



Optimizing Switching Behavior of Organic Electrochemical Transistors

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Abstract

Organic electrochemical transistors (OECTs) are pivotal in bioelectronics due to their ability to translate ionic signals into electronic ones. Their high transconductance, low-voltage operation, and compatibility with aqueous environments make them ideal for interfacing biological systems with electronics. However, the transient behavior of OECTs, particularly the effect of drain potential in switching kinetics, remains poorly understood. This study analyzes the influence of drain potential on the transient response of p-type accumulation mode OECTs. We observed significant current overshoot and slow relaxation times (> 60 sec) when the transistor switches from the depleted to doped regime, caused by drain potentials. These phenomena are attributed to over-depletion of the channel, which can impact the device's reliability in practical applications. We propose an approach to reduce current overshoot and improve switching response with a current stabilization time of less than 10 seconds. Our findings offer valuable insights for optimizing the operation regime of OECTs, enhancing their switching speed, power efficiency, and reliability.

Keywords: Organic electrochemical transistors; Organic ionic-electronic conductors; Switching transient; Electrochemical doping; Transistor switching response.

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1. Introduction

In biological systems, such as living cells, signal transmission occurs via ionic currents, while in human-made electronic devices, signals are transmitted through electronic currents.^[1,2] To effectively interface biological systems with electronic circuitry, materials capable of efficiently conducting both types of charge carriers are essential.^[3,4] A large class of materials with this dual electrotransport capability, known as mixed ionic-electronic conductors, has been developed. Among these, certain conjugated polymers have been optimized to balance both ionic and electronic conductivity, making them ideal for interfacing biological and electronic systems.^[5] In the literature, these materials are referred to as organic ionic-electronic conductors (OIEMCs).

The next critical step in advancing bioelectronics is the development of devices capable of converting ionic signals into electronic ones.^[6] Organic electrochemical transistors

(OECTs), which use OIEMC channels in contact with an electrolyte, are particularly well-suited for this task.^[7] OECTs have garnered significant attention due to their unique properties, including the highest transconductance among all transistor technologies.^[8] Their organic nature, compatibility with aqueous environments, and ability to operate at low voltages (<1 V) make them especially promising for bioelectronics applications^[1,7]

Despite their potential, the operation of OECTs is complex, as it involves the simultaneous drift of both ionic and electronic charge carriers.^[1,7] At present, there is no comprehensive analytical model that fully describes both the steady-state and transient behavior of OECTs. While the steady-state characteristics of OECTs have been studied extensively,^[1,7,9,10] our understanding of their dynamic response, particularly the kinetics of switching, remains limited.^[7,11-16] Recently, we developed a general transition line model for OECTs that accounts for electronic drift transport and ionic injection and diffusion across the channel, driven by the gate voltage.^[17] This model describes the time-dependent relationship between drain current and gate bias. However, experimental observations are more complex due to the influence of capacitive and resistive elements in the electrolyte and at the interfaces between the gate and the channel.^[18]

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The transient response of OECTs is further complicated by the effect of the drain potential, which also contributes to the asymmetry observed in the output I - V curves.^[19] The ion injection and extraction dynamics are governed by the potential difference between the gate and the channel, while the channel potential itself is determined by the drain bias.^[9] The impact of the drain potential on the transient behavior of OECTs can be significant, yet this effect has not been thoroughly explored in the literature. It has often been overlooked, in part, because it can be mitigated by applying a small drain bias and avoiding operating regimes where the effect is pronounced.

Nevertheless, in practical applications, the influence of the drain potential on ionic injection and extraction dynamics cannot be ignored, as it directly affects the switching transients and may compromise device reliability. In this study, we conduct a detailed analysis of the transient behavior related to the drain potential's impact and propose a straightforward approach to mitigate this effect, thereby improving the reliability of OECT-based devices in bioelectronics.

2. Materials and methods

In this study, the following materials were used: Poly(3-hexylthiophene-2,5-diyl) (P3HT) from Luminescence Technology Corp (LT-S909) and chlorobenzene from Sigma-Aldrich (anhydrous, 99.8%, 284513). Devices were fabricated on glass substrates patterned with interdigitated ITO, with a pre-patterned source-drain from Biotain Crystal. The dimensions of the source-drain are width \times length: 30 mm \times 50 μ m, and the ITO thickness is 90 nm.

