Biomimicry at the hardware level is expected to overcome at least some of the challenges, including high power consumption, large footprint, two-dimensionality, and limited functionality, which arise as the field of artificial intelligence matures. One of the main attributes that allow biological systems to thrive is the successful interpretation of and response to environmental signals. Taking inspiration from these systems, the first demonstration of using multiple environmental inputs to trigger the formation and control the growth of an evolvable synaptic transistor is reported here. The resulting transistor exhibits long-term changes in the channel conductance at a fixed gate voltage. Biomimetic logic circuits are investigated based on this evolvable transistor that implement temperature and pressure inputs to achieve higher order processes like self-regulation of synaptic strength and coincidence detection.

1. Introduction

Biological approaches to information processing are vastly different from those implemented in traditional computing systems, such as in digital signal processors. Among the most remarkable distinctions is that brains combine elements of the analog and digital domains at various scales of time and distance,[1] exhibit many-to-one-to-many connectivity,[2] implement global as well as local control of neuronal signaling,[3] and can reorganize connectivity based on necessity.[4] Taken together, these properties allow the nervous system to operate efficiently, multitask, and retain information for years at a time.

Several of the computational paradigms used by the brain have been implemented in the constantly evolving technological field of neuromorphic computing. For example, architecture-based approaches include the design of systems that replicate neural spiking and complex connectivity in the form of silicon-based spiking array processors.[5] Memristor arrays have been widely implemented in neuromorphic applications, as they feature a high degree of interconnectivity, the capacity for 3D integration, and access to multiple conductance states.[6] Three-terminal devices have also been shown to exhibit behaviors mimicking synaptic plasticity at the level of a single transistor and have been assembled into networks to accomplish classification tasks.[7] Organic electrochemical transistors (OECTs) have proven to be particularly attractive for neuromorphic applications, in part due to the recent advances reported by Keene et al. to increase the density and reduce the volatility of conductance states.[8] Additionally, the low voltages required to switch between conductance states in OECTs translate into low energy consumption.[9–11] Higher order effects, like functional connectivity and the incorporation of sensors and actuators into neuromorphic circuits, have also been explored.[7b,10]

Heretofore reported implementations of neuromorphic analog devices and circuits have proven very effective in executing a panel of biomimetic behaviors like spatiotemporal signal processing, pattern recognition, and global regulation.[12] However, in sharp contrast to biological neuronal networks, the systems investigated to date are fundamentally static in that the interconnectivity and conductance range of each component is predetermined during fabrication. Since the wiring and rewiring of neuronal connectivity is an integral mechanism of neuroplasticity,[13] we were motivated to introduce a comparable process at an analog neuromorphic device. We thus developed the first evolvable OECT (EOECT) that is capable of in situ channel formation through electropolymerization of a conductive polymer between metal source and drain contacts in addition to short-term and long-term learning behaviors.[13] The dynamic nature of the EOECT affords additional degrees of freedom in constructing neuromorphic circuits by making...
variable the conductance range, kinetics, and interconnectivity of each device.

In this work, we highlight the multifunctional nature of the EOECT by exploring various ways in which the device can be employed to process sensory information. To this purpose, we developed simple yet effective biomimetic circuits that use environmental inputs to induce channel formation and conduct growth, analogous to afferent sensory signals informing and modulating biological neural circuitry. Here, the environmental inputs include changes in temperature and pressure, which are detected by an ionic thermoelectric generator (iTEG)\textsuperscript{[14]} and a variable resistance pressure sensor,\textsuperscript{[15]} respectively. We selected temperature and pressure inputs to mimic the thermal receptors and mechanoreceptors in the skin, which also have applications in robotics and prosthetics. The two inputs that we have selected are also useful in demonstrating that the EOECT can be regulated by sensors implementing a diversity of transduction mechanisms (voltage modulation and resistance modulation) and can thus be interfaced with almost any sensor that produces an electrical response.

