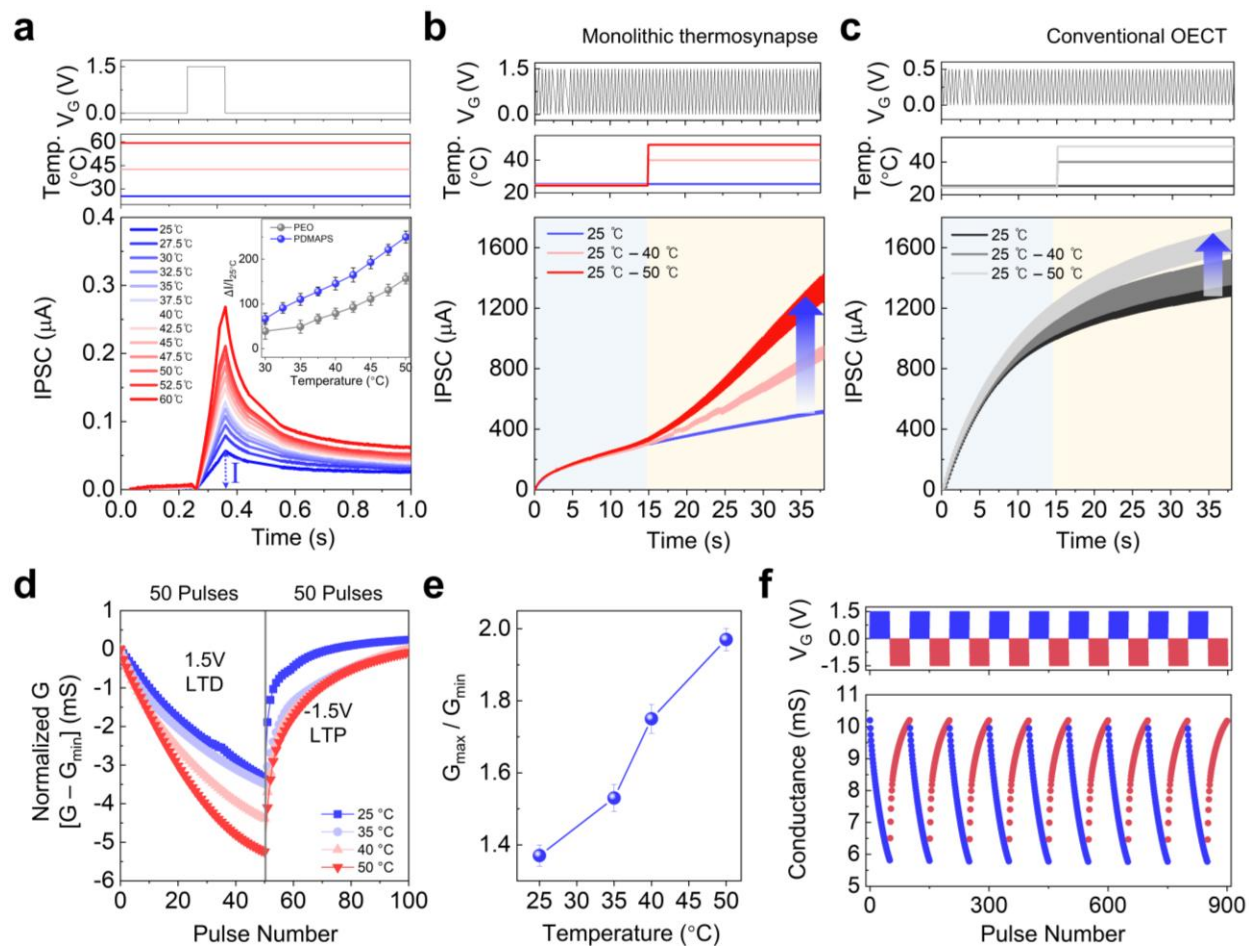


Fig. 3 | Thermally amplified sensing and synaptic modulation in monolithic thermosynapses.