To prepare the P3HT solution, 25 mg of the polymer was dissolved in 1 mL of chlorobenzene. The solution underwent stirring for 3 hours at 50 °C before spin-coating. Prior to deposition, the ITO-patterned source-drain substrates underwent thorough cleaning. Initially, the substrates were sonicated in deionized (DI) water with a detergent for 10 minutes, followed by rinsing three times with DI water. Subsequently, they were sonicated in acetone and isopropyl alcohol (IPA) for 10 minutes each. Finally, the substrates were dried via nitrogen flow and subjected to UV-ozone treatment for 15 minutes to eliminate any residual organics and enhance substrate surface wettability.

The fabrication techniques for P3HT OECTs were adapted from the work of Ginger and his colleagues.^[20,21] The structural design of the P3HT-based OECT is illustrated in Fig. 1. Shortly, P3HT channel layers were deposited using a spin-coating technique. Specifically, 20 μ L of the P3HT solution at a temperature of 50 °C was dispensed onto a substrate spinning at 1000 rpm and allowed to rotate for 1 minute resulting in a 100 nm thick layer. The as-coated P3HT films were annealed at temperature of 120 °C for 20 minutes. A 100 mM tetrabutylammonium hexafluorophosphate in acetonitrile served as the electrolyte, and an Ag wire was employed as the gate electrode.

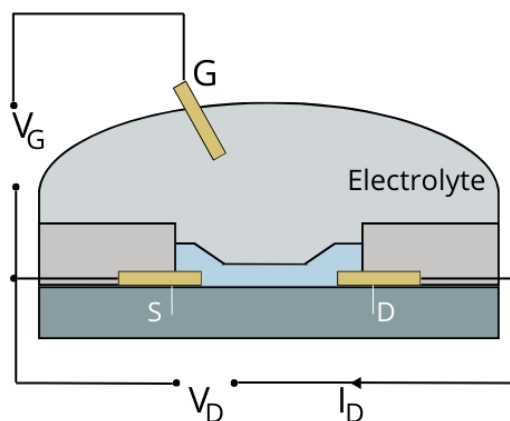


Fig. 1: Schematic illustration of the P3HT-OECT.

The drain current transients of OECTs were measured using a potentiostat (CS350M, Corrtest Instruments) in a chronoamperometry regime. The gate perturbation was performed by signal generator (R&S@SMCV100B, Rohde & Schwarz GmbH & Co. KG)

3. Result and discussion

In OECT, the doping and dedoping of the channel, which occur via ion insertion and extraction, are susceptible to the drain potential.^[19,22] This is because ion drift is governed by the potential difference between the gate and the channel.^[7,10] The potential of the channel is determined by drain bias. It is non-uniform and can be approximated as varying linearly from the value of source potential to the drain potential over its length. Therefore, changes in the drain potential affect both the ionic and electronic currents.

In the case of an accumulation mode OECT with a p-type channel, the standard operating regime is when the drain is biased negatively relative to the source at a fixed potential, and the drain current is modulated by variations in the gate bias (V_g). Here, the sign of the potential difference between the gate and the drain ($\Delta V = V_g - V_d$) is critical, as it determines the doping state of the channel.

When the gate potential is more negative than the drain potential ($\Delta V < 0$ V), the channel is in the doped state. Under this condition, ΔV creates an electrochemical driving force that pushes anions into the channel. The total charge of the injected anions is compensated by the injection of holes from the source, resulting in a boost in the channel's conductivity due to the increased hole density. We will refer to this regime of OECT as the 'doped regime'.

Contrary, when the gate potential is more positive than the drain potential ($\Delta V > 0$ V), the potential difference creates an electrochemical driving force that pulls anions out of the channel (and/or pushes cations into the channel). This renders the channel in a depleted state with decreased conductivity, and we will refer to this regime as the 'depleted regime' of the OECT.

