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Reconfigurable organic logic devices are promising candidates for next generations of efficient computing systems and adaptive electronics. Ideally, such devices would be of simple structure and design, be power efficient. and compatible with high-throughput microfabrication techniques. This work reports an organic reconfigurable logic gate based on novel dual-mode organic electrochemical transistors (OECTs), which employ a self-doped conjugated polyelectrolyte as the active material, which then allows the transistors to operate in both depletion mode and enhancement mode. Furthermore, mode switching is accomplished by simply altering the polarity of the applied gate and drain voltages, which can be done on the fly. In contrast, achieving similar mode-switching functionality with other organic transistors typically requires complex molecular design or multi-device engineering. It in shown that dual-mode functionality is enabled by the concurrent existence of anion doping and cation dedoping of the films. A device physics model that accurately describes the behavior of these transistors is developed. Finally, the utility of these dual-mode transistors for implementing reconfigurable logic by fabricating a logic gate that may be switched between logic gates AND to NOR, and OR to NAND on the fly is demonstrated.

1. Introduction

Efforts to miniaturize conventional complementary metaloxide-semiconductor (CMOS) transistors are not expected to

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result in further improvements in energy efficiency, or to provide significant economic benefits. Several emerging technologies employing new materials and new device structures have been identified as possible candidates for post-CMOS electronics.^[1-7] Among these, reconfigurable logic circuits are of particular interest because they make it possible to simplify circuit design while increasing energy efficiency.^[8–13] The design of reconfigurable logic circuits is based on the use of individual transistors that exhibit multiple functionalities. The most common way to achieve such capabilities is to employ ambipolar field-effect transistors with dynamically gate-tunable doping types. For instance, using double-gate 2D homojunction devices, Pan et al. showed that the polarities of gate voltages determined the major carrier types in ambipolar 2D semiconductors, thus altering the transistors into n-type or p-type.^[8] Using different combinations of applied electrical input

signals, these 2D transistors were used to fabricate configurable ON/OFF switching circuits. Similar strategies for achieving reconfigurable logic functionality with ambipolar transistors have been demonstrated using metal–graphene–silicon–graphene barristors or using ambipolar organic transistors.^[9,10] These excellent works leverage advances in microfabrication in order to create complex device structures, such as vertical heterojunctions or multi-gate transistors, that enable their behavior. The complexities involved could, however, limit their integration in large-scale devices, and result in high fabrication costs. We believe that to simplify the fabrication process of reconfigurable electronics, it is desirable to have devices that could exhibit dynamically configurable functionalities using simpler structures, such as conventional top-gate bottom-contact designs.

Organic electrochemical transistors (OECTs) comprise a recently developed class of organic transistors that has emerged as an electronic platform that is well-suited for reconfigurable electronics. Recently, OECTs have been investigated for many electronic applications, including bio-sensing, wearable devices, and neuromorphic computing systems, where they can serve as artificial synapses or nonvolatile memories.^[5,14–18] A typical OECT consists of three electrodes (source, drain, and gate), a

