Adaptive Biosensing and Neuromorphic Classification Based on an Ambipolar Organic Mixed Ionic–Electronic Conductor

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Organic mixed ionic–electronic conductors (OMIECs) are central to bioelectronic applications such as biosensors, health-monitoring devices, and neural interfaces, and have facilitated efficient next-generation brain-inspired computing and biohybrid systems. Despite these examples, smart and adaptive circuits that can locally process and optimize biosignals have not yet been realized. Here, a tunable sensing circuit is shown that can locally modulate biologically relevant signals like electromyograms (EMGs) and electrocardiograms (ECGs), that is based on a complementary logic inverter combined with a neuromorphic memory element, and that is constructed from a single polymer mixed conductor. It is demonstrated that a small neuromorphic array based on this material effects high classification accuracy in heartbeat anomaly detection. This high-performance material allows for straightforward monolithic integration, which reduces fabrication complexity while also achieving high on/off ratios with excellent ambient p- and n-type stability in transistor performance. This material opens a route toward simple and straightforward fabrication and integration of more sophisticated adaptive circuits for future smart bioelectronics.

1. Introduction

The success of many organic bioelectronic applications results from reduced interface impedance and extraordinarily high electrochemical transconductance (ion-to-electron transduction) in organic mixed ionic–electronic conductors (OMIECs).[1,2] These and other unique properties such as biocompatibility, low voltage operation, simple fabrication processing, and stretchable and flexible materials have opened a wide variety of possible bioelectronic applications such as neural probes[3] and sensors for ions,[3] metabolites,[4] bacteria,[5] and viruses[6] that benefit from the extensive capabilities in tailoring molecular structures and functions via chemical synthesis. Furthermore, the dynamic range and ability to dope these polymers electrochemically have produced tunable analog neuromorphic devices[7,8] with high stability,[9] resulting in proof-of-principle circuits[10,11] as well as biohybrid systems directly modulated by locally secreted neurotransmitters.[12]

Current organic bioelectronic applications are commonly based on commercially available blends of poly(3,4-ethylene dioxythiophene):polystyrene sulfonate (PEDOT:PSS), which is a p-type (hole) conductive polymer that operates in depletion-mode. This material is conductive as-prepared and can be turned off by applying a gate potential. Enhancement-mode p-type devices exist, by design,[13] or by the dedoping of PEDOT:PSS with small molecule amines.[14] However, a combination of p- and n-type materials is required to form logic gates and complementary circuits[15] and, to date, highly stable and high-performance n-type materials are rare.[16] As a result, there is a clear need for stable materials that can operate at biological interfaces and that are able to transduce ionic to electronic signals effectively. In fact,
Figure 1. Schematic of an adaptive neuromorphic biosensor. The adaptive organic electronic circuit can locally process (for example, amplify, rectify, normalize, etc.) biosignals by tuning the gain peak of the inverter using a neuromorphic element. Simultaneously, a hardware-based neural network could classify input signals, for instance, to detect heartbeat anomalies. Both functionalities can be realized using a single OMIEC (P-3O).

despite the vast amount of information available at the physiological level, thus far no smart and integrated bioelectronic circuits exist that can locally adapt to and process information. At the same time, neuromorphic systems have proven successful in typical classification tasks such as digital image recognition, but nevertheless (partly) rely on software and are still outperformed by traditional software neural networks. However, for applications requiring the local, real-time processing of biological and physiological data, the use of software is undesirable and can be problematic. Consequently, bioelectronic circuits integrated with neuromorphic systems could provide the rapid on-site processing and classification of biosignals without the need for external computation and signal analysis to generate relevant optimized actions such as drug delivery or prosthetic movements.

In this work, we show an adaptable circuit that can locally process and tailor biologically relevant signals, such as electromyograms (EMGs) and electrocardiograms (ECGs), that serve as inputs to a neuromorphic classification array and act directly as an in-sensor computing system (see Figure 1). Our system is based on a monolithically integrated semiconducting polymer that forms both the ambipolar inverter for signal amplification and normalization, as well as the neuromorphic memory element to tune the gain properties locally by modulation of the conductance (or synaptic weight). The nonvolatile tuning of the neuromorphic elements also allows for hardware neural network integration and the local classification of signals. The use of a single polymer mixed conductor significantly reduces fabrication complexity, while its unrivaled performance and ambient stability highlight a route toward long-term smart sensing systems and chronic implants.