Depending on its application, an OECT can be operated by switching between the depleted and doped regimes, or by

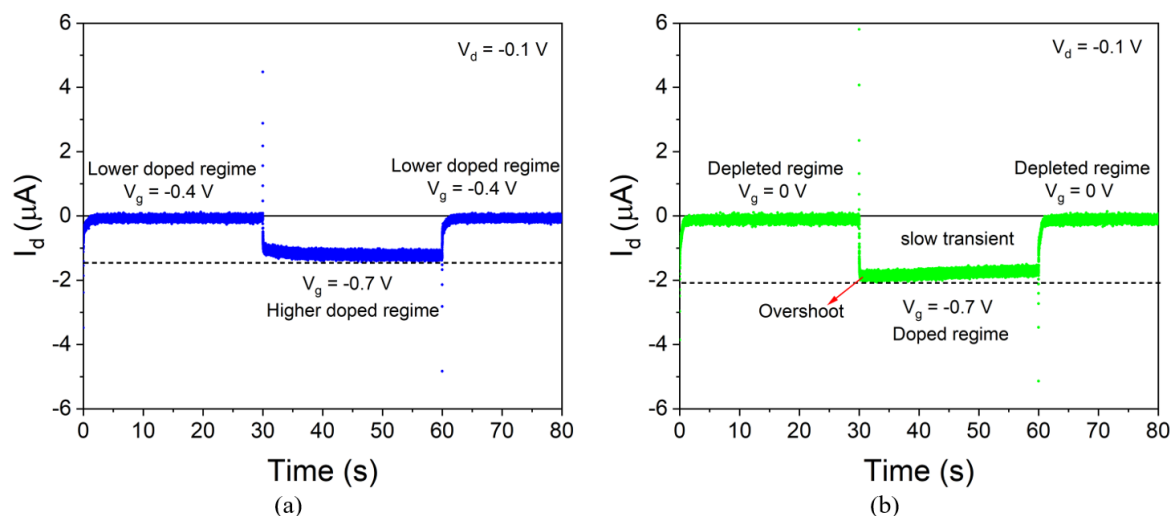


Fig. 2: Drain current transients of an accumulation mode OECT with a P3HT channel: (a) Switching transient between two doped regimes and (b) between the depleted and doped regimes.

switching between two doped states. However, it is worth mentioning that in the depleted regime, the transistor consumes less power. Therefore, in OECT-based logic circuits, it is energetically more efficient to switch between the depleted and doped regimes.

Apart from power consumption, the switching speed of the transistor plays a critical role in the operation of electronic circuits.^[11-17] OECTs are known for their relatively slow switching speeds, primarily due to the slow kinetics of ion movement.^[7] Various studies have been conducted to improve the response rate of OECTs.^[13,15] It has been reported that there is an asymmetry in the switching responses between the switch-on and switch-off processes. Typically, OECTs switch off faster than they switch on, a phenomenon attributed to lateral ion drift within the channel.^[11,13]

However, in OECTs, there is a much slower kinetics in the drain current transient, which, to the best of our knowledge, has not been discussed in the literature. This slow current relaxation is observable only during the transition from the depleted regime to the doped regime. In Fig. 2, the drain current transient of an accumulation-mode OECT with a P3HT channel is shown, illustrating the switching transients between two doped regimes and between the depleted and doped regimes.

In both cases, we observe that the transistor switches off faster in similar manner (the transition from less negative V_g to more negative V_g). In contrast, it turns on more slowly, which is related with lateral ion drift in the channel. However, the turn-on switching transients have distinctive differences. The switching transient from low doped state to highly doped state goes through the spike of current with opposite direction followed by monotonic relaxation to new equilibrium current level (Fig. 3a). This switching transient is well described by our analytical model,^[17] which further was elaborated by J. Bisquert taking into accounting the electrolyte and interfacial limitations.^[18] However, in the case of the transition from the

depleted regime to the doped regime, the current initially exhibits a spike in the opposite direction, followed by an overshoot beyond the new equilibrium level, and then slowly recovers to the equilibrium state.

The origin of the observed current overshoot, followed by slow current stabilization after switching from the depleted to doped regime, has not been discussed in the OECT literature. However, this phenomenon can impact the reliability of OECTs in electronic circuits, as the overshooting and relaxation times can be significant, as will be shown below. The level of current overshoot and the relaxation time depend on both the drain potential and the duration for which the OECT was held in the depleted regime.

As seen in Fig. 3, biasing V_d to more negative values results in an increase in both the current overshoot amplitude and the slope of the slow current relaxation. This indicates that the time required for the OECT to reach a new stable equilibrium state increases as the drain bias becomes more negative. A similar effect is observed with the time duration for which the OECT is maintained in the depleted regime. Fig. 4 illustrates the switching transient at $V_d = -0.3$ V under gate perturbation from 0 V to -0.7 V, with varying durations of the gate square signal. The longer the OECT remains in the depleted regime, the more pronounced the current overshoot becomes, and the steeper the slope of the slow current transient, indicating a longer relaxation time to reach a stable equilibrium.

