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Electrolyte engineering in organic electrochemical transistors for advanced electrophysiology

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Abstract

Electrophysiology is an indispensable tool in the early diagnosis of a wide range of diseases, making the precise, continuous, and stable recording of electrophysiological signals critically important. Organic electrochemical transistors stand out among various electrophysiological recording devices, offering a high signal-to-noise ratio due to their intrinsic amplification capability. However, despite their inherent advantages, several challenges persist in practical scenarios, such as the stability of wearable devices, limited spatiotemporal resolution, and undesired inter-channel crosstalk in implantable systems. Addressing these challenges may require innovative approaches in electrolyte engineering. This perspective summarizes the latest advancements and ongoing hurdles in the electrolyte engineering of organic electrochemical transistors, highlighting their potential to revolutionize advanced electrophysiological applications.

Keywords: Organic electrochemical transistors, electrolyte engineering, electrophysiology

INTRODUCTION

Electrophysiology, depending on its application scenarios, unveils the electrical properties inherent in heart tissues[1,2] and living neurons[3,4]. This empowers scientists to underpin diverse heart rhythm abnormalities[5,6]

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and decode intercellular and intracellular messages[7,8], leading to the establishment of platforms for early diagnosis and treatments of cardiovascular and neurodegenerative diseases[9,10]. Most electrophysiological techniques, e.g., electrocardiograms (ECGs)[11-13], electroencephalograms (EEGs)[14,15], electrocorticography (ECoG)[16,17], *etc*., capture signals by situating bioelectrodes on the tissue surface[18]. Conventional electrodes, typically termed "passive electrodes"[19], have their own sets of challenges. For instance, metal electrodes frequently result in undesired immune responses, making them unsuitable for chronic applications in the biotic environment. Organic electrodes, on the other hand, generally exhibit a low signal-to-noise ratio (SNR) resulting from their limited conductivity[20].

Transistors[21,22], particularly the organic electrochemical transistors (OECTs)[23,24], hold advantages in detecting low-amplitude signals within physiologically relevant time frames owing to their intrinsic amplification capabilities. OECTs excel in on-site signal processing, design flexibility, and biocompatible characteristics. Their unique mechanism, bulky modulation of the active layer, results in a volumetric capacitance, endowing them with high transconductance (g_m) under low-operation voltage, enabling high sensitivity and safe operation for biosensing applications[25,26]. Leveraging these benefits, *in vitro*[27,28] and *in vivo*[29-31] signals have been successfully recorded using OECTs [Figure 1A].

The form factors of OECTs are evolving towards increased stretchability and conformability to the tissue. This necessitates using interfacial materials that are both adhesive and porous, retaining excellent conformability without forming interfacial air gaps, ensuring high oxygen and water vapor permeability, and allowing the underlying tissue to breathe freely [Figure 1B]. Future advancements in implantable devices also demand innovations to engineer these systems to be programmable for enhanced spatiotemporal resolution and biodegradable to reduce the need for subsequent surgical interventions. For chronic application scenarios, including self-healing property further boosts the durability, practicality, and lifespan of OECTs [Figure 1C]. Additionally, integrating self-powered features, such as coupling with energy ultraflexible energy harvesters[32], paves the way for eco-friendly bio-integrated systems. Despite the rapid evolution of this field, achieving high-quality recordings with superior spatiotemporal resolution and stability still presents a formidable challenge.

RECENT PROGRESS IN ELECTROLYTE ENGINEERING OF OECTS

One primary issue arises with the use of liquid electrolytes. Their tendency to evaporate might decrease the reliability of *in vitro* applications[33], and they pose a barrier to individual gating in integrated circuits/arrays for *in vivo* applications in an ion-rich medium. While organic liquid electrolytes demonstrate superior ionic conductivity, many have not been proven to be biocompatible, and fluidic issues persist[34]. Solid-state electrolytes, i.e., ion gels[35,36] based on polymers blended with ionic liquid or based on biomaterials[37] such as hydrogels[38], levan polysaccharide[39], *etc*., offer improved nonvolatility and facilitate stable operation of the devices under dynamic conditions. This is attributed to the crosslinked polymer structure that enhances stability while maintaining ionic conductivity by incorporating ionic-conducting components. Interfacial intimacy can be facilitated through in-situ polymerization, while biocompatibility further necessitates careful material selection. Besides, the electrolyte materials should be mechanically soft to maintain conformal contact between the skin and devices, which is crucial to decrease the impedance for efficient gating and realize stable recording.