In addition to exhibiting growth in response to a temperature stimulus, the circuits reported here exhibit higher order neuromorphic behaviors, including self-regulation through negative feedback and the detection of coinciding inputs. One remarkable property of the EOECT is that it requires very few additional circuit components like voltage amplifiers or relays to translate a temperature input to a persistent change in channel conductance. By coupling the EOECT to the iTEG, we use the thermal voltage and current generated by small differences in temperature to induce the electropolymerization of a transistor channel. Depending on the connection scheme of the EOECT and the iTEG, the channel growth is either limited by the maximum attainable channel thickness or by a threshold value of channel conductance that drives the voltage supplied by the iTEG below the electropolymerization voltage. The coincidence detection circuit that generates channel growth during simultaneous application of temperature and pressure inputs relies on voltage inputs of opposite polarity below the polymerization threshold. In this configuration, a negative thermal voltage is applied to the gate while a positive voltage, modulated by a pressure sensor, is applied to the drain. Thus, the polymerization threshold is only surpassed when temperature and pressure inputs are applied simultaneously. While various neuromorphic devices have demonstrated coincidence detection previously\textsuperscript{[7g,h],[13]} our work is the first demonstration of self-regulated transistor growth.

### 2. Results and Discussion

Prior to circuit construction, we evaluated the performance of the iTEG component independently from the EOECT. Organic iTEGs are a new class of energy harvesting tools that can be used to convert generally wasted heat fluxes into electrical energy.\textsuperscript{[16]} Organic iTEGs can access this conversion by taking advantage of the Soret effect, or the formation of a concentration gradient along a temperature gradient (ΔT) in a multicomponent mixture. In a mixture of ionic species, the thermally generated concentration gradient produces a thermally generated voltage. By applying a range of temperature differences between the two electrodes and measuring the open circuit voltage, a Seebeck coefficient, defined as the thermal voltage (V_\text{Th}) generated per degree of temperature difference applied, of 15.5 mV K\textsuperscript{-1} was obtained (Figure S1a, Supporting Information). Then, we characterized the output current and power of the iTEG by incorporating a load resistance in the circuit (Figure 1, inset). From this circuit configuration, the output voltage across, and current through different loads can be obtained by applying a fixed temperature difference (Figure S1b, Supporting Information). Figure 1 shows the output current and power of a single iTEG as a function of output V_\text{T} when a temperature difference of 14 K is applied. Benefiting from the large Seebeck coefficient, the output V_\text{T} can reach 100 mV at the highest output power.

Commonly, a redox couple or a redox pair is required to generate an output current in an electrolyte-based device. The Seebeck coefficients of devices implementing redox couples are generally on the order of 1–2 mV K\textsuperscript{-1}.\textsuperscript{[16]} Here, in our iTEG device, however, the electrolyte does not contain any intentionally added redox couples. Several clues in the data help us better understand this effect. We noticed that the output current increases linearly with ΔT without a threshold voltage, which thus excludes the possibility of the oxygen reduction reaction contributing to the Faradaic injection of charge into the circuit (Figure S1c, Supporting Information). The possible degradation of the electrolytes during operation was also investigated using fourier-transform infrared spectroscopy. As shown in Figure S2 in the Supporting Information, the characteristic peak of the electrolyte before use and also after 2 h of use remains the same, which indicates that the electrolyte composition does not change during operation. Our hypothesis is that the observed current is due to a small amount of electrochemically active groups generated when dissolving NaOH in PEG. We suspect that this reaction is catalyzed by the Au electrodes because no output current was observed in devices based on fluorinated tin oxide electrodes (Figure S3, Supporting Information).

Regardless of the origin of the observed current, the Faradaic reaction that generates it proves stable and useful to serve as the counter-reaction for the oxidative electropolymerization of the EOECT channel. As shown in Figure S4 in the Supporting Information, when a constant ΔT is applied, the output current...
remains stable for at least 40 min. Although the iTEG current is lower than the current generated by a typical thermogalvanic cell, this application necessitates that the generated voltage surpasses the polymerization threshold, which is much more easily attained with the iTEG since its Seebeck coefficient is an order of magnitude higher than that of a thermogalvanic cell.

We then independently characterized the electropolymerization of the sodium 4-(2-(2,5-bis(2,3-dihydrothieno[3,4-b][1,4]dioxin-5-yl)thiophen-3-yl)ethoxy)butane-1-sulfonate (ETE-S) monomer on a flat gold substrate by cyclic voltammetry (CV) (Figure S5, Supporting Information). A clear oxidation peak is present above 0.2 V versus Ag/AgCl, which corresponds to the oxidative polymerization of ETE-S to produce PETE-S. It is also apparent from the CV data that the polymerization threshold occurs at a higher potential in the first cycle than in the second cycle, indicating that the thermodynamic barrier to nucleation at a bare gold electrode is higher than that of chain elongation at an established nucleation site.