The origin of the observed current overshoot, followed by slow relaxation in the switching transient from the depleted regime to the doped regime, is not yet fully understood. However, its dependence on the drain potential and the duration for which the OECT remains in the depleted regime suggests that it is related to the dedoping of the channel. We attribute this phenomenon to over-depletion of the channel occurring during the depleted regime.

To support our assumption, we first maintain the channel

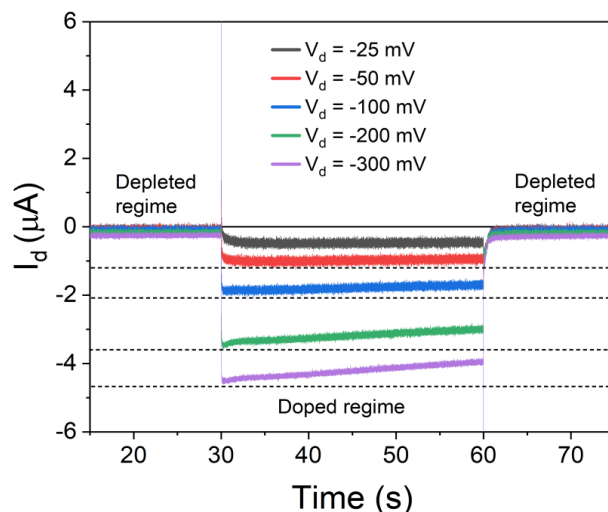


Fig. 3: Drain current transients of an accumulation mode OEET with a P3HT channel at different drain biases (V_d).