The radar chart in Figure 2 provides a comparative analysis of key performance metrics across diverse electrolytes currently in use. These encompass aqueous electrolytes such as NaCl solution and phosphate-buffered saline (PBS), organic liquid electrolytes such as 1-ethyl-3-methylimidazolium tetrafluoroborate ([EMIM][BF_4]) and 1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl) imide ([EMIM][TFSI])[34],

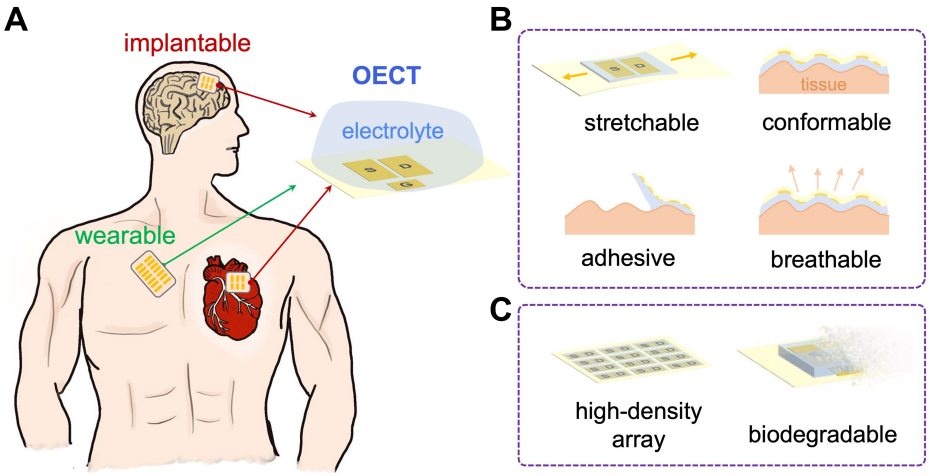


Figure 1. OECTs for advanced electrophysiology. (A) Schematic of wearable and implantable OECTs, with a detailed depiction of a typical OECT device on the right. Here, S, D, and G denote the source, drain, and gate electrodes, respectively; (B) Evolutionary trends in OECT development, including the stretchable, conformable configurations, adhesive interfaces, and breathable device structure; (C) Future directions for OECTs in advanced implantable systems, highlighting the importance of developing high-density arrays, alongside biodegradable and self-healing features tailored for diverse application scenarios. OECTs: Organic electrochemical transistors.

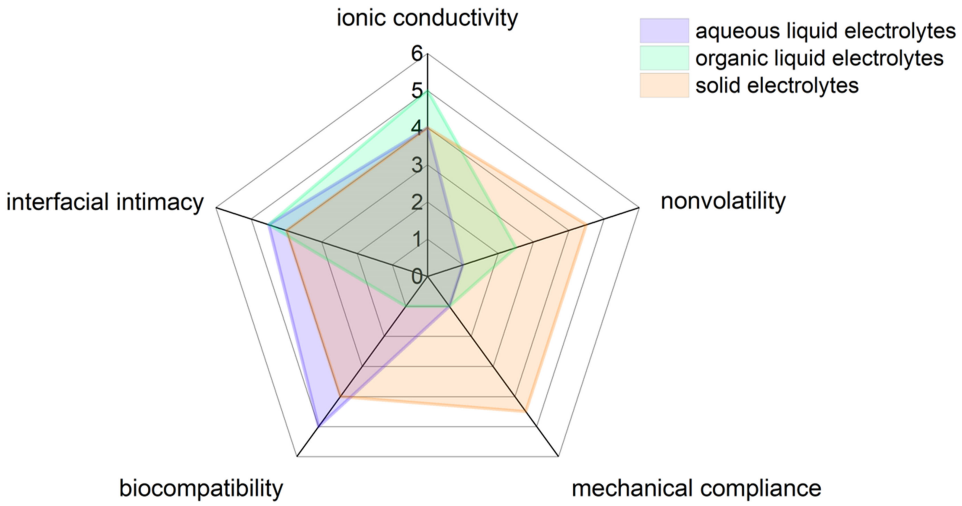


Figure 2. Radar chart comparing performance metrics of existing electrolytes. The interfacial intimacy is used to assess the contact property between the electrolyte and the active material.