a, Inhibitory postsynaptic current (IPSC) responses of the monolithic thermosynapse to a single gate pulse at different temperatures (25~60 °C). **b,c**, IPSC responses to consecutive gate pulses for monolithic thermosynapse (**b**) and conventional OECT (**c**), showing stronger thermal amplification in the thermoresponsive device. **d**, Normalized conductance modulation during long-term depression (LTD) and long-term potentiation (LTP) at different temperatures (25, 35, 40 and 50 °C) for monolithic thermosynapses. **e**, Temperature dependence of the dynamic conductance range, $G_{\max}/G_{\min}$. **f**, Reversible conductance modulation during repeated alternating gate-pulse stimulation at 40 °C, demonstrating stable synaptic operation.

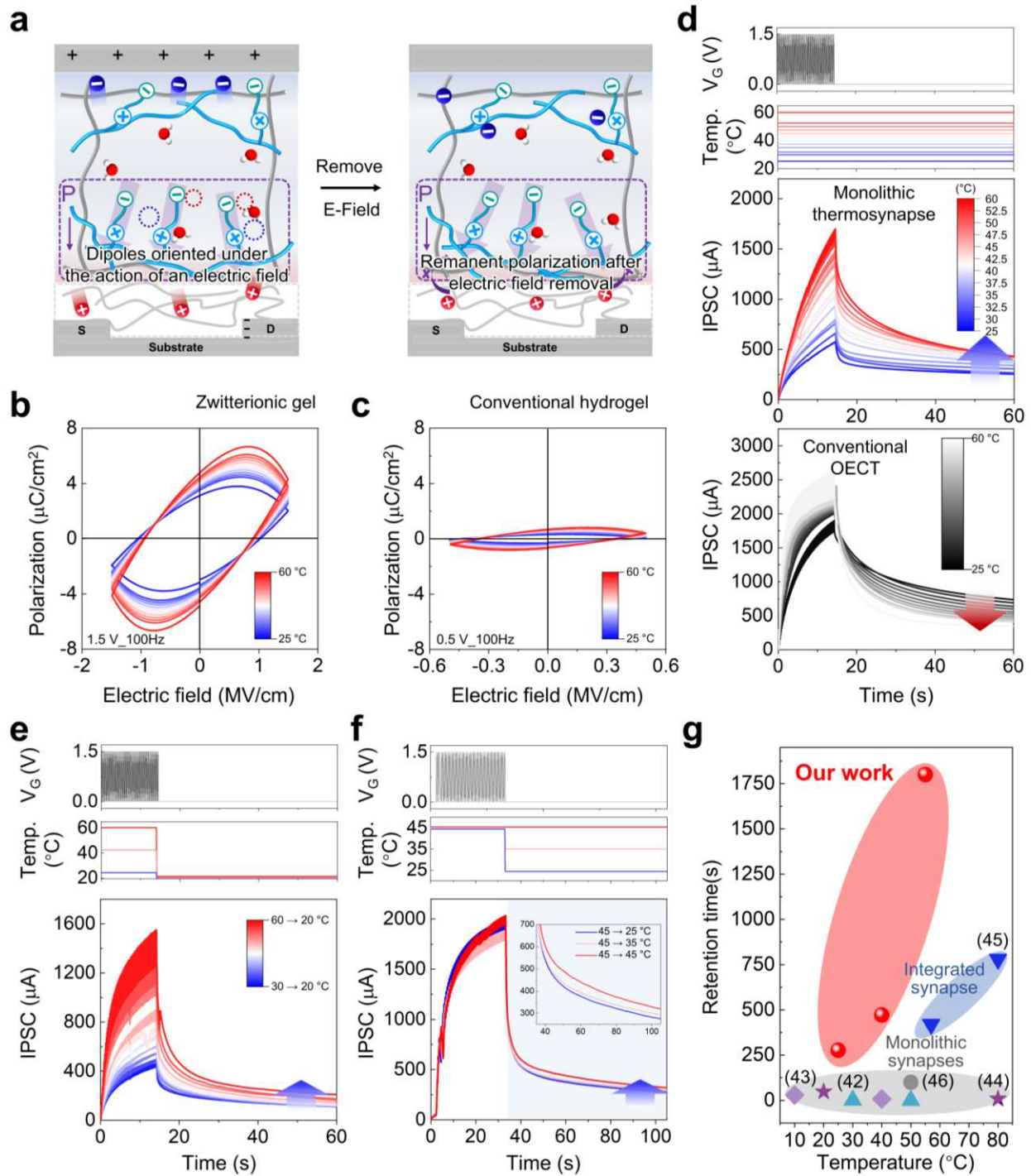


Fig. 4 | Thermal-history encoding through remanent polarization in monolithic thermosynapses. **a**, Schematic illustration of field-induced orientation of zwitterionic side chains and the remanent polarization retained after removal of the electric field, which suppresses ion back-diffusion and stabilizes the written ionic-state. **b,c**, Temperature-dependent polarization-electric field (P-E) hysteresis loops of the thermoresponsive zwitterionic gel (**b**) and conventional

hydrogel **(c)**. **d**, Inhibitory postsynaptic current (IPSC) responses during 100 consecutive gate pulses and subsequent field-off retention at different temperatures for monolithic thermosynapse and conventional OECT. The thermoresponsive device exhibits enhanced ionic-state retention with increasing temperature, whereas the conventional device shows thermally accelerated relaxation. **e**, Decoupled writing protocol in which the temperature is varied during gate-pulse stimulation and returned to a common temperature of 20 °C during retention, revealing temperature-dependent ionic-state writing. **f**, Decoupled retention protocol in which the ionic-state is written at a common temperature of 45 °C and retained at different temperatures, revealing temperature-dependent stabilization of the field-off state. **g**, Comparison of temperature-dependent retention times of previously reported conventional synapses with our work.