2. Results and Discussion

Electronic circuits operating at the interface with biology require both p-type and n-type OMIECs. To date, the number of n-type semiconductors as well as their performance is still inferior compared to their p-type counterparts. The key issues are that doped n-type materials are generally unstable when exposed to air and achieving high electron mobilities has proved more difficult than for their p-type counterparts. To reduce the fabrication complexity ambipolar materials are desirable as it decreases the number of lithography steps. At the same time, ambipolar materials impose the challenging constraint of being able to be doped by both cations and anions as well as conducting both holes and electrons. One strategy to design ambipolar polymer mixed conductors is copolymerizing an electron deficient moiety (acceptor), such as naphthalenediimide (NDI) or diketopyrrolopyrrole (DPP), with an electron rich moiety (donor) like thiophene to obtain a narrow bandgap. These donor–acceptor (D–A) polymers are modified with hydrophilic ethylene glycol (EG) type side chains for aqueous electrolyte gated organic electrochemical transistors (OECTs). The EG chains support highly efficient electrochemical doping within the whole volume and enables low voltage (≤1 V) operation, which is essential when using an aqueous electrolyte. These OECTs exhibit p- and n-type characteristics, and are integrated into ambipolar inverters for logic circuits and biosignal amplifiers. In order to operate stable OMIEC devices properly in n-type mode in water, a lowest unoccupied molecular orbital (LUMO) level below −4 eV is required to avoid side reactions with water and oxygen. High-performance OMIECs additionally require balancing ionic and electron/ hole mobility, while maintaining unperturbed crystalline structures upon electrochemical doping. It has been reported that embedding sp3-N into polymer backbones results in a more planar structure, enhancing the π−π stacking and increasing the electron mobility. Capitalizing on these properties, the material we present here is an NDI-bithiozale D–A copolymer (referred to as P-3O, shown in Figure 1) functionalized with EG side chains, which allows for a stable transistor operation both with a solid electrolyte as well as in aqueous environments.

Ideally, a multi-sensor approach for on-site classification and personalized actions based on biosignals requires a method for local signal processing. The ability to amplify low signal-to-noise ratio biosignals is also important, particularly for small signals like EEG. However, any computational action such
as a decision or classification is still made externally. In order to judge and process a plethora of signals, local amplification, and normalization is needed. At the same time, in-sensor computing requires the modulation of signals over a variety of inputs, while ensuring that signals are correctly tuned to the synaptic weights for use in (neuromorphic) classification algorithms. Furthermore, relevant information could be lost if signals from one sensor (e.g., ECG) outweigh others (e.g., EMG). Our proposed adaptive neuromorphic circuit is schematically shown in Figure 1, where various electrophysiological signals can serve as an input for local adaptive processing (such as normalization or rectification). The circuit consists of an ambipolar inverter combined with an adaptive memory element and is based entirely on the ambipolar material P-3O.

The ambipolar inverter comprises two complementary transistors, one operating in n-type and one in p-type mode. An all-solid-state environment is desirable in applications associated with electronic circuitry, logic circuits, and neuromorphic computing, as the operation of circuits in aqueous environments can lead to interference with the surrounding electronics and neighboring devices. From a technological point of view, the use of liquid electrolytes limits the high-density integration of devices. The solid electrolyte consists of an ionic liquid (1-ethyl-3-methylimidazolium bis(trifluorosulfonyl) imide, EMIM:TFSI) within a polymer matrix (poly(vinylidene fluoride-co-hexafluoropropylene), PVDF-HFP), which has been demonstrated to be able to operate at kHz frequencies.

Figure 2A shows the architecture of the solid electrolyte gated...
device. The devices can be designed with a side gate, which simplifies the fabrication by avoiding a complex layer-by-layer process to construct a stacked structure. Figure 2B,C shows the output and transfer characteristic of the P-3O OECT exhibiting near-symmetric ambipolar electrical behavior when operated in the ambient environment with a solid gel-electrolyte. The polymer achieves an on/off ratio of 10^4 and stable operation for more than 20000 cycles for both p- and n-type in ambient conditions (Figure 2D,E).

Since both the p- and n-type transfer curves have near-identical behavior (shown in Figure 2C), the ambipolar inverter can be constructed. An inverter is a fundamental building block of electronic circuits and is used in both digital circuits such as logic gates, as well as in analog circuits such as voltage amplifiers. It inverts the applied input voltage \( V_{\text{in}} \) from a high to low voltage and vice versa (see Figure 2F,G). At the transition region, it shows a high gain which makes organic inverters relevant for a wide variety of applications in several fields, including printed electronics, imperceptible and wearable electronics, sensors and bioelectronics.[22,31,32] Figure 2F shows the performance of the solid-state ambipolar inverter with a gain of 12 at \( V_{DD} = 0.8 \) V, along with Figure 2G presenting the logic NOT gate operation where a high input results in a low output voltage.