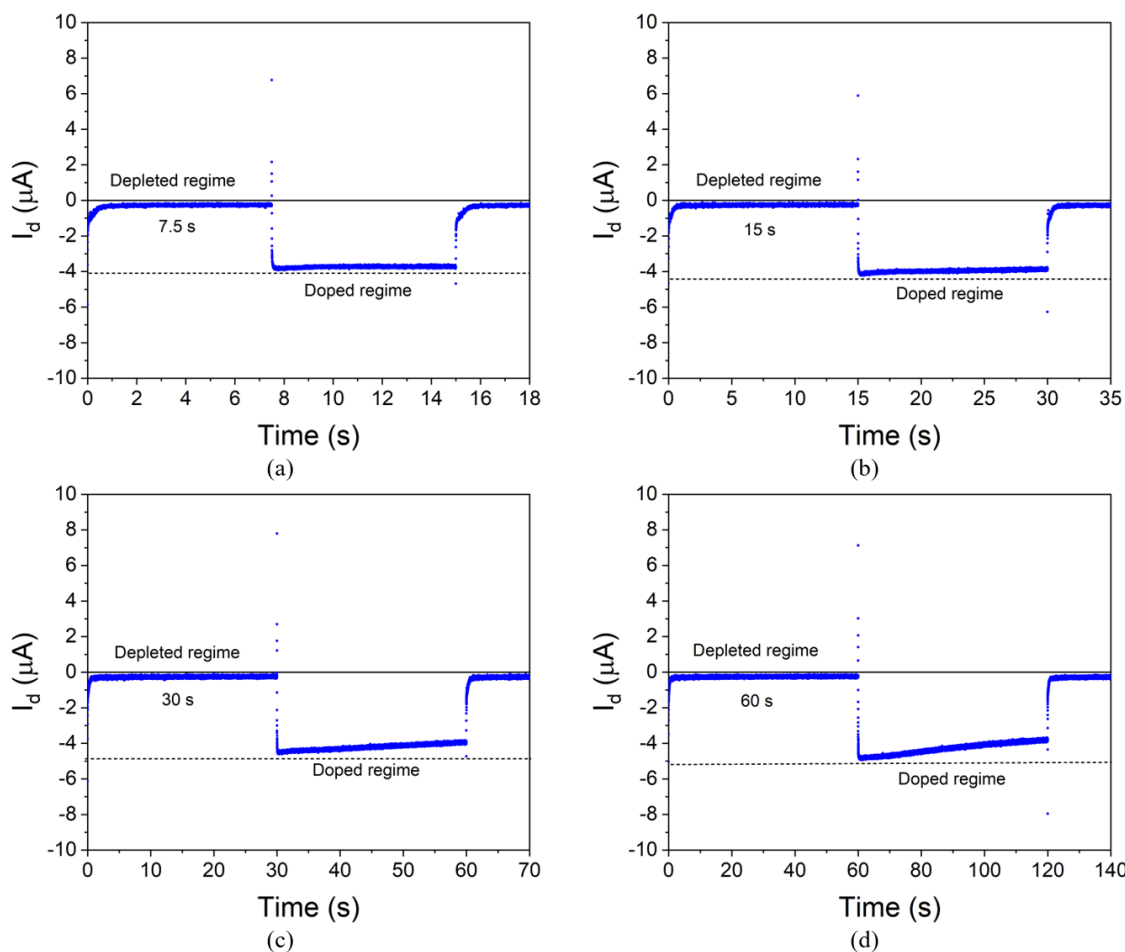


Fig. 4: Drain current transients of an accumulation mode OEET with a P3HT channel at different gate perturbation periods: (a) 7.5 sec, (b) 15 sec, (c) 30 sec and (d) 60 sec (Drain bias (V_d) is -0.3 V).

in a ‘neutral’ state, where there is no potential difference between the gate and the channel, and then apply a gate perturbation to observe the switching transient. This neutral state can be achieved by placing the gate in an open-circuit condition, which is equivalent to connecting a high impedance in series with the gate circuit. In this case, $\Delta V=0$ V, meaning

no doping or dedoping of the channel occurs. We will refer to this state as the ‘neutral regime’ of the OEET.

Fig. 5 shows the switching transient from the neutral regime to the doped regime, and for comparison, the transient from the depleted regime to the doped regime is also provided. The transient from the neutral regime does not result in current

overshooting. Instead, it behaves similarly to the switching between two doped states, with a current spike followed by a monotonic relaxation to the equilibrium state. However, when the gate is switched back to the open-circuit condition, the current recovers to its equilibrium level extremely slowly. This slow recovery is attributed to the fact that, in the open-circuit condition, the extraction of anions is driven solely by the gradient in anion concentration.

To further support our assumption that when V_d is more negative than V_g , the channel is depleted, we measured the impedance spectra (IS) of the OECT between the source and drain electrodes under two different conditions. Impedance spectroscopy is a small perturbation technique that allows the extraction of electrical parameters of a device in steady-state operation without disrupting its established condition.

First, the IS of the OECT was measured in the steady-state condition with the gate floating and with the drain biased at -0.2 V. The resulting IS is shown in Fig. 6 (black dots). Then, the IS was measured under steady-state conditions with

$V_g=0$ V and with the drain biased at -0.2 V (Fig. 5, red dots).

The measured impedance spectra of the OECT are quite complex and a full interpretation is beyond the scope of this work. However, in general, the active resistance (Z') measured at $V_g = 0$ V is higher, indicating that the channel conductivity is lower compared to the floating gate condition. This supports our conclusion that when V_d is more negative than V_g , the channel is depleted. The primary cause of this depletion is the electrochemical force induced by the potential difference between the gate and drain (the channel), which extracts anions (or inserts cations).

As we have demonstrated, the switching from the neutral to the doped state occurs more rapidly, with a current stabilization time of less than 10 seconds, showing no current overshoot, and followed by a relatively slow recovery to equilibrium. In contrast, switching from the depleted state to the doped state exhibits pronounced current overshoot, followed by a much slower recovery process taking more than 60 seconds. The former switching scheme can be more