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channel comprising a mixed organic ionic-electronic semiconductor connecting the source and the drain, and an electrolyte connecting the semiconductor and the gate electrode.^[19] In such a transistor, the application of a gate voltage results in ion migration into the semiconductor channel. This induces electrochemical doping/dedoping reactions of the channel material, thereby modulating the source-drain current.^[20] Advantages of OECTs include low operating voltages (<1V), very fast switching time compared to other organic devices, and very low power consumption projected of 35 aJ per switching event for a channel of 0.3 μ m × 0.3 μ m large (compared to ≈10 fJ for biological synapses and ≈1.23 fJ for nanowire-based organic fieldeffect transistors).^[5,21-24] As organic-based devices, OECTs can be simply and easily printed into large-area electronic circuits using room temperature organic materials.^[25,26] Due to these attributes, OECTs are attractive for reconfigurable electronics. However, the development of long-term stable ambipolar OECTs is currently at an early stage as most n-type active materials in OECTs can easily be degraded.^[27,28] Despite recent progress in creating complementary from ambipolar OECTs,^[28-31] reconfigurable OECT transistors with dynamically gate-tunable doping types have not been demonstrated. Another viable approach would be to dynamically tune the operation mode of OECTs. OECTs operate either as enhancement-mode transistors or as depletion-mode transistors. Depletion-mode transistors are in an ON state at zero gate voltage, while enhancementmode transistors are in an OFF state at this condition. Enhancement mode OECTs have the active channel of an organic mixed ionic-electronic semiconductor that can be doped following the ion uptake, for example, poly(2-(3,3'-bis(2-(2-(2-methoxyethoxy) ethoxy)ethoxy)-[2,2'-bithiophen]-5-yl)thieno[3,2-b]thiophene) (p(g2T-TT)),^[32] while depletion-mode OECTs are based on semiconductors that are in a doped state, but become dedoped upon ion uptake, as in poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate) (PEDOT:PSS).^[20,33] The operation mode of an OECT is usually an intrinsic and static property that can be designed but cannot be dynamically changed. For instance, to switch between operation modes using PTEB-S, Zeglio et al. blended an enhancement-mode material, sodium poly(2-(3-thienyl) ethoxy-4-butylsul- fonate) (PTEB-S), with a depletion-mode poly(4-(2,3-dihydrothieno-(3,4-b)-(1,4)dioxin-2-yl-mematerial. thoxy)-1-butanesulfonic acid, sodium salt) (PEDOT-S), and controlled the operation-mode of conjugated polyelectrolyte-based OECTs by precisely balancing stoichiometry of the two active materials.^[34] More recently, Cea et al. showed that by adding PEI in PEDOT:PSS, a common material for depletion-mode OECTs, the new blend becomes an excellent active material for enhancement-mode OECTs.^[35] Another route to switching the operation mode of OECTs is via device engineering. Doris et al. created a separate compartment for the gate in their OECTs, in which several redox couples are used to control the gate electrochemical potential. This allowed them to vary the threshold voltage of the OECTs. Depending on the specific redox couple, and therefore the threshold voltage, the operation mode of their OECTs can be either depletion or enhancement with the same active material, PEDOT:PSS.^[36] Relying on the same principle, Tan et al. designed two-chamber OECTs, in which the electrochemical potential of the gate can be controlled in an independent separate electrochemical cell containing redox-active

electrodes and analytes.^[37] Their OECTs can in principle used as mode switching transistors, but the slow response time of redox reactions (on the order of 100 s) makes this approach unattractive for electronics. Finally, the only work using OECTs for reconfigurable electronics is found in microwave resonators, where Bonachini and Omenetto employed PEDOT:PSSbased OECTs in conjunction with metamaterial structures to modulate the bulk conductivity of the channel material.^[38] While not directly related to reconfigurable logic circuits, their work highlights the simplicity of OECTs, the robustness of the transistors' performance, and the possibility of effectively using the transistors in electronics.

In this work, we take advantage of the rich flexibility in material design and the simplicity in device engineering that OECTs have to offer, and report the fabrication of dynamically operation-mode switching OECTs with a self-doped conjugated polyelectrolyte poly[2,6-(4,4-bis-potassium butanylsulfonate-4H-cyclopenta-[2,1-b;3,4-b']-dithiophene)-alt-4,7-(2,1,3-benzothiadiazole)] (PCPDTBT-SO₃K, aka, CPE-K) as the active material. We show that a CPE-K OECT can operate in both enhancement mode and depletion mode, depending on the polarity of the applied voltages. This dual-mode behavior is attributed to the tunability in electrical conductivity of the active material CPE-K: it can both be doped by injection of anions and dedoped by injection of cations. We develop a device physics model that explains the devices' working mechanism and correctly predicts its behavior in both operation modes. To highlight the potential of these dual-mode organic transistors for reconfigurable logic circuits, we demonstrate a reconfigurable logic gate that can switch from a NOR gate to an AND gate on the fly, by simply reversing the voltage polarity.