and solid-state electrolytes[35-39] such as poly(vinyl alcohol) (PVA) hydrogel and poly (vinylidenefluofide-co-hexafluoropropylene [P(VDF-HFP)]-[EMIM][BF_4]). Generally, aqueous electrolytes exhibit advantages in terms of tunable ionic conductivity, interfacial intimacy with the channel material, and biocompatibility. Their main limitation is the volatility and the lack of robust mechanical properties. Organic liquid electrolytes, on the other hand, possess favorable characteristics in interfacial intimacy and nonvolatility and excellent ionic conductivity. Yet, they are constrained by limited biocompatibility and mechanical compliance. Solid electrolytes present tunable mechanical compliance, interfacial intimacy, and nonvolatility. With careful engineering, they can achieve satisfactory levels of both ionic conductivity and biocompatibility.

Using ion gels, formulated with gelatin, PBS, and glycerin[36], in conjunction with the well-established active material poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate) (PEDOT:PSS), successful and stable mapping of ECG signals was accomplished via an OECT array [Figure 3A]. The devices retained high performance after medium-intensity exercise, demonstrating their potential for sports-related applications. Moreover, using a gelatin-glycerol electrolyte as the substrate facilitated the construction of a highly stretchable OECT, designed to adapt to skin deformation, ensuring stable recording and conformability[40].

For the long-term application of wearable OECTs on the skin, it is crucial to ensure both the operational stability of the devices and the comfort of the users. The integration of nonvolatile solid electrolytes could significantly enhance the operational stability. For instance, glycerol gel has been used as a solid electrolyte to realize continuous electrophysiological monitoring for several hours and demonstrates stability in the air for more than seven days[33]. In addition, gas-permeable materials and devices have been developed[41,42], effectively mitigating potential skin irritation or device degradation. For example, a gas-permeable solid-state polymer electrolyte (SPE) [Figure 3B] was applied in fibrous nano-mesh OECT, realizing "breathable" OECTs that exhibit high-quality electrophysiological recordings[41]. Ultrathin hydrogel films with a thickness down to 10 μm have been developed recently and applied as universal biocompatible interfaces towards breathable, skin-integrated electronics [Figure 3C][42].

For *in vivo* recordings, high spatial and temporal resolution are crucial for capturing the intricate dynamics of intercellular and intracellular communication and accurately recording high-frequency signals. Although the *in vivo* environment is naturally aqueous, and the tissue fluid can function as an electrolyte, electrolyte engineering can still solve persisting challenges.

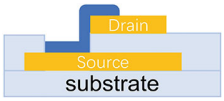
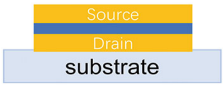
To realize high temporal resolution, the conductivity of the electrolyte and channel capacitance are pivotal factors, as determined by[43]

$$\tau = R_E \cdot C_{ch} \tag{1}$$

where τ denotes response time, R_E is electrolyte resistance, and C_{ch} stands for channel capacitance. Generally, high ionic conductivity of the electrolyte would decrease the response time of OECTs, thus enabling high temporal resolution. Additionally, strategies for controlling the channel capacitance through active material and channel geometry engineering have been explored. Regarding the active material, p-type and n-type materials operating in an accumulation or depletion mode have been demonstrated, as summarized in Table 1. Various factors, such as the polymer backbone structure, side chain symmetry, and localized microstructure of the organic films, were identified as influential on the device response time[49,50]. For instance, the inclusion of ethylene glycol side chains has been observed to enhance transient characteristics by regulating hydration levels[51]. Bithiophene units functionalized with triethylene glycol side chains represent a promising building block for accumulation-mode OECTs, facilitating rapid temporal responses and robust operational stability[52].

