Journal of
Materials Chemistry C

COMMENT

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Cite this: *J. Mater. Chem. C*, 2026, **14**, 6534

Comment on "Electron-interfered field-effect transistors as a sensing platform for detecting a delicate surface chemical reaction" by G. Choi, K. Lee, S. Oh, J. Seo, E. Park, Y. D. Park, J. Lee and H. S. Lee, *J. Mater. Chem. C*, 2021, 9, 8179

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Received 15th July 2025,
Accepted 9th March 2026

DOI: 10.1039/d5tc02689j

rsc.li/materials-c

This comment critically examines the recently proposed electron-interfered field-effect transistors (EIFETs), which are a modification of organic charge-modulated field-effect transistors (OCMFETs). Despite the potential of EIFETs, as suggested by Choi *et al.* (*J. Mater. Chem. C*, 2021, **9**, 8179, https://doi.org/10.1039/D1TC01073E), discrepancies in experimental results and theoretical predictions raise questions about their operational mechanisms, which this comment seeks to address. Through an analysis of the device structure and operating principles, we demonstrate that the proposed EIFET cannot detect molecular adsorption because charge accumulation generates a gate current rather than the claimed threshold voltage shifts.

Organic electronics have emerged as an attractive option for next-generation devices owing to their essential advantages, including flexibility, low-cost production, large-area production, and compatibility with various substrates. This introduction seeks to contextualize the importance of advancements in organic field-effect transistors (OFETs) technology, highlighting the shift towards organic charge-modulated field-effect transistors (OCMFETs) and the innovative electron-interfered field-effect transistors (EIFETs). The critical examination of EIFETs presented in this comment is predicated on understanding these technological strides and their implications for sensing applications.

OFETs have gained significant attention among organic electronic devices owing to their potential applications in sensing, logic circuits, and flexible displays. The OFET device can be described as a three-electrode system, where the charge transport in the OFET device can be described using the gradual channel approximation proposed by Shockley.[1] In this case, the driving force of the charge transport is the drain–source electric field, while the gate electrode serves to charge the channel region capacitively. Various OFET device geometries have been suggested, including vertical OFETs, organic static-induction transistors, electrolyte-gated organic field-effect transistors, and ion-sensitive organic field-effect transistors. Recent research in the field of biosensors has focused on a particular type of OFET known as an OCMFET, which exhibits unique characteristics that distinguish it from conventional OFETs.

To contextualize our critique of EIFETs, it is essential to understand their predecessors, particularly OCMFETs, within the broader spectrum of organic electronics. The OCMFET operates on the principle of charge modulation, wherein variations in the charge concentration on a floating gate directly affect the conductivity of the organic semiconductor in the channel region. In this architecture, the control gate is the electrode to which external voltage is applied for device operation, while the floating gate is an electrically isolated electrode that accumulates charge from the sensing environment. This modulation is provided *via* capacitive coupling between the floating and control gates, allowing precise control of the electrical properties of the device. The existence of a sensing region on the floating gate exposed to the external environment (analyte) enables the OCMFET to convert perturbations in the surrounding charge distribution into quantifiable electrical signals. The intrinsic sensitivity of OCMFETs to charge variations renders them highly attractive for sensing applications.

It is essential to distinguish the OCMFET from an extended-gate OFET (EG-OFET), as illustrated in Fig. 1. Both the OCMFET and EG-OFET employ a sensing electrode, but the primary distinction is in their charge modulation mechanisms. In an EG-OFET, the sensing electrode is directly linked to the gate electrode, and the sensing mechanism depends on variations in the work function of the sensing electrode owing to its interaction with the analyte. In most cases, it is utilized using redox

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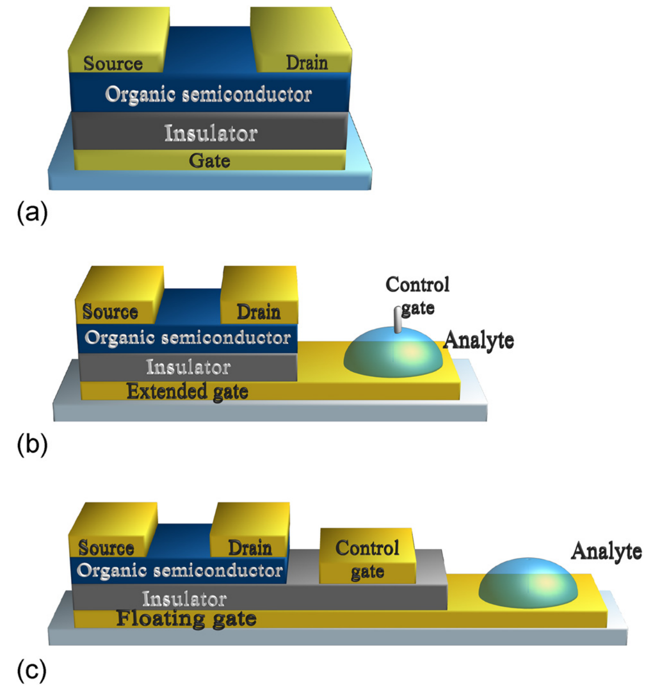

Fig. 1 Device geometries of (a) OFET, (b) EG-OFET, and (c) OCMFET.

reactions. In contrast, the OCMFET utilizes a floating gate that is electrically insulated from the control gate, and the sensing mechanism relies on charge accumulation on the floating gate generated by the analyte. Here, it is assumed that the charge varies owing to molecular adsorption on the floating-gate surface. These differences in operational principles result in differences in sensitivity, response time and possible applications.