2. Results and Discussion

2.1. Device Structure and Dual-Mode Behavior

The structure of a typical OECT in this study (Figure 1A) consists of an interdigitated contact, a spin-coated CPE-K that connects the source and drain, an electrolyte (100×10^{-3} M KCl), and an Ag/AgCl gate electrode (see Experimental Section and Supporting Information Section S1 for details on the fabrication). A drain voltage V_d is applied on the source and drain to create a source-drain current Id. To control the ion injections from the electrolyte to the channel, a gate voltage V_d is applied from the gate to the source electrode. We observed that CPE-K OECTs operate in both enhancement mode and depletion mode. To operate CPE-K OECTs in the enhancement mode, the drain voltage and gate voltage were set to negative values ($V_d < 0$ and $V_q < 0$). From the transfer characteristics of an OECT at different drain voltages, shown in Figure 1B, it is observed that the drain current increased with the increase in the amplitude of the gate voltage. The higher the drain current, the steeper the slope of the transfer curves, with the change of slope from $V_{\rm d}$ = -0.2 V to $V_{\rm d}$ = -0.4 V being more pronounced than that from $V_d = -0.4$ to $V_d = -0.6$ V, indicating that the device reaches saturation at more negative drain voltage (semi-log plot in Figure S2A, Supporting Information). In an ON/OFF switching experiment (Figure 1C), in which the gate voltage is switched





Figure 1. Dual operational mode organic transistors with CPE-K OECTs. A) Top: The chemical structure of PCPDTBTSO₃K (aka CPE-K); Center: Schematics of an OECT based on CPE-K; Bottom: the CPE-K OECT can work in two operation modes depending on voltage polarity. B–E) Transfer characteristics and ON/OFF switching of a CPE-K OECT operating in enhancement mode ((B) and (C), respectively), and in depletion mode ((D) and (E), respectively).

between 0 V and -0.3 V, the transistor switches between the OFF state ($V_{\rm g} = 0$ V) to the ON state ($V_{\rm g} = -0.3$ V). The amplitude of the switching experiment was chosen so that the device is not in the saturation regime to avoid the risk of sample degradation or instability.^[39] The ON/OFF switching of the output current was in phase with the switching of the gate voltage, which is a characteristic of an enhancement-mode transistor.

With the same device, we observed that under a reversed polarity, $V_{\rm d} > 0$ V and $V_{\rm g} > 0$ V, the operation mode changed to depletion. The transfer characteristics of the transistors in these conditions are presented in Figure 1D, showing the drain current being suppressed as the gate current increases, with the rate of change (the slope of the transfer curves) increasing with the drain voltage (semi-log plot in Figure S2B, Supporting Information). The depletion-mode characteristics are evidenced further with an ON/OFF switching experiment (Figure 1E): with the gate voltage alternating between 0 and 0.3 V, the output current switched between the ON state ($V_g = 0$ V) to the OFF state $(V_g = 0.3 \text{ V})$. As a consequence, CPE-K OECTs can operate in both operation modes, depending on the polarity of the applied voltages. It is noted that OECTs based on PEDOT:PSS operate in depletion mode and do not switch to enhancement mode by switching voltage polarity as CPE-K OECT (Section S3, Supporting Information).

2.2. Device Physics Model of the Dual-Mode Single-Device OECTs

CPE-K is a self-doped conjugated polyelectrolyte that has recently been used in OECTs.^[40] The dependence of the