Pairing the P-3O ambipolar inverter with electrochemical random-access memory (EC-RAM) at the inverter gate allows for the construction of a neuromorphic inverter that can locally tune the amplification of input signals (see Figure 3A). The P-3O EC-RAMs adopt a similar configuration as the solid-state electrochemical transistor (Figure 2A) and maintain non-volatile memory by employing an open gate circuit in combination with a current limiting resistor. Figure 3B shows the excellent state retention of 6 representative memory states across two orders of magnitude for 5 min in a nitrogen environment. By tuning the neuromorphic memory device to different conductance values, we can directly and locally tune the gain of the adaptive inverter circuit (see Figure 3C), a characteristic feature used to extract, for example, electro-physiological signals.[33] Lowering the conductance results in a shift in the transition region to higher input voltages as well as a broadening of the transition region. The opposite is true for an increase in the conductance, which moves the transition region to lower input voltages and makes it narrower. As such, we can adjust the fully integrated adaptive neuromorphic sensing circuit to modulate and normalize biosignals of different amplitudes without increasing fabrication complexity, assuming signals with a sufficient signal-to-noise ratio. Figure 3D,E demonstrates the signal normalization of EMG signals for two different gestures using the behavior of the neuromorphic inverter (see the Supporting Information). The signal envelope (an essential feature encompassing relevant information of the gesture) of the wrist extension (Figure 3D) is correctly preserved after normalization using adaptive state 1 whereas the signal envelope of a clenched fist is completely lost.

Figure 3. A) Schematic of an adaptable inverter including an EC-RAM in series with the gate of the ambipolar inverter. B) State retention of EC-RAM in a N₂ environment for 5 min at different conductance states over two orders of magnitude. C) The top panel shows the behavior of the neuromorphic inverter (\( V_{DD} = 0.8 \) V) when tuning the EC-RAM to different conductance states and bottom panel shows the corresponding gain. The dashed lines represent the experimental data, and the two solid lines represent the simulated inverter behavior of two different EC-RAM conductance states. State 1 resembles a neuromorphic inverter with a negligible EC-RAM resistance whereas State 2 represents one with a high EC-RAM resistance (\( C = 167 \) nS). D,E) The use of the neuromorphic inverter applied to locally amplify EMG signals from two different gestures, wrist extension and clenching of a fist, respectively. Each signal can be normalized between 0 and \( V_{DD} \), depending on the state of the neuromorphic inverter.
To correctly retrieve the signal envelope after amplification the signal of a clenched fist should be normalized using adaptive state 2 (Figure 3E, red line). While the two different gestures require a different amplification of the biosignals to correctly scale and retrieve the signal envelope, the adaptive nature of the neuromorphic inverter makes it generalizable to local processing applications such as smart autonomous robotics,[34] in-sensor classification and prosthetic motion control,[19] or for further processing and classification in hardware-based neural networks.[18]

In fact, when integrated into large crossbar arrays (Figure 4A), P-3O EC-RAMs can be used as the synaptic weights in an artificial neural network for local classification. In this array, each weight in the hardware neural network is represented by the conductance of an EC-RAM, which can be randomly accessed over a large range and in an analog fashion, enabling parallel computation[10] and increasing energy efficiency. Organic EC-RAMs are able to operate at low voltage and with high write speeds (20 ns pulse).[35] The P-3O EC-RAMs operate in n-type mode (positive read voltage, ±5 V on the gate circuit with the current limiting 1 GOhm resistor, writing speed less than 300 ms) and can access over 2^5 conductance states, in linear fashion, and with high state retention (see Figure 4B). Operating in ambient conditions mandates encapsulation due to the presence of oxygen and water (see Figure S19 in the Supporting Information). The state retention can be further improved by optimizing the device geometry and purifying the ion gel. Furthermore, to avoid possible crosstalk in neuromorphic array operation, other devices geometries like vertical gate architecture[36] or patterning ion gel[37] can be used. Our