Moreover, the OCMFET distinguishes itself from the conventional OFET by its ability to immediately detect charge variations. In a conventional OFET, the gate electrode regulates channel conductivity; therefore, it does not intrinsically detect charge variations in the surrounding environment. The drain–source current I_{ds} in an OFET device in the saturated region follows relation

$$I_{ds} = \mu C_g (W/2L)(V_{gs} - V_{th})^2,$$

where μ is the charge carrier mobility, C_g is the gate insulator capacitance per unit of area, W and L are the channel width and channel length, respectively, V_{gs} is the gate–source voltage (*i.e.* $V_g - V_s$), and V_{th} is the threshold voltage.[2] Fig. 2 illustrates the common structure of the OCMFET fabricated on a heavily doped silicon substrate.

On the other hand, the OCMFET includes a floating gate and sensing region, addressing this issue, and provides direct charge-sensing capabilities. In the OCMFET, the effective gate voltage is the floating gate potential V_{FG}, so the drain–source current follows the relation

$$I_{ds} = \mu C_g (W/2L)(V_{FG} - V_{th})^2,$$

where $V_{FG} \approx V_{gs} - (Q_{sens}/C_{cf})$. Here Q_{sens} is the immobilized charge in the sensing area and C_{cf} represents the capacitance

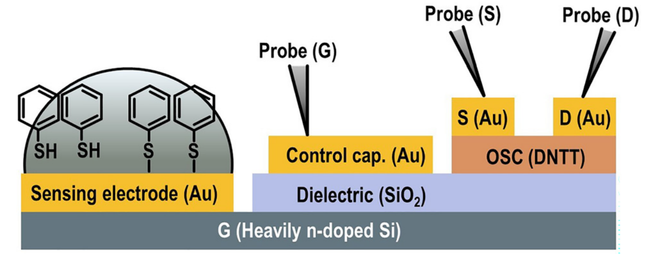

Fig. 2 Operating scheme of OCMFET device. Control cap. denotes the control capacitor formed between the control gate and floating gate. The scheme is based on modification of the device proposed by Choi *et al.*[3] Used with permission of Royal Society of Chemistry, from Choi *et al.*, *J. Mater. Chem. C*, 2021, **9**, 8179; permission conveyed through Copyright Clearance Center, Inc.

between control and floating gates, determining the coupling efficiency. The distinctive features of OCMFETs enable numerous possible applications. Their high sensitivity to charge variations makes them ideal candidates for biochemical sensors, environmental monitoring applications, and medical diagnostics.

Given this background on OCMFETs, we now turn our attention to electron-interfered field-effect transistors (EIFETs) to elucidate the critical points of divergence and concern. In detail, the recent work of Choi *et al.*[3] proposed the EIFET as a modification of the OCMFET device, as shown in Fig. 3. Here, the gate potential is applied directly to the extended gate, while the control gate electrode, denoted as the interference electrode, is a floating electrode connected to the analyte. The research report of Choi *et al.* suggested that the variations in the threshold voltage are proportional to the chemical reaction of the thiol molecules on the interference electrode (IE) surface.

EIFETs were proposed to overcome specific limitations of OCMFETs, aiming to enhance sensitivity to molecular adsorption. However, our analysis of the proposed mechanism reveals fundamental issues. The electrochemical reaction between thiol-containing molecules and the gold surface generates electrons that accumulate on the IE, with the total charge Q_{IE} directly proportional to the number of reacted molecules

$$Q_{IE} = -neN_r,$$

where n is the number of electrons per reaction (typically 1), e is the elementary charge, and N_r is the number of reacted

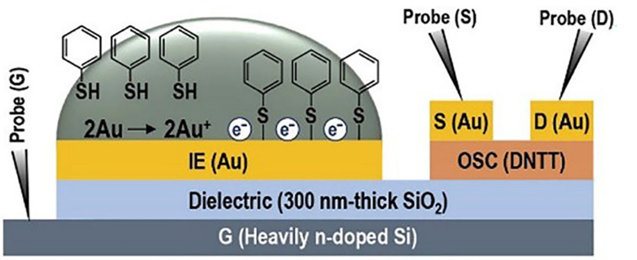

Fig. 3 Operating scheme of EIFET device proposed by Choi *et al.*[3] Used with permission of Royal Society of Chemistry, from Choi *et al.*, *J. Mater. Chem. C*, 2021, **9**, 8179; permission conveyed through Copyright Clearance Center, Inc.