operation mode on the polarity suggests that both cation injection and anion injection into the CPE-K film would result in electrochemical reactions that change the doping state of the active channel. To investigate this behavior, we conducted a spectroelectrochemical study of a CPE-K film. Our spectroelectrochemical cell (Figure 2A) consists of a cuvette containing an electrolyte KCl 100×10^{-3} M (the same electrolyte used in the OECTs). A CPE-K film deposited on an ITO substrate was used as a working electrode. As a self-doped p-type semiconductor, the positive polarons in the CPE-K backbone are stabilized by the SO₃⁻ groups.^[24] In the cell, an Ag/AgCl pellet was used as the counter electrode. A voltage is applied between the working and the counter electrode: a positive voltage would draw anions (Cl⁻) into the CPE-K film and therefore a higher doping level is expected while a negative voltage would result in cation (K⁺) migration into the film leading to a lower doping level due to the compensation of SO_3^- by the K⁺ ions. These ion migrations would in turn result in electrochemical reactions, changing the CPE-K between a doped state to a dedoped state indicated in Figure 2A (bottom), which could be tracked with the UV-vis absorption of the film. The absorbance of the CPE-K at different biases is shown in Figure 2B. We observed three typical peaks: the one at \approx 400 nm corresponds to the π - π transition, the one ≈700 nm corresponds to the intra-charge transfer, while the one at ≈1180 nm is a polaron absorption.^[41] The intensity of the polaron peak reflects the level of charge carrier density inside the CPE-K film.^[41,42] With a voltage sweeping from -0.8 to 0.8 V, the three peaks follow a reverse trend: the π - π and charge transfer peaks decrease and the polaron peak increases. The increase of the polaron peak compared at the expense of the other two peaks indicates the increase of polarons in the

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Figure 2. Spectroelectrochemical behavior of CPE-K. A) Left: Schematics of the spectroelectrochemical experiment; Top right: Schematics of ion uptake in CPE-K; Bottom right: Doping and dedoping electrochemical reactions of CPE-K. B) Absorption of CPE-K depending on an applied voltage (vs Ag/AgCl). C) The relative absorption of CPE-K at different V with respect to that of V = 0 V. D) The relative change in absorption of the polaron peak ($\lambda = 1180$ nm) as a function of applied voltage V.

backbone of CPE-K, i.e., higher doping level. This can be explained with the following mechanism: under the application of a voltage, ions in the electrolytes would migrate into the film, which then interact with the anion SO_3^- group. If cations K⁺ are injected, the Coulombic interaction between K⁺ and the $SO_3^$ groups would compensate for the ionic sidechains and would destabilize the polarons. As a result, the number of polarons decreases and the film is dedoped (less conductive). In case of anion Cl⁻ migration, Cl⁻ anions would stabilize the positive polarons as well as interact with the cationic counter-ions in the CPE-K and free up SO_3^- groups to stabilize the polarons on the backbone, hence increasing the hole carrier density of the CPE-K film. These electrochemical processes can be described as the following:

Dedoping: $PCPDTBT^{+}SO_{3}^{-} + K^{+} + e^{-} \rightarrow PCPDTBT^{0}SO_{3}^{-}K^{+}$ (1)

Doping: PCPDTBT⁰SO₃⁻K⁺ + Cl⁻ \rightarrow PCPDTBT⁺SO₃⁻ + KCl + e⁻ (2)

This reversed trend between the charge transfer peak and the polaron peak upon a sweeping voltage is evidenced further in Figure 2C, plotting the adjusted absorption value with respect to the absorption at V = 0 V. From this curve, the difference in intensity of the polaron peak to that at V = 0 V is plotted with regard to the applied voltage (Figure 2D). In this plot, around the negative value of the applied voltage, corresponding to cation injection into the film, we observed that the polaron peak plateaued at high amplitude V (more negative). This could be attributed to an exhaustion of available polaron inside the CPE-K film. Indeed, the concentration of the polaron in dehydrated CPE-K is in the order of 10^{18} cm⁻³,^[42] whereas the concentration of K⁺ in 100×10^{-3} M KCl is in the order of 6 $\times 10^{19}$ cm⁻³. The excess of cation in the film could compensate for all SO₃⁻ sidechains, impeding the self-doping

of CPE-K. As the voltage goes to the positive range, we observe a linear increase of the polaron peak intensity. This behavior could be attributed to the volumetric capacitance characteristics of the organic semiconductor,^[43] with the number of net ion density written as: $-[K^+] + [Cl^-] = q = (c^*/e)V$, c^* being the volumetric capacitance of the CPE-K films, and *e* the elemental unit charge (1.6×10^{-19} C). In this experiment, the voltage range stayed not higher than 0.8 V to avoid the risk of electrolysis of the electrolyte. However, one could expect another saturation. in which all backbones in CPE-K possess a mobile charge and the conductivity cannot increase further upon further ion injection. We note that the proportional change in charge carrier density, indicated by the polaron peak, at both polarities of applied voltages, has not been shown in conjugated polymers (e.g., p(g2T-TT)),^[44] conjugated polyelectrolyte copolymers (e.g., poly[6-(thiophen-3-yl)hexane-1-sulfonate-*co*-3-(hexylthiophene)] copolymers PTHS-TMA⁻-P3HT/PTHS-TMA⁺-co-P3HT),^[45] or conjugated polymer blends (e.g., PEDOT:PSS).[46] Only selfdoped polyelectrolytes, such as CPE-K (this work) and PEDOT-S and their derivatives are reported with this feature.^[34,47]