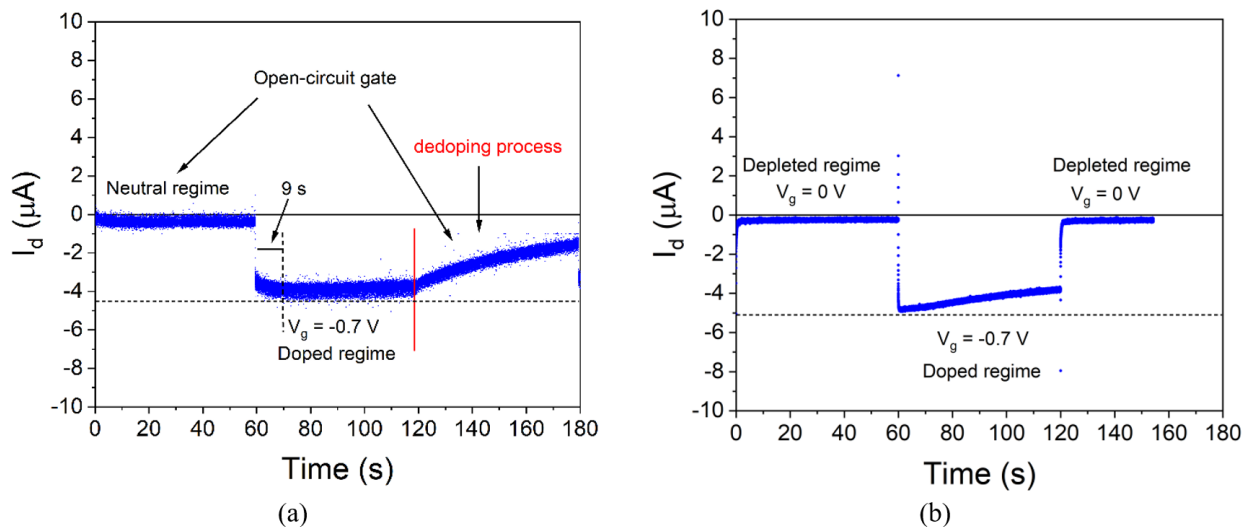


Fig. 5: Drain current transients with different gate perturbation schemes: (a) Transition from the neutral to the doped state and (b) Transition from the depleted to the doped state.

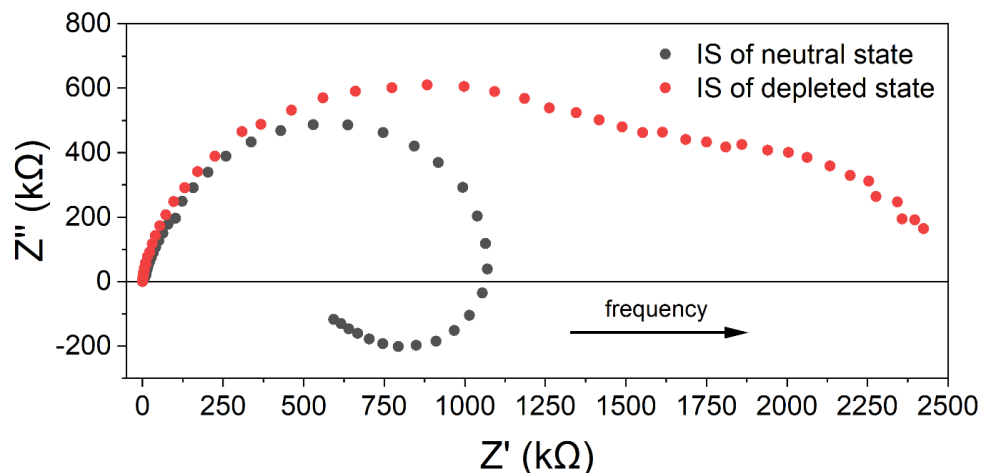


Fig. 6: Impedance spectra of the channel measured in the neutral state and depleted state. In the neutral state, the gate was left open-circuit, and in the depleted state, it was held at 0 V relative to the source. The drain was biased at -0.2 V in both cases.

reliable for OECT-based electronic circuits. The following extremely slow switch-off transient can be easily mitigated by temporarily grounding the gate and then returning it to a floating condition. Maintaining the gate at open-circuit (or in series with high impedance) prevents the over-depletion of the channel that leads to the slow transient kinetics observed after switching on. This approach will be addressed in our upcoming study.

4. Conclusion

In this study, we examined the impact of drain potential on the transient behavior of accumulation mode OECTs with a P3HT channel. Our findings highlight the significant effect of drain potential on switching kinetics, particularly during transitions from the depleted to doped regimes. The observed current overshoot and slow relaxation, lasting longer than 60 seconds, are attributed to over-depletion of the channel, which is enhanced by both negative drain bias and prolonged depletion periods. These effects can compromise the reliability of OECTs in electronic circuits. However, switching from a neutral regime, where the channel is neither doped nor depleted, effectively mitigates current overshoot and improves switching reliability, with a current stabilization time of less than 10 seconds. This insight provides a promising avenue for enhancing OECT performance in bioelectronics applications, ensuring faster, more reliable switching with lower power consumption. Future studies will focus on refining this approach and further exploring the role of gate modulation in optimizing OECT behavior.

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Conflict of Interest

There is no conflict of interest.

Supporting Information

Not applicable.

Reference

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