Figure 4. A) Schematic representation of a neuromorphic array based on P-3O EC-RAMs. B) 14 cycles of 32 conductance states showing linear potentiation and depression (pulse width 300 ms and delay 500 ms). C) 5 different ECG classes represent the normal sinus rhythm (SNR) and 4 heartbeat anomalies. D) Neural network architecture consisting of 125 input neurons, 1 hidden layer with 100 neurons and 5 neurons in the output layer. E) Classification accuracy as a function of the training epochs for both the hardware neural network based on P-3O (red) and the ideal software limit (blue). F) Confusion matrix for all 5 classes using the hardware-based network.
n-type P-3O-based EC-RAM exhibits comparable performance to p-type polymer based EC-RAMs such as PEDOT:PSS,[39] p(g2T-TT),[15] and P3HT.[16] Even though the P-3O polymer is ambipolar, in p-type mode (negative read voltage) EC-RAM decays fast, even in nitrogen atmosphere (see Figure S20 in the Supporting Information), which might indicate that the energetic barrier for back diffusion of the anions (TFSI⁻) is low;[38] and the generated holes are unstable due to the low-lying HOMO (Table S1, Supporting Information).[39] The linear and symmetric behavior of potentiation and depression of the conductance (Figure 4B) results in efficient training and inference of the neural network. To highlight this, we simulate the behavior of a neuromorphic array based on the P-3O EC-RAM, in its performance to recognize and classify heartbeat anomalies in ECG signals (see the Supporting Information). Figure 4C shows the five different ECG classes serving as the input for the neural network depicted in Figure 4D. ECG signals are generally very similar and known to be difficult to classify,[40] hindering real-time diagnoses. Local processing and computation using a hardware-based neural network could facilitate on-site real-time classification. Figure 4E shows that training an EC-RAM based neural network approaches ideal numerical accuracy (over 70% training accuracy after 20 epochs), which is the theoretical limit for this algorithm. The corresponding confusion matrix is presented in Figure 4F, demonstrating the successful classification of the five ECG classes based on the EC-RAM hardware array. Note that increasing the complexity of the neural network architecture can lead to higher accuracies. However, since in this work we focus on hardware neural networks and specifically the EC-RAM functioning within a neural network, we have chosen to restrict the neural network architecture to a single fully connected layer with one hidden layer. On the other hand, this network structure still faces a significant integration challenge which would benefit from downscaling the network structure and sacrificing accuracy or implementing more complex neural network architectures (e.g., convolutional and pooling layers, or a recurrent neural network) or additional preprocessing steps (e.g., Fourier transform).

Conventional complementary circuits based on a combination of a p- and n-type material commonly require multiple or complex fabrication steps. Using our ambipolar conductive polymer, we can significantly decrease this complexity by monolithic fabrication, including the added neuromorphic traits. This expands the capabilities of the complementary circuits in bioelectronics and sensors, in which OMIECs have already proven to be powerful due to their large volumetric ionic capacitance, which allows for significant amplification when transducing biological signals. In biological, aqueous environments, stable p-type OMIECs are common, whereas high-performance n-type materials, in terms of stability and electrical behavior, are not. However, for biosensors detecting anions or some metabolites like glucose, stable n-type materials are crucial.

We studied the behavior of a 100 × 10⁻³ m NaCl aqueous electrolyte gated OECT using P-3O as the active channel, and Ag/AgCl as the gate electrode (shown in Figure 5A). In this configuration P-3O also exhibits ambipolar behavior in transistor performance (see Figure 5B,C for the output and transfer curves), which indicates that the polymer could also be doped by both cations (Na⁺) and anions (Cl⁻). In n-type mode, the P-3O polymer works exceptionally well with a stable on/off ratio of approximately four orders of magnitude for more than 21 600 cycles, shown in Figure 5D. After continuously operating for more than 60 h, the on-current only exhibits a slight decrease (~25%) but maintains the on/off ratio at similar orders of magnitude (see Figure S13 in the Supporting Information). The remarkable stability is a result of the precise tailoring of the energy levels (LUMO ~4.30 eV, see Figure S10 and Table S1 in the Supporting Information) with respect to oxygen and water.[21] Despite the extremely high stability of the p-type mode transistor using ion-gel, the material performance in liquid electrolytes in p-type mode is less (see Figure S14 in the Supporting Information), showing a two order of magnitude on/off ratio for over two hours operation. The aqueous electrolyte operated ambipolar inverter and the tuning of it using the EC-RAM is demonstrated in Figures S22 and S24 in the Supporting Information. We further show the exceptional n-type performance of the P-3O polymer in an aqueous environment by demonstrating its behavior as a neurotransmitter sensor (see Figure 5E), which has potential in advanced biohybrid systems.[18] We adopted the Au side-gated device configuration in Figure 2A and monitored the drain current at a drain potential of Vd = 0.1 V, while oxidizing the dopamine at the gate electrode by applying a gate potential of Vc = 0.6 V. A faradaic gate current was observed when different concentrations of dopamine analyte were added (Figure 5E bottom panel), which lowers the potential drop at the Au gate and electrolyte interface, leading to more effective gating at the channel (see Figure S15 in the Supporting Information). Meanwhile, the drain current increases in response to increased dopamine concentrations (Figure 5E top panel and Figures S16 and S17 in the Supporting Information). These results prove the interaction of P-3O as an excellent electron conductor with neurotransmitters like dopamine, showing its potential in adaptive biohybrid applications.