Using this understanding from the spectroelectrochemical measurements of the CPE-K, we developed a model to describe the dual-mode behavior of CPE-K OECTs behavior based on Bernards' model for depletion mode OECTs.^[20] A transistor, whose structure shown in the inset of **Figure 3**A, is considered, with KCl as the electrolyte and Ag/AgCl gate. As shown above, the hole density in CPE-K can be described as

$$p(x) = -[K^+] + [Cl^-] - p_0 = \frac{c^*}{e} (V_x(x) - V_g) - n_0$$
(3)

with n_0 being a constant related to the initial charge carrier density of the CPE-K film, c^* the volumetric capacitance of the material, e the unit charge (1.6 × 10⁻¹⁹ C), and with $V_x(x)$ being

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Figure 3. Output characteristics of a CPE-K OECT. A) Output curves, full range, with device structure and dimension of the channel in the inset at the top-right corner. B) Output curves of the quadrant $V_d < 0$ V and $V_g < 0$ V, showing enhancement-mode behavior. C) Output curves of the quadrant $V_d > 0$ and $V_g > 0$, showing depletion-mode behavior. The solid lines in (B) and (C) are calculated from the device physics model (Equation 8) with the same fitting parameters of $V_p = 32.9$ mV and G = 1.66 mS.

the voltage along the *x*-direction (inset of Figure 3A). Note that the charge carrier density in Equation 3 is a positive number, when $V_{\rm d} < V_{\rm g} + p_0 e/c^*$, the charge carrier density becomes 0.

Using the microscopic form of Ohm's law, the current density along the *x*-direction j_x is given as:

$$j_{x} = \sigma(x) \frac{\partial V_{x}(x)}{\partial x} = \left(\frac{C^{*}}{e} (V_{x}(x) - V_{g}) - p_{0}\right) \mu e \frac{\partial V_{x}(x)}{\partial x}$$
(4)

Here, we assume that the hole mobility μ is constant throughout the film, then the conductivity of CPE-K is given as $\sigma(x) = p(x)\mu e$. By definition, $j_x = -I_d/(Wd)$, with W and *d* the width and the thickness of the CPE-K film, we obtain:

$$\frac{I_{\rm d}}{Wd}dx = \left(\frac{C \star}{e} \left(V_x\left(x\right) - V_{\rm g}\right) - p_0\right) \mu e dV_x\left(x\right)$$
(5)

By integrating Equation 5 from 0 to *L* along the *x*-direction, we get:

$$\int_{0}^{L} \frac{I_{d}}{Wd} dx = \int_{0}^{V_{d}} \left(\frac{C^{*}}{e} \left(V_{x}(x) - V_{g} \right) - p_{0} \right) \mu e dV_{x}(x)$$
(6)

or

$$\frac{I_{\rm d}}{Wd} L = \left(\frac{C \star}{e} \left(\frac{1}{2} V_{\rm d} - V_{\rm g}\right) - p_{\rm o}\right) \mu e V_{\rm d} \tag{7}$$

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To simplify Equation 7, we define a conductance $G = \frac{Wd}{L} \mu e p_0$ and a threshold voltage $V_p = (p_0 e)/c^*$; thus, Equation 7 becomes

$$I_{\rm d} = -G \left(1 + \frac{V_{\rm g}}{V_{\rm p}} \right) V_{\rm d} + \frac{1}{2} \frac{G}{V_{\rm p}} V_{\rm d}^2 \tag{8}$$