molecules. Owing to capacitive coupling through the insulating SiO_2 layer, this charge accumulation induces an equal and opposite image charge on the gate electrode (silicon substrate). Since the gate electrode is maintained at constant potential, the formation of image charges Q_i requires continuous charge redistribution within the silicon, manifesting as gate current I_g flow from the voltage source

$$I_g = \partial Q_i/\partial t = \partial Q_{IE}/\partial t = -ne\partial N_r/\partial t,$$

where t is the time and $\partial N_r/\partial t$ represents the reaction rate. Note that we use the partial derivative $\partial Q_i/\partial t$ to emphasize that the gate current depends specifically on the time-evolution of accumulated charge while other parameters remain constant. Choi *et al.*[3] reported thiophenol immobilization on the IE for 2 h. Assuming a thiophenol footprint of approximately 0.3 nm^2, the molecule adsorption generates an average gate current of 10 nA per 1 cm^2 of IE. It should be noted here that even though the calculated gate current should be easily detectable with standard semiconductor parameter analyzers, it is generally recognized as a leakage current through the gate insulator layer, and it is not monitored in most experiments. However, the absence of reported gate current measurements in the original study represents a significant oversight, as this current would be the direct evidence for the proposed mechanism.

Notably, while Choi *et al.* claimed that 'obtained variations of the EIFET device performance were almost the same' for thiophenol, dodecylthiol, and pentafluorobenzenethiol, they presented detailed data only for thiophenol. The absence of comparative data for molecules with significantly different chemical properties (aromatic *vs.* aliphatic, fluorinated *vs.* non-fluorinated) makes it difficult to verify the claimed chemical specificity of the sensing mechanism.

According to this analysis, the EIFET device cannot be sensitive to the chemical reaction in terms of the threshold voltage shift, and the observed variation of the threshold voltage probably originates from two opposing phenomena. The fast voltage shift to negative values most probably originates in the charge trapping on the organic semiconductor/gate insulator interface. The observed voltage shift corresponds to the trap charge density of 10^{12} cm^{-2}, which corresponds to commonly reported values.[4] The slow recovery process requires careful consideration. While Choi *et al.* performed blank experiments with toluene solvent that showed smaller threshold voltage shifts compared to thiol solutions (addressing potential thermal or mechanical effects), this control experiment does not conclusively validate their proposed sensing mechanism. The observed differences between toluene and thiol solutions could arise from several factors: (i) different solution volumes or application methods affecting thermal equilibration, (ii) variations in mechanical stress from repeated measurements, or (iii) the presence of thiol molecules affecting charge trapping dynamics at the semiconductor/insulator interface without necessarily involving the proposed electron accumulation mechanism on the IE.

Furthermore, the recovery of threshold voltage to initial values during continuous bias application remains physically inconsistent with genuine chemical detection. Molecular adsorption and Au–S bond formation would produce permanent or slowly-reversible changes rather than the rapid recovery observed within 80 minutes. This recovery pattern is more consistent with the relaxation of trapped charges or mechanical stress[5] than with a chemical sensing mechanism. In addition, the observed recovery pattern (return to original threshold voltage values during continuous bias application) is physically inconsistent with known organic transistor behavior. In other words, the reported timescales (initial shift within 15 minutes, recovery over 80 minutes) are consistent with well-documented bias stress effects in organic transistors, where charge trapping occurs rapidly upon bias application followed by apparent stabilization as the device reaches thermal and mechanical equilibrium. Genuine electronic effects such as charge carrier trapping would result in threshold voltage shifts that are conserved at their shifted values once equilibrium is reached, not recovery back to the original state during continuous operation. The reproducibility of this pattern across multiple devices supports its origin as a systematic experimental artifact. In addition, the similarity of the responses across different thiol molecules suggests that the effect is not chemically specific.

Conclusions

Our analysis demonstrates that the proposed EIFET cannot function as a chemical sensor due to a fundamental design flaw: charge accumulation on the interference electrode must generate gate current rather than threshold voltage shifts. The absence of gate current measurements in the original work, combined with the observed threshold voltage recovery pattern inconsistent with molecular adsorption, indicates that the reported effects likely originate from well-documented artifacts in organic transistors (interface charge trapping and thermal/mechanical relaxation) rather than chemical sensing. This critical limitation prevents EIFETs from detecting surface reactions as claimed.

Author contributions

M. Micjan and M. Weis: investigation; M. Weis: formal analysis; M. Weis: writing – original draft: M. Micjan: funding acquisition.

Conflicts of interest

There are no conflicts to declare.

Data availability

No primary research results, software, or code were included, and no new data were generated or analyzed as part of this comment.

Acknowledgements

This work was supported by the EU NextGenerationEU through the Recovery and Resilience Plan for Slovakia under the project no. 09I03-03-V04-00178.

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