Considering the oxidation potential of ETE-S, we found that we can induce the formation of a conductive channel using the electric power generated by the iTEG and thereafter dynamically control the channel conductance using a thermal input in the absence of the monomer. This behavior closely mimics the growth and function of sensory neurons, which grow in response to biochemical and electrical stimuli and once formed, convert environmental information to electrical stimuli that are then sent to the brain. As illustrated in Figure 2A, the hot side (negative electrode) of the iTEG was connected to the gate of the EOECT and the cold side (positive electrode) was grounded together with the source electrode. A small load of 1 kΩ (not shown in Figure 2A) was connected in series to the iTEG to measure the generated thermal current during operation, by measuring the voltage across the load. A drain voltage ($V_D$) of $-50 \text{ mV}$ was applied to monitor the connectivity between the source and the drain electrodes throughout the experiment. In order to enhance the generated $V_T$, we connected two iTEGs in series, such that a temperature gradient of 14 K results in an open circuit voltage of 380 mV, which thus is more than sufficient to electropolymerize the ETE-S monomer at the source electrode (Figure S6, Supporting Information). As shown in Figure 2B, $V_T$ increases rapidly when a $\Delta T$ is applied, occurring at 30 s, and gradually decreases after reaching a peak at about 6 min. The decay in $V_T$ indicates a reduction of the load resistance. This is likely due to an increase in the leakage.

Figure 2. Channel formation and device characteristics. A) Schematic illustration of the device structure prior to and after the formation of the conductive channel. In this implementation, two iTEG devices were connected in series. B) Channel formation is characterized by the current (blue) that is produced by the thermally generated voltage (red) upon the application of a temperature gradient of 15 K for the span of time between 1 and 20 min. A low $V_D$ ($-50 \text{ mV}$) was applied to characterize the $I_D$. C) Transistor characteristics under instrumental gating and thermal gating at a $V_D$ of $-200 \text{ mV}$. D) Channel formation, characterized by optical microscopy, is analogous to the growth of sensory neurons in response to a stimulus. The channels used in this work are 15 µm in length and 150 µm in width. E) Once the sensory neuron is formed, it can respond to a temperature stimulus.
current that accompanies a thicker EOECT channel, since this decay is not evident when the iTEG is coupled to a fixed transistor (see Figures S1b and S4 in the Supporting Information). The drain current \( (I_D) \) exhibits an initial phase of linear growth, after which it shows signs of saturation due to the reduction in the thermal \( V_T \) and possibly also due to side-reactions or stearic blocking of reactive polymerization sites on the linear polymer chain. A decrease in the electropolymerization efficiency with increasing polymer thickness is also evident from the CV characteristics (Figure S5, Supporting Information), as each subsequent cycle produces a reduced change in capacitance.

We demonstrate that the transistor can also be thermoelectrically modulated within the range of maximum transconductance (Figure 2C). Since the channel material is intermediately doped in its pristine state, the transconductance \( (\partial I_D/\partial V_T) \) peaks at the relatively low value of \(-90 \text{ mV}\) and renders this device an effective amplifier of temperature signals produced by the thermoelectric generator. Channel formation between the source and drain contacts was verified by optical microscopy (Figure 2D). The resulting evolved channel is a functional OECT that behaves similarly to the previously reported EOECT that implements an electropolymerized PETE-S channel, analogous to a formed sensory neuron reporting information to the brain (Figure 2E).[13]

Biological mechanisms of self-regulation and negative feedback are essential for living systems to maintain homeostasis and optimal function at various scales.[18] At the level of the synapse, for instance, presynaptic neurons are equipped with autoreceptors that detect the neurotransmitter in the synaptic cleft and act to inhibit its production and release (Figure 3A).[19] We take advantage of the iTEG as a load-dependent voltage generator to construct self-regulating devices, thereby mimicking autoreceptor behavior. By altering the wiring between the two devices such that the load on the iTEG changes dramatically during channel formation, we can attain self-regulated channel growth. When the source and drain are connected to opposite sides of the thermoelectric generator, as they are in Figure 3B, the formation of the channel leads to a dramatic drop in the total load resistance in the circuit and a corresponding drop in the output voltage across the thermoelectric device. Initially, when a temperature gradient is applied across the iTEG (after \( \approx 5 \text{ min} \)), ionic thermodiffusion produces an increase in voltage between the source and the drain (Figure 3C). No detectable current is evident, however, until the voltage surpasses the electropolymerization threshold at \( 0.2 \text{ V} \) with the onset of Faradaic processes at the gate and drain electrodes. Once the PETE-S channel closes the gap between the source and drain electrodes, a sharp current increase is observed, accompanied by a marked output voltage drop due to the reduction in the load resistance (here defined as the EOECT channel that is developing between the source and drain). Figure 3D highlights the behavior of the system when the connection is made. Considering that a time of \( 20 \text{ s} \) is required from the onset of polymerization to channel formation, we can calculate that about \( 0.19 \mu \text{C} \) of charge is required to form a new connection. Curiously, the current increase persists even as the voltage applied to the drain dips below the value required.