Regarding channel geometry, vertical structures[23,48] [Table 1], are favored for achieving faster responses due to reduced channel length and, consequently, decreased volumetric capacitance. Specifically, in a step-type vertical structure, the channel length corresponds to the thickness of the interfacial layer, typically around 1 μm. In a sandwich-type vertical structure, the channel length is determined by the thickness of the active layer, typically around 0.1 μm. Notably, across various device configurations, molecular orientation plays a crucial role in determining both ionic drift [53] and charge carrier transport pathways[54], thereby affecting

Table 1. Summary of documented electrolyte materials, channel active materials, and device architecture in OECTs

Electrolyte		Active layer			Architecture	Ref.
Material	**Type**	**Material**	**Type**	**Working mode**	**Schematic**	
0.1M NaCl	Aqueous	BBL	n	Accumulation	Planar	[44]
0.1M NaCl	Aqueous	BBL:PEI	n	Depletion		[45]
[EMIM][BF_4]	Organic	p(g1T2-g5T2)	p	Accumulation		[46]
Glycerol gel	Solid	PEDOT:PSS	p	Depletion		[33]
0.1M NaCl	Aqueous	PEDOT:PSS	p	Depletion	Vertical (step-like)	[23]
PBS	Aqueous	p(g2T-TT)	p	Accumulation		[47]
PBS	Aqueous	p(C_6NDI-T)	n	Accumulation		[47]
PBS	Aqueous	HOMO-gDPP	n	Accumulation	Vertical (sandwich-like)	[48]
[EMIM][TFSI]:PEGDA	Solid	PIDTPEG-BT	p	Accumulation		[35]

OECTs: Organic electrochemical transistors; BBL: poly(benzimidazobenzophenanthroline); PEI: polyethyleneimine; p(g1T2-g5T2): poly[3,3'bis(2-methoxyethoxy)-2,2'-bithiophene]-co-[3,3'-bis(2-(2-(2-(2-(2methoxyethoxy)ethoxy)ethoxy)ethoxy)ethoxy)-2,2'-bithiophene]; p(g2T-TT): poly[2-(3,3'-bis{2-[2-(2-methoxyethoxy)ethoxy]ethoxy}-[2,2'-bithiophen]-5-yl)thieno[3,2-b]thiophene]; p(C_6NDI-T): naphthalene diimide thiophene-based (NDI-T) backbone functionalized with ethylene glycol side chains.

transient responses. Meticulously selecting device architecture, channel materials, and electrolytes makes it possible to significantly improve the transient response and overall performance of the OECT devices.

An innovative approach to reduce the response time (τ) involves integrating the channel material with the electrolyte. In particular, an internal ion-gated OECT (IGT), a unique type that incorporates hydrated ion reservoirs within a conducting polymer channel, was innovated[55]. This design eliminates the reliance on an external electrolyte, and significantly reduces the time of ions participating in the de-doping process, thereby enabling enhanced operational speed [Figure 3D]. Based on this structure, a rapid response time of 2.6 μs was achieved, resulting in an effective bandwidth of 380 kHz, and ensuring a high SNR within the physiological frequency bands. Moreover, when IGTs are adopted together with a vertical device configuration, known as vIGT[56], high spatial and temporal resolution (sub-μs domain) can be realized at the same time [Figure 3E]. Local field potential (LFP) patterns corresponding to wakefulness, rapid eye movement (REM) sleep, and non-REM sleep were recorded precisely using a vIGT-based recording system [Figure 3F]. In addition to embedding ions directly into the active layer, using an in situ π-ion gel as the active material and an internal gate capacitor - a design known as π-ion gel transistors (PIGTs)[57] - also effectively reduces response time (down to 20 μs). This approach enhances the device performance by maximizing the interfacial area between the ionic liquid and the semiconducting fibers, facilitating rapid ionic transport and electronic responses. The above-mentioned strategies work effectively toward high-speed OECTs.

To achieve high spatial resolution *in vivo*, it is essential to develop channels with a high density. Preventing the inter-channel crosstalk would be a prerequisite to ensure the quality of signals. Recent research has made progress by integrating OECTs with organic electrochemical diodes, which serve as switches. This approach leads to negligible signal interference across the channels and allows for the multiplexing of amplified LFPs within the active recording pixel (26-μm diameter)[58]. To realize individual gating of high-density OECT arrays, vIGTs represent a noteworthy example[56]. They boast an impressive density of approximately 155,000 transistors per square centimeter and demonstrate stable performance in an aqueous environment. Strategically incorporating an H-via design [Figure 3E] and embedding ions in the active layer effectively minimized crosstalk.