3. Conclusion

Due to the intrinsic ionic–electronic coupling, organic mixed ionic–electronic conductors exhibit great potential not only in bridging electronics and biological systems, but also in operating as non-volatile memory devices in hardware neural networks. In this work, we have presented the capability to integrate individual devices into circuits for applications like adaptive biosensors and brain-inspired computing, utilizing the ambipolar ND1-bithiazole conjugated polymer P-3O. Though ambipolar materials do not always have better performance, the remarkable characteristics of this semiconducting polymer, including symmetric p- and n-type transfer curves, high performance, and ambient stability, allow for straightforward monolithic fabrication and integration of single devices, including transistors, logic gates, neuromorphic memory, and sensors. Though neural network simulations based on experimentally obtained behavior of P-3O EC-RAM demonstrate the capabilities of local classification of biosignals, full hardware integration remains a challenge. This is partly because of the EC-RAM encapsulation requirements, but mostly due to the complexity in hardware-based learning rules (e.g., gradient descent and
were prepared in chloroform at the concentration of 5 mg mL\(^{-1}\).

Monolithically integrated adaptive sensor clearly highlight a path toward next-generation adaptive and personalized bioelectronic applications.

4. Experimental Section

Materials: The P-3O synthesis details and characterization are described in the Supporting Information. Chloroform, 1-ethyl-3-methylimidazolium bis(trifluorosulfonyl)imide, poly(vinylidene fluoride-co-hexafluoropropylene), and dopamine hydrochloride are purchased from Sigma-Aldrich and used as received. The ion gel solution was prepared following the reported method\(^{[29]}\) without further baking the ionic liquid. Ionic liquid (EMIM:TFSI) and poly(vinylidene fluoride-co-hexafluoropropylene) (4:1 w/w) were dissolved in acetone with the following proportions: 17.6 w/o ionic liquid, 4.4 w/o polymer, and 78 wt% solvent. The resulting ion gel solution was stirred at 40 °C for at least 30 min. The interdigitated microelectrodes IDA-Au-6 (channel length l 5 µm, individual channel width, w, 1.8 mm, number of pairs 30, number of channels, n, 59, total channels width, W = n x w = 10.62 cm) are purchased from MicruX technologies.

Device Fabrication and Characterization: The polymer solutions were prepared in chloroform at the concentration of 5 mg mL\(^{-1}\). The interdigitated microelectrodes were treated with UV ozone for 15 min, following spin-coating the polymer solutions at 1000 rpm for 30 s. The samples were annealed on the hotplate at 100 °C for 30 min. For side gate devices, the gate and active channel were separated by excimer laser ablation. The ion gel solution was drop-cast on the top of the active area and dried in the fume hood. For EC-RAM measurements, the devices were stored in the nitrogen atmosphere glovebox. The electrical characterization of EC-RAMs is recorded by Keithley 2602B with Labview and the inverter data is acquired with a NI DAQ USB-6363 and with Keithley SMU 2636B controlled with Matlab software.

Supporting Information

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

Acknowledgements

Y.Z. and E.R.W.v.D. contributed equally to this work. Y.Z., E.R.W.v.D., and Y.v.d.B. acknowledge financial support from The European Union’s Horizon 2020 Research and Innovation Programme, Grant agreement no. 802615. G.Y. and J.S. acknowledge The National Natural Science Foundation of China 61620106016/61835009/61775145. Additionally,
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
adaptive sensing, ambipolar inverters, neuromorphic computing, organic mixed ionic–electronic conductors

Received: January 13, 2022
Revised: March 15, 2022
Published online: April 17, 2022

[1] G. Y. acknowledges The China Postdoctoral Science Foundation Funded Project Grant 2020M672771 and Guangdong Basic and Applied Basic Research Foundation 2020A1515110636.

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

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