Figure 3. Self-regulation: A) Self-regulation is evident in a biological synapse when autoreceptors (blue) block the synthesis and/or release of neurotransmitters (pink). B) Connection scheme of a self-regulating device. C) Behavior of the \( I_D \) and \( V_T \) upon application of a 15 K temperature gradient across the thermoelectric generator. The time interval during which a temperature gradient was applied is marked in gray. An arrow marks the time at which the transistor was disconnected from the thermoelectric device. D) An expanded graph showing the behavior of the \( I_D \) and the \( V_T \) upon the onset of electropolymerization and when an electrical connection is made between the source and drain contacts.
for electropolymerization. This effect is necessarily a result of an increase in channel conductance, since the drain voltage and gate voltage are also decreasing at this time. One possible explanation for this effect is that radicals generated within the channel persist beyond the timeframe of the pulse due to the considerably slower diffusion within the solid channel matrix. Experiments are currently underway to fully understand this behavior. When the EOECT is taken out of the circuit after \( \approx 12 \) min, the \( V_T \) across the iTEG quickly reverts to the open circuit values while the current drops to values below the noise level of the measurement. As the temperature gradient is removed after \( \approx 15 \) min, the \( V_T \) also drops to baseline levels.

Coincidence detection is another important biological function that can be replicated by providing independent inputs to the gate and drain contacts of an EOECT. We first demonstrate this concept using a two-channel potentiostat to apply the input voltages (Figure 4A). For channel growth to occur, a minimum voltage difference of 0.2 V must be applied between the gate and either the source or the drain contact, with the negative terminal located at the gate. To achieve coincidence detection, which can also be described as an AND gate-type behavior with respect to channel growth, we take advantage of principles commonly applied in passive matrix addressing. Voltages of opposite polarity that lie below the ETE-S polymerization threshold (+/- 0.15 V) were selected. A \( V_D \) of -0.2 V at a \( V_G \) of 0 V was used to monitor conductance before and after the interrogation of a set of conditions. It was observed that an electrical connection between the source and the drain is only made when +0.15 V is applied to the drain and -0.15 V is applied to the gate (Figure 4A, red line) and not when either voltage is applied on its own (Figure 4A, blue and black lines). Splitting the voltage required for electropolymerization between two sources presents several advantages. In mimicking the biological process of coincidence detection, the voltage splitting configuration is beneficial in that it requires the simultaneous application of two independent inputs to induce channel growth. In applications where small fluctuations in only one input voltage are to be measured, the second voltage can be set to just below the electropolymerization voltage to suppress the inherent threshold of this technique.

We implement a more relevant form of coincidence detection by tracking sensory inputs from two environmental monitors—a temperature and a pressure sensor—into a simple circuit where the inputs control the channel growth of an EOECT (Figure 4B). As in the self-regulated iteration of the device, a temperature input is applied to the gate while a variable resistance pressure sensor[15] is connected in series with the drain voltage of 0.1 V. A 22 k\( \Omega \) regulation resistor in parallel with the EOECT channel was employed to obtain the desired voltage division to enable the growth of the channel, together with the resistive pressure sensor. A characterization of the resistance of the pressure sensor in the on and off states, as well as negative controls excluding the possibility of polymerization being initiated by either of the inputs alone, are outlined in Figure S7 in the Supporting Information. The circuit response to a series of three 30 s pressure pulses at a range of temperature gradients was then evaluated (Figure 4C). A pressure of 3 kPa was used to evaluate this system, which is near the pressure threshold detectable by human mechanoreceptors (≈5 kPa)[15] and well below the human pain threshold limit of 330 kPa.[20] The error bars reflect the change in the \( I_D \) over the course of a single pressure pulse. Below a certain temperature gradient...
threshold, which falls between 2 and 4.2 K, minimal changes in channel conductance are observed, whereas above this threshold, the channel exhibits an increase in conductance. The temperature gradients of 2 and 4.2 K applied at the iTEG correspond to voltage differences of 60 and 125 mV, respectively. Again, indications of subthreshold polymerization are observed between pulses when the pressure is removed. As in the self-regulated device, subthreshold electropolymerization can also be explained by the persistence of reactive monomer and polymer radical species within the channel after the voltage is removed. When the temperature gradient is removed, the channel conductance stabilizes at a constant value.