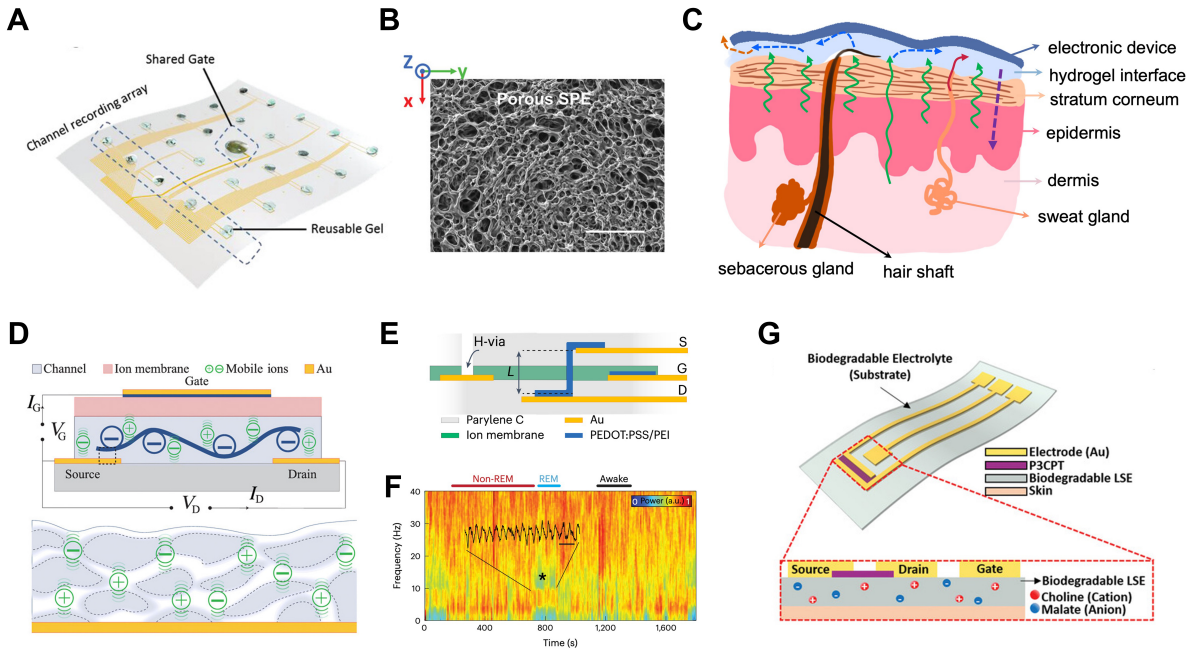


Figure 3. Electrolyte engineering of OECTs for electrophysiological recording. (A) Demonstration of wearable OECT array adopting solid gel electrolyte[36]; (B) The SEM image of a SPE, showing porous structure. Scale bar, 200 μm[41]; (C) Schematic of breathable skin-integrated electronics using an ultrathin hydrogel film as the skin/electronics interface[42]; (D) Schematic of an internal IGT, which has mobile ions embedded in the active layer[55]; (E) Schematic showing the cross-section of a vIGT. *L* denotes the vertical channel length. *S*, *D*, and *G* stand for the source and drain contacts and the gate electrode. The H-via shows a micro-conduit from the device surface through the ion membrane layer to permit hydration of the channel[56]; (F) The time-frequency spectrogram of the neural data captured and wirelessly transmitted by a vIGT-based standalone device. The characteristic local field potential patterns associated with wakefulness, REM sleep, and non-REM sleep are revealed. A superimposed raw time trace highlights the theta oscillation characteristics during REM sleep. Scale bar, 250 ms; (G) Schematic of a biodegradable OECT based on biodegradable electrolyte[39]. Figure 3A adapted with permission from ref.[36], Copyright 2023 John Wiley and Sons; Figure 3B adapted with permission from ref.[41], Copyright 2022 John Wiley and Sons; Figure 3C adapted with permission from ref.[42], Copyright 2022 John Wiley and Sons; Figure 3D adapted with permission from ref.[55], Copyright 2019 Authors, licensed under Creative Commons Attribution NonCommercial License 4.0 (CC BY-NC); Figure 3E and F adapted with permission from ref.[56], Copyright 2023 Authors, licensed under Creative Commons Attribution 4.0 International License; Figure 3G adapted with permission from ref.[39], Copyright 2020 John Wiley and Sons. OECTs: Organic electrochemical transistors; SEM: scanning electron microscopy; SPE: solid-state polymer electrolyte; IGT: ion-gated electrochemical transistor; REM: rapid eye movement.