710

711

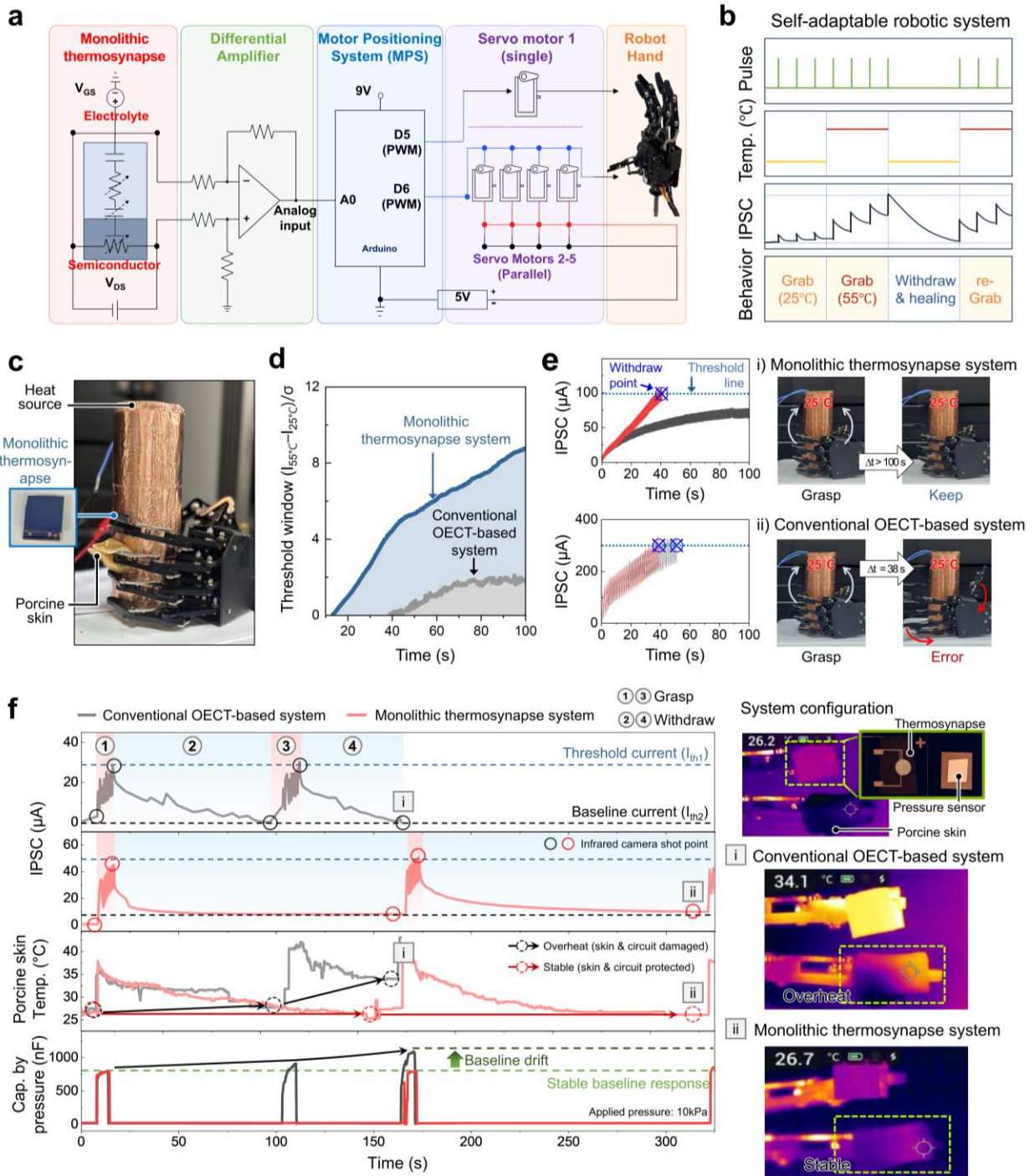


Fig. 5 | Thermal-history-dependent closed-loop robotic protection with a monolithic thermosynapse. **a**, Schematic of the integrated system comprising the monolithic thermosynapse, differential amplifier, motor-positioning controller, servo motors and robotic hand. **b**, Operating sequence of the closed-loop robotic system, in which temperature-dependent ionic-state evolution governs withdrawal and subsequent recontact. **c**, Photograph of the integrated robotic platform incorporating the monolithic thermosynapse and a porcine-skin layer as a tissue surrogate for

monitoring cumulative thermal exposure. **d**, Temperature-dependent threshold window for the monolithic thermosynapse and conventional OECT-based systems, showing greater separation of temperature-dependent electrical trajectories in the thermosynapse. **e**, Temperature-selective actuation under benign (25 °C) and elevated (55 °C) thermal conditions. The monolithic thermosynapse maintains contact at 25 °C but reaches the withdrawal threshold at 55 °C, whereas the conventional OECT-based system exhibits reduced temperature selectivity. **f**, Correlated evolution of IPSC, porcine-skin temperature and pressure-sensor capacitance during repeated grab-withdraw-regrasp cycles. Rapid state relaxation in the conventional OECT-based system shortens the interval before recontacting, resulting in cumulative heating and a shifted pressure-sensor baseline. By contrast, the retained ionic-state of the monolithic thermosynapse prolongs the waiting interval, limits cumulative heating and preserves the pressure-sensor baseline response. Infrared images show the corresponding thermal states of the integrated system.

Supplementary Files

This is a list of supplementary files associated with this preprint. Click to download.

- [SupplementaryInformationDoHwanKim.pdf](#)
- [SupplementaryVideo1DoHwanKim.mp4](#)
- [SupplementaryVideo3DoHwanKim.mp4](#)
- [SupplementaryVideo2DoHwanKim.mp4](#)