3. Conclusion

The results presented here are meant to demonstrate the diversity of biomimetic functionalities that can be attained when EOECTs are integrated into fairly simple circuits. From our experiments, we establish that environmental inputs can be used to produce unregulated and regulated EOECT growth as well as coincidence detection, but there is ample space to further develop the concept of in situ fabrication and modification of electronic components. The research field of evolvable devices and hardware promises to be especially fertile because evolvability is not limited to OEFTs, but can also be extended to produce evolvable resistors, capacitors, and diodes. There is an ever-growing number of known mechanisms by which the brain interprets and retains information. Incorporating a chemical regulatory pathway into an electrical circuit is analogous to adding a supplementary, parallel level at which this system can be manipulated and trained. The additional degrees of freedom provided by the chemical capacity to activate latent synapses and modulate their conductance in situ directly parallel the information-processing strategies of neural remodeling. By adding redundant modes of modulating the synaptic weight, we increase the complexity of the system and the number of levels on which it can be controlled, leading to a greater likelihood of observing emergent properties.

4. Experimental Section

Fabrication of the Thermoelectric Device: Au electrodes were prepared through thermal evaporation of 5 nm Cr followed by 40 nm Au on glass substrates. PDMS with thickness of 1 mm was used as spacer between the two electrodes to form a chamber. NaOH pellet was directly added as the gate electrode. Device images were collected using a Nikon light microscope and processed using the NIS-Elements Viewer software. The electrolyte was injected into the chamber by syringe. Fabrication of the Pressure Sensor: Three components: 1) poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate) (PEDOT:PSS, Heraeus Clevios, PH1000, 1.3 wt%), 2) glycidoxypropyl trimethoxysilane (GOPS, Alfa Aesar, 97 wt%), and 3) carboxymethylated nanofibrillated cellulose (NFC, 0.5 wt%) were mixed in a dispersion by an ULTRA-TURRAX T-10 disperser. The mixture dispersion was then dropped into an aluminum mold and covered by a glass slide. Dispersant in the mold was frozen by liquid nitrogen, followed by freeze drying (Benchtop Pro) with a setting of −60 °C and 200 jbar for 12 h. An aerogel with the same dimension (18 mm length, 9 mm width, and 4 mm height) of the dispersion was produced by the freeze-drying process. The aerogel was then put into an oven at 140 °C for 30 min. This aerogel was connected with two electrodes (PEDOT:PSS coated glass slides) on the bottom and top. Flexible carbon fibers were then used to connect the electrodes with the remaining parts of the device in series.

Synthesis and Characterization of ETE-S: Detailed information on the synthesis and characterization of the ETE-S monomer can be found in the Supporting Information.

Fabrication of the Source and Drain Contacts for the EOECT: The gold source and drain contacts and the SU-8 insulating layer were fabricated on 4 in. silicon wafers (1 µm thermally grown oxide) using conventional photolithography protocols, which were followed by gas-phase modification of the exposed silicon oxide by aminopropyl triethoxysilane (APTES). Detailed information on the microfabrication can be found in the Supporting Information. An Ag/AgCl pellet (MultiChannel Systems) was used as the gate electrode. Device images were collected using a Nikon light microscope and processed using the NIS-Elements Viewer software. Electropolymerization of the channel was carried out in a standard solution containing 1 mg mL⁻¹ ETE-S and 10 × 10⁻³ m NaCl electrolyte.

Thermoelectric Characterization: A custom thermoelectric setup, consisting of a Peltier heating element connected to a Keithley 2230 DC voltage power source, was used to apply the temperature gradient across the thermoelectric device. The Vₜ was measured using Keithley Nanovoltmeter (Model 2182A) between the cold and hot electrodes of the device. The current in the thermoelectric circuit was determined by measuring the voltage across a resistor of known value (1 kΩ) connected in series with the thermoelectric device (using the second channel of the same Nanovoltmeter). When the EOECT was connected to the iTEG, a potentiostat (Bio-Logic SP300) was used to apply the V₀ and measure the I₀ in Figure 2D (30 mV), Figure 2E (−0.2V), Figure 4A (variable voltage), and Figure 4C (0.15 V).

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

coincidence detection, mixed ion-electron conductors, neuromorphic computing, organic electrochemical transistors, self-regulation