For stable electrophysiological monitoring *in vivo*, the inherent movement of tissues, such as brain and cardiac tissues, during their functions poses a challenge in maintaining consistent signal acquisition over the long term[59,60]. To overcome this challenge, adhesive and biocompatible materials should be integrated with the OECTs. For instance, the N-hydroxysuccinimide (NHS) ester group was used to form covalent bonds with the primary amine groups on tissue surfaces, thus enhancing the bio-adhesion[61]. In a different approach, a bio-adhesive polymer semiconductor (BASC) film established rapid and robust adhesion to biological tissues. In this regard, bio-adhesive solid electrolytes, including hydrogels[42], can act as a conformable interface, providing mechanical cushioning to ensure stable contact and maintain the recording fidelity.

Can electrolyte engineering address the aforementioned challenges comprehensively and generate high-performance, high-speed OECTs in large-scale integrated circuits with minimal crosstalk in physiological media? In an *in vitro* setting, employing a solid electrolyte could facilitate the separation of individual channels, enabling individual gating[53]. When applied in an aqueous *in vivo* context, a solid-like electrolyte that exhibits biocompatibility, adhesive properties to tissues, and the ability for patterning, thereby

supporting high-density configurations, has the potential to enable high-quality signal mapping with enhanced spatial resolution and reduced crosstalk. Exploring such systems could simplify device architecture while preserving their functionalities. This area remains largely unexplored, and efforts are yet to be made.

Finally, to circumvent the secondary surgical interventions, developing biodegradable and bioresorbable bioelectronics has become a pressing research priority. For example, a biodegradable electrolyte composed of all edible materials, i.e., levan polysaccharide and choline-based ionic liquid, was developed that concurrently serves as the device substrate [Figure 3G][39]. Such biodegradable OECTs demonstrated successful recording of ECG signals from the heart surface of a rat. These findings underscore the potential that biodegradable electrolytes hold for the application in implantable medical devices.

SUMMARY

Despite the considerable progress enabled by electrolyte engineering in OECTs, particularly within electrophysiology, challenges exist in realizing high-quality signal acquisition under dynamic physiological conditions and across prolonged durations. For wearable OECT applications, it is essential to ensure the user comfort and the device sensitivity simultaneously. These OECTs must exhibit mechanical flexibility and softness commensurate with biological tissues while allowing the skin underneath to breathe and move naturally.

In the context of implantable OECTs, ensuring robust interfacial adhesion with the soft and dynamic tissues remains a nascent challenge. Developing bio-adhesive and biocompatible electrolytes possessing requisite ionic conductivity is anticipated to be a key strategy. Such advancement would enable seamless integration and sustained interfacing with biological tissues, thereby facilitating chronic biomedical investigations and interventions. Additionally, the engineering of OECTs with controlled degradation pathways remains an ongoing challenge, which would minimize long-term adverse effects from device residue and hold promise for enhancing the practicality of these devices in therapeutic settings.

A further challenge within the liquid *in vivo* environment is achieving high spatial and temporal resolution in high-density arrays and integrated circuits without interference- the "crosstalk"- between adjacent channels. Engineering the patternable, bio-adhesive, and solid-state electrolytes could fundamentally solve such issues, yet substantial efforts are still necessitated in this field.

Finally, the interplay between device architecture, channel materials, and electrolyte engineering remains an open question. Consider the innovative IGT as a case in point; beyond the predominant PEDOT:PSS channel material, there remains huge potential of broadly applying this strategy to embed ions into other channel materials. In common OECTs, the intricacies and control of ion penetration and transport at the electrolyte/channel material interface are yet to be fully understood. Besides, unveiling the localized microstructure is crucial for comprehending the ionic/electronic interactions and the subsequent electronic conductivities, both of which demand further in-depth investigation.

To summarize, strategic innovations in electrolyte materials could redefine the capabilities of OECTs, advancing electrophysiological devices that combine high performance and stability, biocompatibility and comfort, with the dynamic intelligence demanded for cutting-edge biomedical applications.

DECLARATIONS

Authors' contributions

Conceived the topic and supervised the research: Xu X
Wrote, discussed, and revised the manuscript: Zhu Y, Xu X

Availability of data and materials

Not applicable.

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Conflicts of interest

Both authors declared that there are no conflicts of interest.

Ethical approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

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