For validation of the model, we obtained the output characteristics of the OECT at different gate voltages (Figure 3A), and used the Equation 8 to fit experimental data. The experimental output curves of the transistors show two quadrants of saturation: when $V_q > 0$ V and $V_d < 0$ V, the film showed a very low conductivity, attributed to exhaustion of polarons due to excess K⁺ cation injections. When $V_{\rm g}$ < 0 V and $V_{\rm d}$ > 0 V, the film showed very high conductivity, reaching saturation of polarons due to the excess of Cl- anions in the film. Only in the other two conditions, namely ($V_g > 0$ V, $V_d > 0$ V) in Figure 3B and $(V_{\alpha} < 0 \text{ V}, V_{d} < 0 \text{ V})$ in Figure 3C that the device showed gating behaviors. Figure 3B shows a typical output characterization of an enhancement-mode OECT: increasing the amplitude of the gate voltage leads to an increase in the drain current. On the other hand, the depletion-mode behavior is observed in Figure 3C: higher amplitude gate voltage lowers the output curve. The governing Equation 8 can be used to fit both behaviors using the same set of fitting parameters $V_p = 32.9$ mV and G = 1.66 mS (solid lines in Figure 3B,C).

Equation 8 provides another way to validate the model with experimental data. The transconductance of the transistor $\partial I_d / \partial V_g$ can be both calculated from this governing equation and can be measured from the slope of the transfer curves (Figure S4, Supporting Information). An expression of the transconductance at non-saturation can be derived from Equation 8

$$g_{\rm m} = \frac{\partial I_{\rm d}}{\partial V_{\rm g}} = -\frac{G}{V_{\rm p}} V_{\rm d} \tag{9}$$

This linear dependence is observed with both enhancement mode ($V_d < 0 \text{ V}$, $V_g < 0 \text{ V}$) and depletion mode ($V_d > 0 \text{ V}$, $V_g > 0 \text{ V}$), showing the slope $g_m/V_d = (32.85 \pm 0.25) \times 10^{-3} \text{ F cm}^{-1} \text{ V}^{-1} \text{ s}^{-1}$ for enhancement mode and g_m/V_d = (34.90 ± 0.20) × 10⁻³ F V⁻¹ s⁻¹ for depletion mode. These similar fitted values $g_{\rm m}/V_{\rm d}$ obtained from the transfer characteristics of two modes are in the same order of magnitude with the one obtained from the output characteristics $g_{\rm m}/V_{\rm d}$ = $G/V_{\rm p}$ = 45 \times 10⁻³ F V⁻¹ s⁻¹. This good fit confirms the validity of our model, which then allows for understanding the principle of the dual-mode OECTs. On the one hand, for the device to operate in enhancement mode, the initial condition is set in the way that the conductivity of the film is very low. It is achieved by applying a negative V_d , so that cations are present in the film even without the gate voltage. A negative small-amplitude V_d (≈ 1 V) might be enough for the film to reach exhaustion of polarons (evidenced by the spectroelectrochemical measurement). From this initial condition, applying a negative gate voltage ($V_g < 0$ V) would draw K⁺ cations out of the film and to drive Cl- anions into the film, allowing for more SO₃⁻ groups to be available for compensating polarons in the backbone of the CPE-K, thereby increasing further the conductivity of the film and the drain current. On the

other hand, to operate the transistor in depletion-mode, a necessary condition is to have a positive drain voltage ($V_d > 0$ V). In this condition, at zero gate voltage there is an excess number of Cl⁻ anions in the CPE-K film, assuring a high conductivity. Increasing the amplitude of a positive V_g would result in more K⁺ cations in the films which interact with the sulfonate SO₃⁻ in the sidechains, weakening the self-doping efficiency of the CPE-K. This results in a lower conductivity of the channel, and therefore, a smaller drain current.

2.3. Demonstration of a Dynamically Reconfigurable Logic Gate

With the OECTs operating in both enhancement mode and depletion mode, and switching between modes being done on-the-fly via switching the polarity of applied voltage, we constructed reconfigurable logic gates which are the basic building blocks of digital electronics. A logic gate produces a binary output from one or more binary inputs, and any complex logic operation can be built from the most common logic gates, such as AND, OR, NAND, and NOR. Reconfigurable logic gates allow to modify the binary output from the same set of digital input without interrupting its operation and without modifying its physical built.^[2] We designed a logic gate from CPE-K OECTs as shown in Figure 4A. The inputs of the logic gate $V_{in}(1)$ and $V_{\rm in}(2)$ were applied to the gate electrode of the two transistors, respectively, and the output signal is the drain current I_{out} . With a $V_{\rm DD} = -0.5$ V and the input voltage $V_{\rm in}(1) = V_{\rm in}(2) = -0.3$ V, the transistors operated in the enhancement mode. In this condition, the logic gate works as an AND gate. At zero gate voltages in one of the two transistors, the current was suppressed (output = 0). Only when both gate voltages were ON the transistors are both in an ON state, allowing for the current to be nonzero (output = 1). Figure 4B shows the input signals, the analog output signal and the digital output signal over time. In the analog signal, the ON/OFF switching may be observed from the spike of the output current. This spike can be eliminated by device engineering (e.g., using thinner active materials).^[26] Here, the spike helps to illustrate the operation of two individual transistors. The switching time of the transistors in this logic gate in the AND gate is 3.8 ms (Figure S5A, Supporting Information).

When the polarity of the applied voltage was reversed, without any hardware alteration, the logic gate became a NOR gate. Indeed, for $V_{DD} = 0.5$ V and $V_{in}(1) = V_{in}(2) = 0.3$ V, the transistors operated in depletion mode. As a consequence, at zero gate voltage, the output current was in an ON state. When one of the transistors was turned into an OFF state by a positive applied input voltage, the current was zero. Figure 4C presents the input voltage sequences and the response of the output signal in analog and in digital forms, respectively. Similar to the AND gate configuration, in the NOR gate configuration, the ON/OFF switching of each transistor could be seen by a spike in the output current, and the digital output was obtained using a binary filter. The switching time of the NOR gate was 8.4 ms (Figure S5B, Supporting Information).

Similarly, we realized an OR gate with two enhancementmode OECTs in parallel. This device was reconfigured to a NAND gate when the transistors were switched to depletion



Figure 4. Dynamically reconfigurable logic gates with CPE-K OECTs. A) The circuit diagram of the logic gate (top) and its output based on the sign of gate and drain voltages (bottom). B,C) For the same input pattern, the logic gate is an AND gate for $V_{DD} = -0.5$ V and $V_{in}(1) = V_{in}(2) = -0.3$ V (B), and a NOR gate for $V_{DD} = 0.5$ V and $V_{in(1)} = V_{in(2)} = 0.3$ V (C).

mode (Figure S6, Supporting Information). To our knowledge, this is the first demonstration of logic based on OECTs. Compared to previous reconfigurable organic logic gates based on organic field-effect transistors (OFETs),^[9] this OECT-based gate requires far smaller voltages (the amplitude of $V_{\rm g}$ and $V_{\rm DD}$ is 0.3 V and 0.5 V here, respectively, compared to 60 V and 60 V for the OFETs), yet achieves similar output (ON) current to OFET devices ($I_{ds} \approx 0.2$ mA). This feature is the result of the higher transconductance of transistor components of the two systems. The average transconductance of OECTs in our circuit at $V_d = 0.3$ V is 10.74 \pm 0.45 mS, while the average transconductance of the p-type OFETs in the previous work was 1.6 µS (a four order of magnitude difference).^[9] The low-operatingvoltage and high-transconductance of OECTs stem from their inherent ion-to-electronic transductions as well as from the bulk conductivity of the channel materials,^[48,49] highlighting the advantages of using this type of transistor as platforms for reconfigurable electronics.

After successfully demonstrating the reconfigurability of the logic gate with CPE-K OECTs, we believe that there are some areas for further improvement of these devices. For instance, for the circuit to work, the voltage signs of the logical inputs would need to be changed. This is non-standard in digital logic, and would require additional components in order to achieve. This can be addressed by configuring the functionality of the circuit with appropriate connections. Another point of improvement is to replace liquid electrolytes with solid-state OECTs. Indeed, because the operation of OECTs is based on ion migration, liquid electrolytes are used for their high ionic mobility. Highly ion-conducting solid electrolytes, such as gels or ionic liquid will allow for more stable and easier to miniaturize devices, with a possibility of integration into the largescale high-throughput printing process.^[23,25,26,35] For example, one particular interest lies in the fabrication of reconfigurable instrumentation amplifier circuits, which are relevant for physiological sensing such as electroencephalography measurement^[50] and noninvasive hormone monitoring.^[51] Another promising development lies in combining these reconfigurable CPE-K OECT circuits with organic solar cells for self-powered flexible on-skill integrated electronics.^[52]

3. Conclusions

We have reported in this communication the dual operation mode behavior of organic transistors with single-device conventional top-gate bottom contact architecture. The transistors are CPE-K- OECTs that operate in both depletion mode and enhancement mode. Dual-mode single-device transistors have not been demonstrated with organic counterparts because operation modes in organic transistors are static and switching between two modes requires complex molecular design or device engineering. As for our transistor, this is done via simply changing the polarity of the applied gate and drain voltages. We conducted a study of the underlying working mechanism using spectroelectrochemistry and developed an analytical model to describe this behavior that matched well with experimental data. The model provided insights into the transistor operation, and revealed that the dual operation mode feature stems from the unique concurrent doping following anion injection and dedoping following cation injection into the CPE-K film. The dual-mode transistors were used to construct a configurable logic gate, for example, one that switches from an AND to a NOR gate and vice versa. This finding paves the way toward a simple and low-cost fabrication of low-power consumption reconfigurable electronics, and opens new opportunities for organic transistor designs that are relevant to currently emerging applications of OECTs in neuromorphic computing and biosensing.

4. Experimental Section

Materials: CPE-K was synthesized following a protocol detailed elsewhere.^[41] KCI and Ag/AgCI pellets were bought from Sigma–Aldrich and used as received.



OECT Fabrication: Interdigitated contacts were fabricated on Silicon substrates with a lift-off process, using two photoresists (SF11 and AZP4110, MicroChemical). Photoresist patterning was done with a mask aligner SUSS MA6, and photoresist development was done with AZ400k (for AZP4110) and MicroChem 101A Developer (for SF11). Au evaporation was done via thermal evaporation and lift-off was done by immersing the evaporated wafers in an *N*-methyl-2-pyrrolidone (NMP) bath in 60 °C. Detailed parameters of the photolithography and lift-off process could be found elsewhere.^[53] Active material CPE-K (10 mg mL⁻¹ with 0.1% v.v. (3-glycidyloxypropyl)trimethoxysilane (GOPS)) was spin-coated on the interdigitated contact, with a thickness of 75 nm, the channel area was defined via solution removal with a Q-tip, then was annealed at 110 °C for 30 min. Devices were kept in a desiccator if not used.

Spectroelectrochemical Characterization: UV-vis-NIR spectra were obtained using a Perkin-Elmer Lambda 750 UV-vis-NIR spectrometer using a tungsten lamp for the visible to near-IR region and a deuterium lamp for the UV region. A 100% transmission blank was obtained using a clean indium tin oxide/glass substrate in KCl 100 \times 10⁻³ $\,$ M for all spectra.

OECT Electrical Characterization: Characteristics of OECTs were measured with a Keithley semiconductor parametric analyzer (model 4200-SCC). Inputs for the logic gates were done with two function generators and were recorded using a Teledyne Lecroy Waveace 2034 oscilloscope and a function generator. The voltage sweeps of the transfer curves in both modes started at 0 V, the voltage sweeps of the output curves started at $V_d = -0.3$ V. The hold time for each data point was 100 ms, two orders of magnitude higher than the switching time of the devices. Forward and backward sweeps of the transfer curves in both modes showed negligible hysteresis (Figure S2C,D, Supporting Information).

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

dynamically gate-tunable transistors, organic electrochemical transistors, organic electronics, reconfigurable logic gate

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